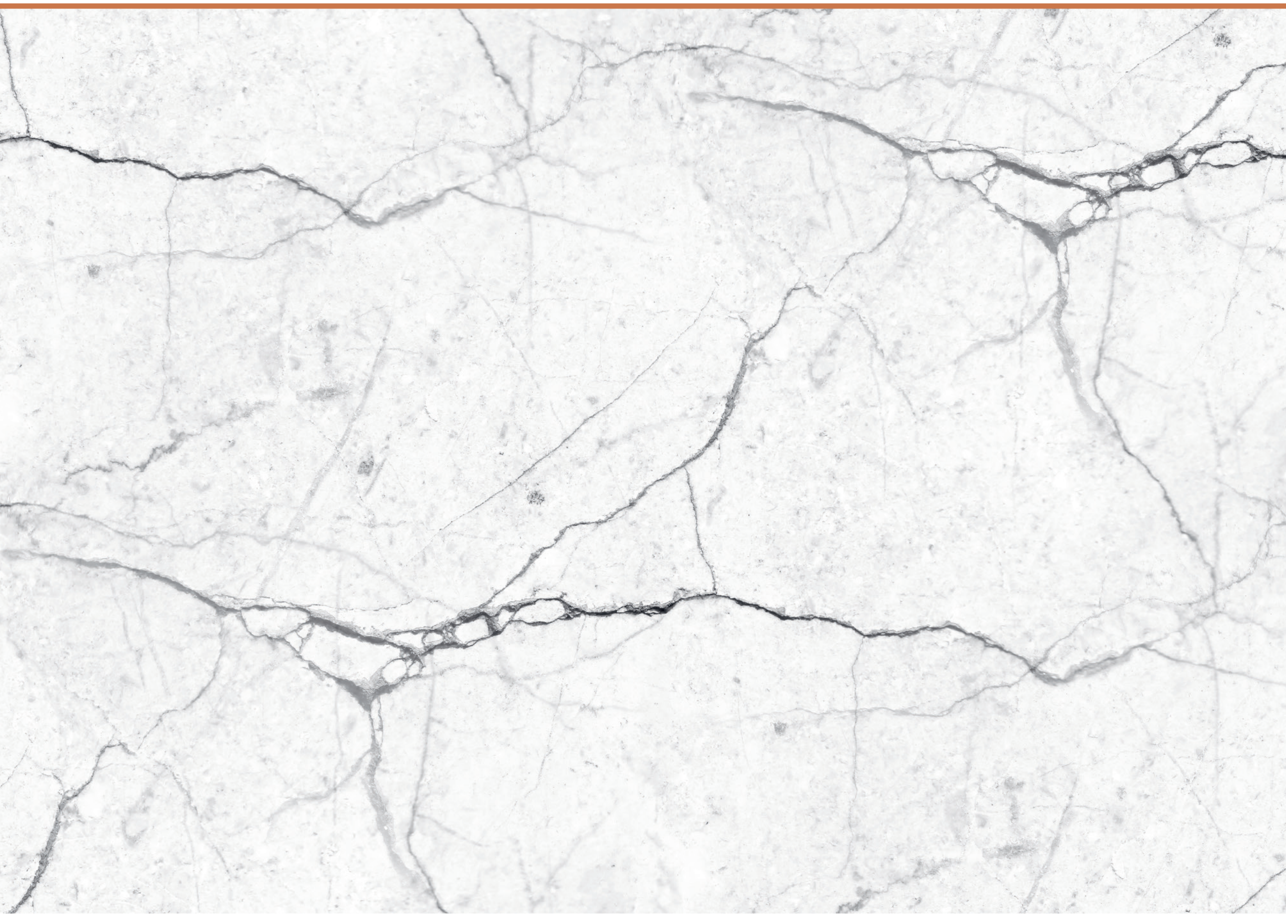
RIGA TECHNICAL UNIVERSITY

Sandra Guzlēna

CRACK PREVENTION IN CERAMIC AND CEMENT BASED MATERIALS

Doctoral Thesis



RTU Press
Riga 2022

RIGA TECHNICAL UNIVERSITY
Faculty of Materials Science and Applied Chemistry
Institute of Technical Physics

Sandra Guzlēna
Doctoral Student of the Study Programme “Materials Science”

CRACK PREVENTION IN CERAMIC AND CEMENT BASED MATERIALS

Summary of the Doctoral Thesis

Scientific supervisors
Senior Researcher Dr. sc. ing.
GITA ŠAKALE
Professor Dr. habil. phys.
MĀRIS KNITE

Scientific consultant
Dr. sc. ing.
SERGEJS ČERTOKS

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To be granted the scientific degree of Doctor of Science (Ph. D), the present Doctoral Thesis has been submitted for the defence at the open meeting of RTU Promotion Council "RTU P-02" on 21 December 2022 at 11:30 online at https://rtucloud1.zoom.us/j/91056890359?pwd=dGtIYlRiT1dyL0Y3MnM3YUZGYzZFdz09.

OFFICIAL REVIEWERS

Professor Dr. sc. ing. Jānis Ločs
Riga Technical University

Assistant Professor PhD Marta Roig Flores
Jaume I Univeristy, Spain

Dr. sc. ing. Līva Dzene
University of Upper Alsace, France.

DECLARATION OF ACADEMIC INTEGRITY

I hereby declare that the Doctoral Thesis submitted for the review to Riga Technical University for the promotion to the scientific degree of Doctor of Science (Ph. D) is my own. I confirm that this Doctoral Thesis has not been submitted to any other university for the promotion to a scientific degree.

Sandra Guzlēna………………………. (signature)
Date: ………………………

The Doctoral Thesis has been written in English as a summary of scientific publications. It consists of a summary and seven scientific publications. The publications are in English and their total length is 51 pages. The Bibliography contains 92 titles.

LODE SIA
Lodes iela 1, Liepa, Liepas pagasts, Priekuļu novads, LV-4128
Reģistrācijas numurs: 50003032071
Pievienotās vērtības nodokļa maksātāja numurs: LV50003032071
Tālrunis: 64195246, 64122512, fakss: 64107721
e-pasts: info@lode.lv, http://www.lode.lv
Mārketinga un pārdošanas birojs: Brīvības iela 155, Rīga, LV-1012
Tālrunis: 67378020, fakss: 67378022

Confirmation of the industry

The research carried out within the framework of the doctoral thesis "**CRACK PREVENTION IN CERAMIC AND CEMENT BASED MATERIALS**" developed by Sandra Guzlēna is important for the development of the company and improvement of product quality. Within the framework of the doctoral thesis, topical issues for the company on ways to reduce cracks in full bricks using innovative solutions have been studied: adding surfactants, using fiberglass reinforcement, as well as optimizing the granulometric composition of on clay mass based raw materials available in the clay quarry.

LODE SIA
Ane factory production manager
Sergejs Čertoks

Confirmation of the industry

The research carried out within the framework of the doctoral thesis "**CRACK PREVENTION IN CERAMIC AND CEMENT BASED MATERIALS**" developed by Sandra Guzlēna on the addition of glass fiber reinforced concrete (GRC) with polymer additive and crack self-healing after the addition of crystalline additive is important for the company's development and future projects.

Skonto Concrete Cladding
Production manager
Roberts Gulbis

ACKNOWLEDGMENTS

This Doctoral Thesis has been developed in collaboration with various RTU institutes and faculties, the Latvia University of Life Sciences and Technologies, and together with two large Latvian companies – LODE Ltd and Skonto Concrete Cladding Ltd. By combining the theoretical knowledge available in universities and the need of companies to solve practical problems, this collaboration has resulted in additional quality of this dissertation.

Already when developing my Master's Thesis at the Institute of Technical Physics, it was clear that with such a team of creative, talented, and curious colleagues it is possible to overcome any obstacles. First of all, I would like to express my gratitude to my supervisor Gita Šakale who has always supported and encouraged me, including the time during the vicissitudes of completing this promotion work. I am grateful for the advice in planning and implementing work assignments as well as in processing and compiling the data obtained. I would also like to thank Professor Māris Knite of the Department of Materials Physics of Institute of Technical Physics for the advice regarding the completion of the promotion work, for the opportunity to attend international conferences and participate in the doctoral conferences for young students and young scientists organized by COST CA 15202, as well as for the opportunity to work in the Laboratory of Materials Physics. Thanks also go to the colleagues in the Institute of Technical Physics (Valdis Teteris, Astrida Berzina, Artis Linarts, Linards Lapcinskis, Vija Brilte.) who helped moving the samples, implementing experiments, and obtaining data, shared healthy criticism about the dissertation, and assisted in solving organizational issues.

I would also like to express my gratitude to LODE Ltd for the opportunity to use materials from the Liepa quarry and the equipment available in the laboratory of the factory, and for the advice provided by the staff. Special thanks go to the laboratory assistant Gunta, Aija Liepa, and Sergejs Čertoks.

I would like to express my gratitude to Skonto Concrete Cladding Ltd for the opportunity to use materials and equipment to make and test samples. I would like to thank Oskars Matisons for his advice and Inese Žigalova and Ernests Matisons for their help in testing the samples.

I would like to thank Ilze Vircava from the Institute of Soil and Plant Sciences of the Latvia University of Life Sciences and Technologies for the permission to use the laboratory premises and the equipment of the Faculty of Agriculture, and to especially thank Ināra Dižakova for the opportunity to determine the granulometry of the clay sample.

I am grateful to the RTU Faculty of Civil Engineering and the Department of Building Materials and Construction Products. Thanks go to Ģirts Būmanis for the help in preparing, testing and analysing concrete samples.

I am thankful to the RTU Riga Biomaterials Innovation and Development Centre for the XRD and SEM measurements and to the RTU Institute of Silicate Materials for the possibility to use their laboratory and equipment. Many thanks are due to Ruta Švinka for valuable advice.

Thanks go to researcher Pēteris Lesničenoks from LU ISP for obtaining SEM images.

I would like to express my deepest gratitude to my family which has grown larger over the years. It would not have been possible to make this true without the support, help, and faith of my husband, sister, and parents.

Contents

Abbreviations

AR – alkali resistant
CMC – critical micelle concentration
LOP – level of proportionality
MOR – modulus of rapture
OPC – ordinary Portland cement
GRC – glass fibre reinforced concrete
PGRC – glass fibre reinforced polymer concrete
UK – United Kingdom
SEM – Scanning electron microscope
XRD – x-ray deflection
FTIR – Fourier-transform infrared spectroscopy

General description of the research

The aim of the research

1. To identify the possible causes of cracks in ceramic and cement-based facade materials.
2. To find the most effective way to reduce defects using inert and active fillers.

Tasks

1. To identify and study the types of clays available in the Liepa quarry and their granulometric composition:
   - 1.1. to reduce the formation of cracks in solid bricks during drying by adding the surfactant Triton X-100 to the clay mass;
   - 1.2. to reduce crack formation in solid bricks by adding fibreglass to the clay mass during mixing;
   - 1.3. to identify the optimal granulometric composition of the clay mass in order to reduce the formation of cracks in solid bricks made using raw materials from the Liepa quarry.
2. To develop fibreglass reinforced concrete (GRC) compositions with and without added acrylic polymer and to determine:
   - 1.1. the effects on flexural properties depending on the length and amount of glass fibre in the composition;
   - 1.2. the autonomous self-healing mechanisms of concrete and to add crystalline additives to the composition to reduce crack growth after their initiation.

Scientific novelty

1. Ways to reduce cracks in bricks made of clay available in Latvia's largest clay quarry using innovative solutions have been studied: surfactants have been added, as well as the granulometric composition of clay mass has been optimized.

2. For the first time the effectiveness of crystalline additives for self-healing cracks in cement-based materials – GRC and PGRC – was studied.

Practical novelty

The use of ceramic and cement-based materials in construction is an environmentally friendly solution. Such materials combine properties such as high strength and durability and the ability to control humidity, creating a healthy indoor microclimate. However, in the production process of construction materials there is a relatively large amount of defective products (30–40 %), which is caused by cracks and due to which the manufactured products are rejected. Also, cracks in the finishing materials during deployment significantly reduce their longevity.

The Thesis has a significant practical aspect, as it was developed in cooperation with two Latvian companies LODE Ltd and Skonto Concrete Cladding Ltd. In cooperation with LODE Ltd, an analysis of clays from the Liepa quarry was performed and their impact on the formation of cracks in ceramic materials was assessed, as well as solutions for crack reduction at various technological stages was sought. Skonto Concrete Cladding Ltd is a company that manufactures innovative products with fibreglass reinforced concrete (GRC). In cooperation with this company, the material was investigated by changing the raw materials and investigating the effect of AR fibreglass on the flexural properties. With the goal of eventually creating an effective material, the self-healing of GRC cracks using crystalline additives was investigated. Such a study is important for any building materials plant to be able to predict and prevent the formation of cracks in the material by improving its quality.

Structure of the Thesis

The Thesis is a summary of scientific publications focused on crack reduction in ceramic and cement-based facade materials.

Approbation and publications

The results of the dissertation have been published in 7 SCI publications (5 of which are publications in conference proceedings) and have been presented in 10 international conferences.

List of publications

Publication No.	Reference
Publication I	**S. Guzlena**, G. Sakale, S. Certoks. Clayey Material Analysis for Assessment to be Used in Ceramic Building Materials. *Procedia Eng.*, Vol. 172, pp. 333–337, **2017**. https://doi:10.1016/j.proeng.2017.02.031
Publication II	**S. Guzlena**, G. Sakale, S. Certoks, L. Grase. Sand size particle amount influence on the full brick quality and technical properties. *Constr. Build. Mater.*, Vol. 220, pp. 102–109, **2019**. https://doi.org/10.1016/j.conbuildmat.2019.05.170.
Publication III	**S. Guzlena**, G. Sakale G. Alkali Resistant (AR) Glass Fibre Influence on Glass Fibre Reinforced Concrete (GRC) Flexural Properties. In: Fibre Reinforced Concrete: Improvements and Innovations. BEFIB **2021**. RILEM Book series, vol. 30. Springer, Cham. https://doi.org/10.1007/978-3-030-58482-5_24
Publication IV	**S. Guzlena**, G. Sakale, S. Certoks, A. Spule. Crack Reduction during Drying Process by Using Surfactant. MATEC Web Conf., Vol. 278, pp. 2–5, **2019**. https://doi.org/10.1051/matecconf/201927801008
Publication V	**S. Guzlena**, G. Sakale, S. Certoks, L. Grase. Effect of the Addition of Fibreglass Waste on the Properties of Dried and Fired Clay Bricks. *IOP Conf. Ser.: Mater. Sci. Eng.*Vol. 251, pp. 1–7, **2017.** https://doi:10.1088/1757-899X/251/1/012014
Publication VI	**S. Guzlena**, G. Sakale. Self-Healing Concrete with Crystalline Admixture – A. *Technol. Mater. Sci.* Ed., Vol. 34, pp. 1143–1154, **2019**. https://doi:10.1088/1757-899X/660/1/012057
Publication VII	**S. Guzlena**, G. Sakale. Self-healing of glass fibre reinforced concrete (GRC) and polymer glass fibre reinforced concrete (PGRC) using crystalline admixtures. *Constr. Build. Mater.*, vol. 30, p. 120963, **2021**. https://doi.org/10.1016/j.conbuildmat.2020.120963

Results of the research were presented at the following conferences

1. **S. Guzlena,** G. Sakale. Alkali resistant glass fibre influence on glass fibre reinforced concrete (GRC) flexural properties. *RILEM-fib International Symposium on FRC (BEFIB 2020).* Valencia, Spain, September 21–23, **2020**.
2. **S. Guzlena**, G. Sakale, D. Bajāre. Crack healing of glass fibre reinforced concrete using crystalline admixtures. *PhD Students' and Early Career Investigators' Meeting – Self-healing concrete structures*. Novi Sad, Serbia, March 7–8, **2019**.
3. **S. Guzlena**, G. Sakale. Self-healing concrete with crystalline admixture – a review. *4th International Conference "Innovative Materials, Structures and Technologies" (IMST 2019).* Riga, Latvia, September 25–27, **2019**.

4. **Guzlena S**, Sakale G. Self-healing concrete with crystalline additives. *20th International Conference on Fiber-Reinforced (ICFRC 2018)*. UK, London, November 19–20, **2018**.
5. **Guzlēna, S**., Spule A. Šakale, G., Čertoks, S. Triton X100 addition influence on brick formation. *2nd International Conference on Building Materials and Materials Engineering - ICBMM 2018*, Portugal, Lisbon, 26–28 September 2018.
6. **Guzlēna, S**., Spule A., Šakale, G., Čertoks, S. Crack reduction in clay bricks by surfactants. *In 8th International Conference on Silicate Materials "Balt Silica"* Latvia, Riga, 30 May – 1 June **2018**.
7. **S. Guzlēna**, G. Šakale, S.Čertoks. Fiberglass additive effects on the technological properties of the clay bricks. *The 3rd International Conference on Innovative Materials, Structures and Technologies (IMST 2017)*. Riga, Latvia, 27–29 September **2017**.
8. **S. Guzlēna**, G. Šakale, S. Čertoks. Sand size particle amount influence on the ceramic building material properties *Riga Technical University 57th International Scientific Conference "Materials Science and Applied Chemistry"*. Riga, Latvia, October 21–22, **2016**.
9. **S. Guzlēna**, G. Šakale, S. Čertoks. Clayey material analysis for assessment to be used in ceramic building materials. *12th International Conference "Modern Building Materials, Structures and Techniques*. Vilnius, Lithuania, May 26–27, **2016**.
10. **Guzlēna, S**., Šakale, G., Tirzmale, D., Čertoks, S. Clay Granulometry Influence on the Formation of Cracks in the Ceramic Building Materials. *1st International Interdisciplinary Symposium "Clays and Ceramics"*, Latvia, Riga, 28–29 January **2016**.

Declaration of authorship in publications I–VII

Sandra Guzlēna conducted a major part of the experiments, evaluated the results, and wrote all of the appended papers. In general, the co-authors contributed with experiment planning and constructive criticism of the obtained results which increased the scientific quality of the publications.

Publication No.	Reference	Corresponding author	Evaluation of Guzlēna 's contribution
Publication I	S. Guzlēna, G. Šakale, S. Čertoks, "Clayey Material Analysis for Assessment to be Used in Ceramic Building Materials," *Procedia Eng.*, vol. 172, pp. 333–337, 2017.	S. Guzlēna	S. Guzlēna developed 100 % of the experimental work, analysed and formatted all of the results of the research in accordance with the requirements of the journal, and fully prepared the publication manuscript.
Publication II	S. Guzlena, G. Sakale, S. Certoks, L. Grase, "Sand size particle amount influence on the full brick quality and technical properties," *Constr. Build. Mater.*, vol. 220, pp. 102–109, 2019.	S. Guzlēna	S. Guzlēna developed 90 % of the experimental work, analysed and formatted all of the results of the research in accordance with the requirements of the journal, and fully prepared the publication manuscript.
Publication III	S. Guzlena and G. Sakale, "Alkali Resistant (AR) Glass Fibre Influence on Glass Fibre Reinforced Concrete (GRC) Flexural Properties," *RILEM Bookseries*, vol. 30, no. September, pp. 262–269, 2021.	S. Guzlēna	S. Guzlēna developed 100 % of the experimental work, analysed and formatted all of the results of the research in accordance with the requirements of the journal, and fully prepared the publication manuscript.
Publication IV	S. Guzlena, G. Sakale, S. Certoks, A. Spule, "Crack Reduction during Drying Process by Using Surfactant," *MATEC Web Conf.*, vol. 278, p. 01008, 2019.	S. Guzlēna	S. Guzlēna developed 80 % of the experimental work, analysed and formatted all of the results of the research in accordance with the requirements of the journal, and fully prepared the publication manuscript.
Publication V	S. Guzlena, G. Sakale, S. Certoks, L. Grase, "Effect of the addition of fibreglass waste on the properties of dried and fired clay bricks," *IOP Conf. Ser. Mater. Sci. Eng.*, vol. 251, no. 1, 2017.	S. Guzlēna	S. Guzlēna developed 90 % of the experimental work, analysed and formatted all of the results of the research in accordance with the requirements of the journal, and fully prepared the publication manuscript.

Publication VI	S. Guzlena, G. Sakale. Self-Healing Concrete with Crystalline Admixture – A Review. J. Wuhan Univ. Technol. Mater. Sci. Ed., Vol. 34, pp. 1143–1154, 2019.	S. Guzlēna	Following the instructions of the supervisor, S. Guzlēna fully prepared the review article in accordance with the requirements of the journal and submitted the paper as a corresponding author.
Publication VII	S. Guzlena and G. Sakale, "Self-healing of glass fibre reinforced concrete (GRC) and polymer glass fibre reinforced concrete (PGRC) using crystalline admixtures," *Constr. Build. Mater.*, vol. 30, no. September, p. 120963, 2019.	S. Guzlēna	S. Guzlēna developed 90 % of the experimental work, analysed and formatted all of the results of the research in accordance with the requirements of the journal, and fully prepared the publication manuscript.

Scientific supervisor
Dr. sc. ing. Gita Šakale …………………………………………………

Theses to defend

1. The prevention of cracks in ceramic building materials is a complex process that is affected by the following technological stages – establishing the granulometric composition of raw materials, mixing and forming, drying, and firing:
    a. Adding the surfactant Triton X-100 close to the critical micelle concentration (CMC) decreases the crack area of solid bricks during drying by 30 %. The surfactant molecules help water molecules to move more easily from the brick centre to the surface thus reducing the mechanical stresses between the dry brick surface and the wet brick core.
    b. Increasing of the amount of sand-sized particles to 59 % in the granulometric composition of the raw materials not only reduces the average crack area in dried samples but also reduces the drying shrinkage by 30 %. Reducing of the clay-sized particle amount and thus the absorbed water in the raw materials reduces mechanical stresses during the drying process, which otherwise cause sample cracking.
2. During the first 8 weeks after crack initiation the average self-healing rate per week is 12 µm for GRC samples and 17 µm for PGRC samples. The self-healing rate of PGRC samples is higher than that of GRC samples because the crystalline admixtures are "encapsulated" in thin acrylic polymer film, which prevents them from reacting during mixing, providing more crystalline centres for crack growth after crack initiation.

Crack reduction in facade materials

1. Introduction and background

Urbanization and industrialization in the last decades have caused a great need for living space in cities all over Europe. Urbanization is a complex process which is characterized by population migration, economic development, land-use change, and lifestyle reform [1], [2]. A new significant neighbourhood development with private and public buildings is taking place. Ordinary Portland cement (OPC), due to its many functions and applications ranging from load-bearing elements to facade materials, is a widely used building material, and it is highly available due to the comparatively low costs of raw materials and processing technologies [3]–[5]. In recent years, poured concrete walls left without any post-treatment interior and exterior, called exposed concrete or architectural concrete, have become increasingly popular. Other typical facade materials are glass, aluminium alloy, stone, and ceramics [6]. By mixing all these materials architects can achieve stunning building shapes and colours. To have a sustainable and a highly durable facade, it is important to use high quality materials. However, to achieve high quality materials, highly qualified specialists must be employed and manufacturing technologies must be improved.

During the building process and the lifetime of a building, a lot of energy is used, starting from the raw material production and including the building process itself. Green building is an important measure introduced to deal with energy and environmental problems in the construction sector of the world. The US Environmental Protection Agency has defined the term green building [7]. According to it, green building is the practice of creating structures and using processes that are environmentally responsible and resource-efficient throughout a building's life-cycle from design, construction, operation, maintenance, renovation and deconstruction [8]. Sustainable construction, green construction, high-performance construction, and therefore, the corresponding buildings are one of main research topics in construction science [9]. It is vital to use novel materials and technologies and for the architect to work in close connection with builders and materials manufacturers. Architects are more often looking for something more extraordinary with complex shapes, coluors, and materials. Buildings instead of box shape forms are now more complex and architectonic. In Fig. 1, one can see a social house designed by Mikhail Riches and Cathy Hawley, which has been awarded the biggest prize in UK architecture – the Royal Institute of British Architects Stirling Prize 2019. This is a low-energy, highly sustainable modern building [10].

Fig.1. Goldsmith Street, Norwich, UK [10].

A building of a very complex shape known as The Broad was opened in Los Angeles, USA, in 2015 and is presented in Fig. 2. It is comprised of 2,500 fibreglass-reinforced concrete panels, and 650 tons of steel were used for the facade [11]. This building is representative of the current possibilities to shape a building with novel facade materials.

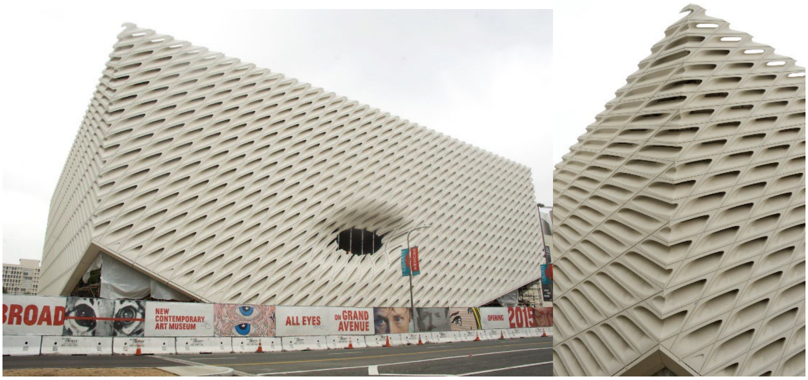


Fig.2. Los Angeles, USA [11].

Buildings by the local firm Blitch/Knevel Architects provide great examples of complex shapes and of the use of many materials, like ceramics, stone, wood, metal, glass, and concrete in harmony. An example is presented in Fig. 3 –public University Medical Center in El Paso, Texas, USA [12].

Fig. 3. University Medical Center, El Paso [12].

Maintenance is vital in achieving long-lasting housing stock and if not done, then the early ageing of building's elements superficially can be accelerated [13]. The building's maintenance and cleaning of the curtain wall accounts for the majority of energy and resource costs. According to Perret [14], about 75–80 % of costs occur during the use and maintenance stage of building with 50 years of service life. So, the right choice of facade materials is essential.

The building envelope, facade, windows, rainscreens, etc. determine the comfort level in the building. In this context, the facade determines the main building appearance and is responsible for permeability of external aggressive environment from outside the building to the inside. A facade consists of many complex components such as windows, claddings, walls, openings, etc. so, it is one of the most complex systems to design, build and maintain. The primary source of defects on a facade is moisture, but dirt, extreme temperature changes, pollution, salt, microorganisms, and loads and stresses influence facade performance in the long term. Main defects caused by the mentioned factors are cracks, stains, and loss of adhesion. According to the research of Pires et al. [8], stains from dirt deposits on facades are the primary defect. Research regarding ceramic tiles used for facades has been done, and for primary defects two types have been detected: staining and change of colour or brightness of the tiles and change of colour of joints. Both defects are related to dirt deposits on the facade. When natural stone claddings are used, according to Neto and de Brito [10], colour variation is the most common defect. To reduce these defects, a maintenance plan which includes inspection, cleaning, surface protection treatment, local repair, local replacements, and minor and major interventions is required [13].

2. Ceramic facade materials

Ceramics based building materials, like tiles and bricks, have been known for more than 2000 years, but nevertheless they remain modern, environmentally friendly facade materials. Ceramics based products are widely used all over the world because of their durability, high strength, good thermal conductivity, resistance to various weather conditions, and the ability to control humidity and ensure a healthy indoor climate [15].

Ceramics production begins by quarrying materials such as clay and sand. Before the material is processed, it is pre-tested at the quarry by drilling and determining the depth, area, and granulometry of the material. Clay from the quarry is stored in cones. Cones are made from 20 to 30 centimetres thick clay layers based on data from previous drillings. Clay can remain in cones throughout the year, as temperature fluctuations make the clay more plastic. The clay is then mixed and then used for production [16].

In addition to the clay mass, various fillers such as sand and tenesite, burnt fillers such as shavings and coal are added. After adding all the necessary fillers, the mass is mixed and then extruded. The product obtained has a moisture content of about 20 %, so it is dried until 3 % and then fired at around 1000 ºC [17].

However, the quality requirements for the finished product continue to grow. Production needs to control and adjust additives to reduce the granulometric variation. Inconsistency of mixture granulometry can affect the quality and properties of the final product [18].

Mixing of the raw material is one of the most important processes before forming the product. A poorly blended raw material can induce defects during the forming, drying and firing processes, and these defects can be detected only at the end of each step. The primary purpose of mixing is to obtain a completely homogeneous mass ready for extrusion, which means that all samples taken from the thoroughly mixed mixture must have the same composition. Unfortunately, it is practically impossible to obtain such a fully blended mixture by mechanical mixing. The mixing process is influenced by:

- component density,
- raw material granulometry,
- particle shape,
- moisture content.

There are several methods for pre-extrusion mixing – using rotary blades, filter pushing, and others – to achieve a fully blended mixture [19].

The second most important process after mixing is the forming by extrusion. Defects occurring during this process may not be noticeable at the time but will appear in the form of cracks or deformations after drying or firing. An auger extruder is usually used in the production of building ceramics, but a piston extruder can also be used. The type of auger used is also important. There are single-screw and two-screw extruders. Their combinations and directions of rotation can be different and customized for the clay mass and mixture properties [19].

During extrusion, cracks develop as a combination effect due to the properties of the raw material and the physical processes caused by homogenisation and forming operations in the extruder, which can lead to different defects in the product.

Attempting to prevent cracks in the material by changing the geometry of the muffler reduces, but does not eliminate the problem, as it is caused by earlier stages of the process, such as clay retention, grinding, or mixing.

It is recommended that the problem causes are divided into three groups (Table 1):

- clay variables,
- technological variables,
- mechanical process variables.

Table 1

Reasons for Crack Formation after Extrusion by Probst [19]

Ceramic variables	Technological variables	Mechanical process variable
• The type of clay used • Mineralogical composition • Granulometric composition • Water quantity/mass flow • Other additives	• Preparation/homogenization • Extruder auger speed • Moisturizing or lubricating the extruder head • Intermediate transportation between the press and dryer and maintenance of moisture • Drying • Firing	• Screw geometry • Pointed screw head • Cylinder housing • Extrusion head • The extruder muffler

Clay minerals such as kaolinite, haloizite, montmorolite, or illite have a particle size of about 5–1000 nm. Clay minerals have large surface areas (5–100 m^2/g), and anisotropic morphology [19]. According to the literature, crack formation is generally classified into the following types:

- crack formation due to flow,
- cracking during beams cutting,
- defects in the air intakes,
- crack formation under vacuum,
- cracks caused by inhomogeneous clay mass [19].

Main defects often observed on facades are joint failure, cracking, delamination/detachment, and efflorescence. The main reason of defect appearance is increased moisture content due to precipitation, rising dampness, condensation, or faulty maintenance [20].

In the literature about clays in new and existing quarries, research on their granulometry and technologic properties for brick production can be found [21]–[23]. Still, papers where research has been done and results discussed about material cracking, crack measurements, detection of cracking, and elimination potential are rare. The research for this dissertation was done with the goal to reduce and analyse cracks. Methods used by authors were changing the granulometry of the mixture, adding surfactants to improve water evaporation during the drying process, and using glass fibres to achieve reinforcement effect during the firing process. After sample drying and firing, crack measurements were done using a microscope to analyse the influence of different variables.

In the author's research, described in **paper I**, **paper II paper IV and paper V,** materials from one of the biggest quarries in Latvia were analysed with the aim to reduce the amount of cracks in solid bricks using different methods. To minimize the formation of cracks, it is necessary to decrease the amounts of fine and anisotropic particles, which are mainly clay minerals in different proportions. Cracking problems amplify with increasing specific surface area of the particles. Other materials, such as talc with laminated structure, also tend to form cracks during extrusion.

On the other hand, clay minerals have the property of plasticity, which is vital for extrusion processes. A balance between plasticity and crack formation must be found. For structural clay products, the optimal grain size distribution is essential. Fractions below 2 μm in size should not exceed 50 % of the total mixture. The empirically derived data is compiled in a diagram in Fig. 4 a). Often cracking can be reduced by increasing the amount of coarse material in the mixture, for example by adding a coarse-grained additive – gravel or crushed fired clay products. At present, coarse grained additives with precise granulometric and chemical compositions are available from various suppliers [19], [24], [25].

The addition of coarse material reduces shrinkage of clay products and reduces the risk of cracking during the drying and firing process. However, replacing the clay with a coarse-grained additive will reduce plasticity [19], [24], [25].

The moisture of the material affects the formation of cracks during technological processes, but the content and homogeneity of the mixture are also important here. Variations in the moisture content of the mix can lead to significant cracks and their consequences. Therefore, drying should be done slowly and gently so that moisture is evenly distributed throughout the body [19].

In Latvia clay sediments of all periods, predominantly Devonian and Quaternary, less so Triassic and Jurassic, are used to produce ceramic building materials. The most significant quarry in Latvia consists of clay soils of Devonian time. The quarry contains different soils – from clay and clay aleurolite to sandstone, but dominant are different red-coloured carbonate-free clay aleurolites. Aleurolites are loose rubble rocks consisting mainly of quartz, feldspar, and mica. Aleurolite granulometry is 0.1–0.01 mm, thus between clay and sand grain size [26], [27]. Three types of clay – grey, red, composite – were used from Latvia's largest quarry. In **Paper I** the granulometry and mineralogy of samples are analysed with the overall aim to produce solid bricks. Granulometry of the clay mass used for manufacturing ceramics is vital because it greatly influences the technological processes of the ceramics products, for example, the behaviour of the material during the forming, drying, and firing processes. High clay-size particle amount will cause a high chance of cracking during the drying process and a high shrinkage of a product during the firing process [28]. According to Mcnally [24], the proposed particle distribution limits for solid bricks, perforated bricks, and roof tiles are shown in a triangular diagram (Fig. 4 a)). The diagram consists of three major soil particle size distributions – clay particles (<2 μm), dust particles (2–20 μm), and sand particles (>20 μm) [24].

Analysing the available clay mass discussed in **Paper I**, the most suitable granulometric composition for ceramic building material production was found to be composite clay due to its balance between all grain size fractions [24], [29], [30]. Grey and red clay have a low amount of sand-size particles, which could cause significant material shrinkage during the drying and firing processes. In a quarry, the available sand can be used as an additive to plastic clays to reduce brick shrinkage during the production. According to XRD results, illite and kaolinite clay minerals were detected in samples. To have clay mass with desired properties, the proportion of illite, kaolinite, and quartz should be controlled. Each of the minerals has influence on every step of the production starting from mixing and extrusion and ending with the ceramic building material firing in a kiln [24].

As concluded in **Paper I** the clay available in the quarry is very plastic due to the high clay-size particle amount. Clay particles provide plasticity to extrude, shape, or press material into a mould. Clay particles also influence material sintering and the mechanical properties of the final product. Evaluating the data available in the literature, it was concluded that the sand-size particles must be added to have the appropriate granulometry for the finished bricks, according to the recommended particle size distribution for solid bricks [24].

In **Paper II** the granulometry of the mixture was changed by varying the sand-size particle amount from 33 % to 59 %. A laboratory extruder was used to make solid brick samples. After formation, samples were dried at 105 ºC and fired at 1000 ºC for 1 h. Sample water absorption, shrinkage, density, and compressive strength were measured after the firing process. A digital camera and an SEM-EDX were used to determinate the crack area after drying and after the firing process.

The sand-size particle function is to control shrinkage and lamination and to provide gas drainage paths during the firing. Addition of sand to clay mass can reduce defects during the drying process. During the firing process silicon dioxide or quartz (SiO_2) which occurs naturally in clay and shale, melts. After firing and during the cooling process, quartz forms a bond between clay particles. During the cooling process the development of cracks can be induced due to quartz volumetric changes of about 0.8 % at 573 ºC, which is caused by its crystallographic modification [31], [32].

After clay mass is mixed and formed, the bricks are dried in a drying tunnel and some defects, like cracks, can be visually observed and a quality check can be performed. As described in **Paper II,** the drying rate must be controlled and set correctly to eliminate a too rapid temperature increase. The brick temperature during the drying process is increased 30 to 80 °C to evaporate the free water from bricks and decrease the moisture content to 3 % [17]. During the next technological step – firing – fracture and cracking are mostly caused by the volume changes of materials. Usually, it happens with SiO_2, which transforms from the alpha to the beta phase and changes its volume as described above [33]. So, the firing must be done at a controlled rate. Kadir et.al. changed heating rates during the firing process from 0.7 to 10 °C/min and concluded that brick properties start to deteriorate if the heating rate is 2 °C/min or more [34]. In **Paper II** samples were dried at 105 °C. The firing was done at 1000 °C for 1h at a heating rate of 5 °C/min. Five different clay mass mixes were made, and the sand-size particle amount was varied from 33 % to 59 %. In Fig. 4 c), the mix granulometric composition of all blends is presented. The positions of the mixes in the triangular particle size diagram are shown in Fig. 4 b).

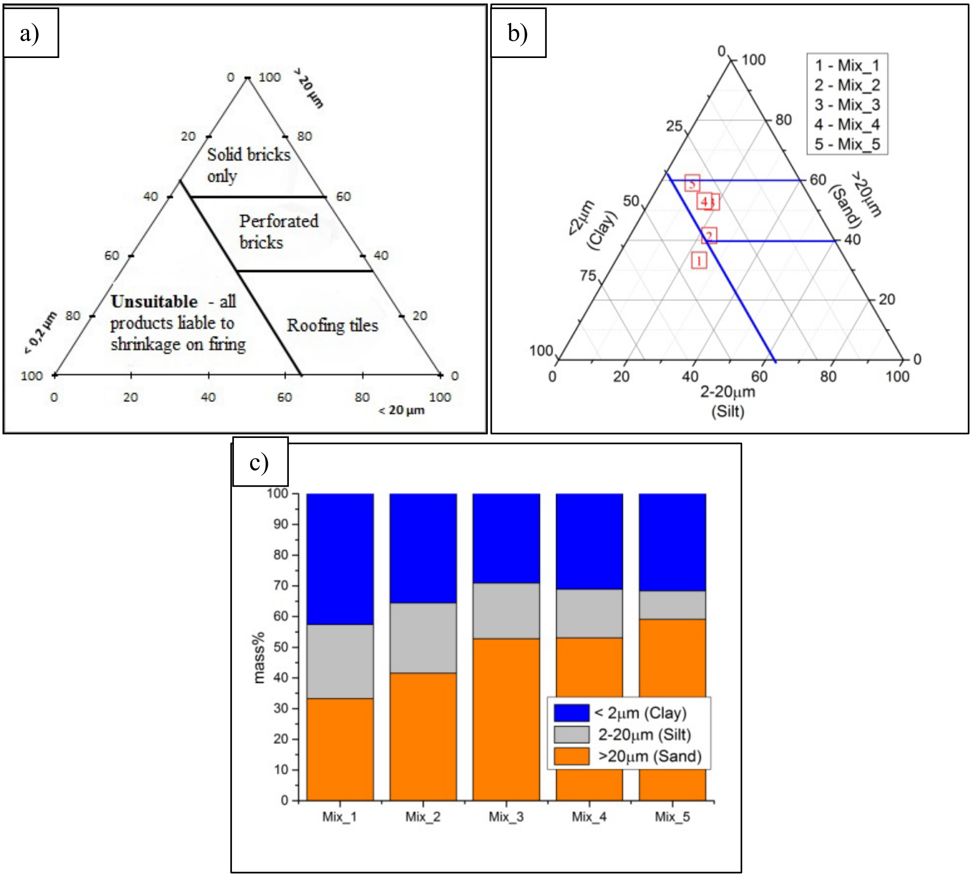


Fig. 4. a) – Suggested particle size range for brick and tile clays [24]; b) – mix location in a diagram; c) – mix granulometric composition [35].

From analysing the dried samples, it was determined, as shown in table 1 in **Paper II,** that the average sample moisture content was determined during forming changes from 14.4 to 20.8 %. But as shown in the third column, the average drying shrinkage does not depend solely on the sample humidity. Instead, the average drying shrinkage also correlates with the clay particle amount in the sample. As is known, clay has a layered structure. There are water molecules between these layers, and during the drying the water molecules evaporate and the layers move closer to each other, which results in the sample shrinkage [21].

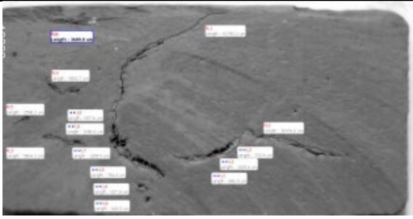
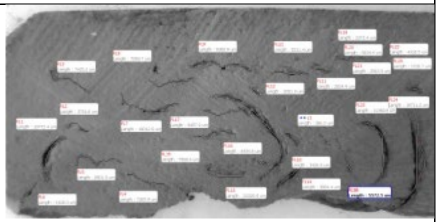
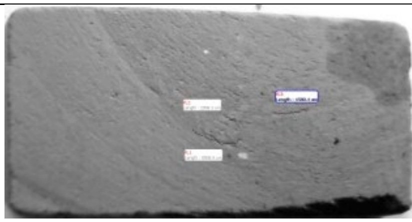
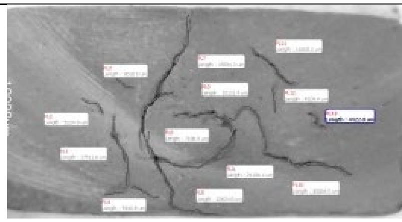
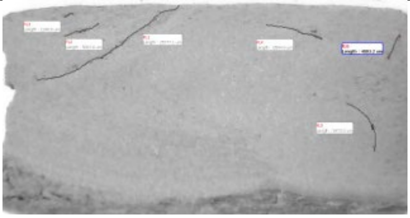
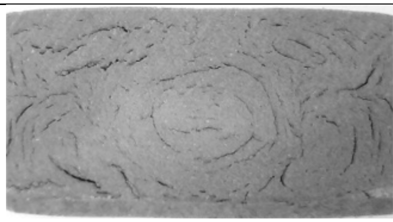
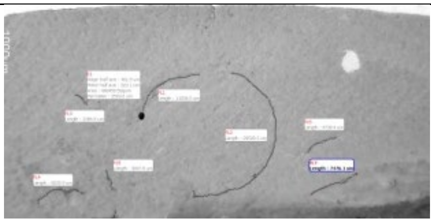
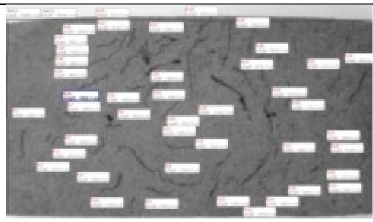
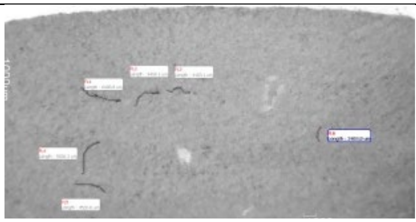
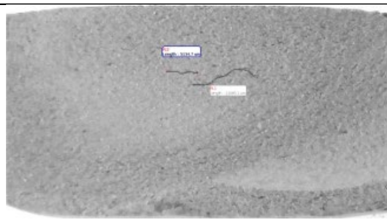
In the table 4 of **Paper II,** the following relationship was demonstrated: by increasing the sand-size particle amount in the sample mix, the average crack area of the dried sample surface decreases. That can be explained as before, where by decreasing the fraction of clay particles which absorb water molecules and by increasing the fraction of sand-sized particles, the drying-induced stresses in the sample decrease and cracks do not form [21], [36]. In addition, the drying rate has a significant influence on cracking during the drying process. If the drying process is fast, brick drying tends to be more irregular, leading to the formation of more flaws, which leads to the generation of more fractures. Therefore, drying must be done evenly [37].

For the fired samples, table 3 in **Paper II** shows the measured compressive strength. it can be concluded that higher compressive strength can be achieved when all fractions in the clay mix are in approximate equilibrium. From the data given in **Paper II,** table 4 one can conclude that the average crack area of the fired samples does not show any correlation to the granulometry or the average crack area of the dried samples. But the average crack area can be related to the average heating weight loss if **Paper II,** table 2 and table 4 are compared. The samples with a lower average crack area have a lower heating weight loss. It can be explained by a rapid temperature increase in two sensitive temperature zones. The first temperature zone is 100–150 °C, where water evaporates from pores. The second temperature zone is 500–900 °C, where hydroxyl water evaporates and where at around 573 °C, quartz changes from the alpha phase to the beta phase [35].

In **Paper II** cracks in the samples are analysed by their length. Visual crack measurements are shown in Table 2. It can be concluded that samples with a larger average crack area, like Mix_1, have a higher number of cracks longer than 5 mm. Differences between dried and fired samples are significant. In every mix that was made, crack area increases after the firing process. After the firing, the largest crack area is for compositions where sand dominates, Mix_3 and Mix_4, excluding Mix_5 the composition of which, according to the clay material granulometric triangular diagram [30], is the closest to the ideal solid brick composition. However, Mix_5 shows the lowest compressive strength result. Optimum compositions by taking into account both the amount of cracks and compressive strength data are Mix_1 and Mix_2. The round-shaped cracks that appear in almost every sample at the centre come from the extruder auger. This type of crack is a combination of flow lamination and the rotational movement of the clay mass. Also, the hallow space created by the auger hub at the centre of the emerging clay mass has not entirely re-joined as a result of either mass characteristics or the geometry of the pressure head and die [19]. These hollow spaces in samples, which do not occur in dried samples, are visible in fired samples. There are two reasons for this type of cracks. First is the nature of quartz to transform from the alpha to the beta phase at around 573 °C, the second – the firing rate [38]. According to Arsenovic et al. [21], the firing temperature must be increased slowly (2–5 °C/min) during the first part of the process. Slow warming should be carried out up to 600 °C to minimize the possibility of cracks occurring during the quartz phase transformation from the alpha to the beta phase at 573 °C. During the second part the heating rate can be increased to 5–10 °C/min. The firing rate that was used in the author's research was 5 °C/min from room temperature to 1000 °C. In this case, probably the warming rate in the first part was too rapid and caused cracking in the samples. As Ukwatta et al. have concluded, the heating rate also influences other properties such as firing shrinkage, compressive strength, and water absorption. As the heating rate increases, the compressive strength of a brick decreases as a result of low vitrification time which bonds clay particles leaving them more porous and brittle [18], [39].

Table 2

Crack Measurements of Dried and Fired Samples [35]

Mix number	**Dried sample**	**Fired sample**
Mix_1		
Mix_2		
Mix_3		
Mix_4		
Mix_5		

After firing, the samples were analysed using SEM as shown in **Paper II,** table 5. Almost in every brick sample, sand grains of size 150 µm and larger were found. The area around grains is cracked due to the quartz phase change. In many papers [38], [40], [41] cracks around quartz grains have been noted. Allegretta et al. in their work [41] also encountered problems with cracking around quartz grains and found that by increasing the firing temperature from 750 °C to 1000 °C the detachment zone and the crack size increased.

If the cracking during the drying process is reduced, there is a high chance to reduce the crack area after the firing. The drying process is often one of the most complex operations in the manufacturing process. During the drying process, a large fraction of moisture is removed and the form and appearance of the material is set for the final product [42]. In **Paper IV** a surfactant was added to clay mass during the formation process to reduce crack generation in the drying process. The first part of the drying process is the most likely time when cracks appear, caused by the tension between the dry brick surface and the wet brick core. Cracks appear due to nonlinear moisture and/or temperature changes in the material. If surfactants are used, the tension between the surface and the material pore walls can be controlled in order to improve moisture transport from the brick core to the surface [43]. As Kowalski et al. [43] have investigated, if dedocilsulfate and fluoric surfactants are used at very low concentrations (0.001 %), crack formation is reduced. For clays with a tendency to swell, which can cause cracking, this risk is decreased and cracking reduced when amido-amin base cationic surfactants are used [44]. Most of the research done limited the concentration range to below the critical micelle concentration (CMC) or near the CMC in order to reach the maximum adsorption [43], [45]. CMC is the concentration where molecules of the given surfactant arrange into micelles and all additional surfactants added to the system go to micelles [43].

In the presented research the Triton X-100 was used around CMC. In **Paper IV**, table 1 show the contact angle measurements between a glass plate and droplets of Triton X-100 at different concentrations, which were made to determine the CMC. The CMC determination is shown in figure 5. The CMC can easily be determined to be at 0.001 %. The surface tension is mainly influenced by the CMC: at concentrations below the CMC the surface tension changes rapidly while above the CMC it remains almost constant. All samples were made with the surfactant concentration at around the CMC in order to achieve improvement in the moisture transport and subsequently less cracking during the drying. Three samples with the same granulometry but different Triton-X concentrations were made.

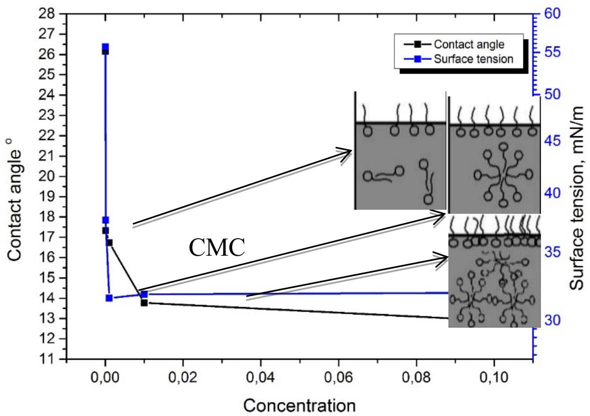


Fig. 5. Contact angle and surface tension dependence on concentration. CMC detection [46].

To evaluate and quantify cracks, the dried brick samples were sawed into pieces of 2–3 mm width. Crack measurements and analysis were performed using a digital camera (Moticam

2000) and the Motic Images Plus 2.0 software. Crack width was measured close to both ends and in the middle of a crack, and the average value was calculated. Crack fractional areas were calculated using Eq. (1).

$$\frac{Crack\ length\ \times Average\ crack\ width}{Sample\ length\ \times Sample\ width} \times 100 = Crack\ fractional\ area\ in\ \% \quad (1)$$

As shown in Table 3, the lowest average crack area of the dried sample was achieved in the sample with the molar concentration above the CMC for the non-ionic surfactant Triton X-100. In different publications there are different opinions regarding results for concentrations below or above the CMC [43]. But R. Guéganet et al. [45] state that concentration for the non-ionic surfactant (C10E3) above or near the CMC gives the maximum adsorption to montmorillonite clay surface. So, it can be concluded that the better is the adsorption of the surfactant to clay surface, the better is the water evaporation from the centre of the brick and the lower is the crack area after the drying.

Table 3

Crack Amount of Samples with Different Triton X-100 Concentrations [46]

Triton X-100 molar concentration	0.01	0.001	0.0001
Crack amount, %	0.07±0.0006	0.09±0.0007	0.10±0.0006

Our proposed explanation for the drying mechanism with surfactants is shown in Fig. 6. The hydrophilic heads of the surfactant molecules have approached the clay structure while the hydrophobic tails remain in pores. During the drying process, the water molecules are rolling along the hydrophobic tails and do not adsorb to other particles or interact with the structure. This mechanism helps water molecules move easier from the brick centre to surface.

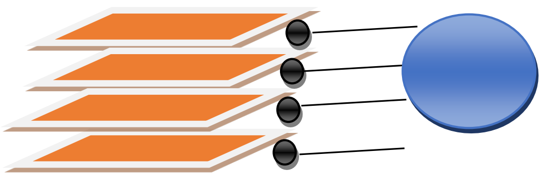


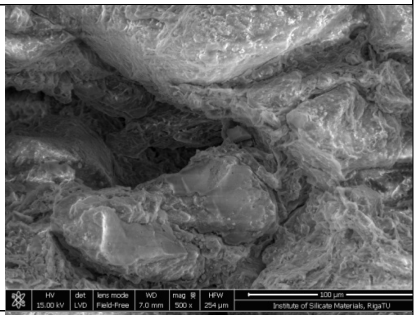
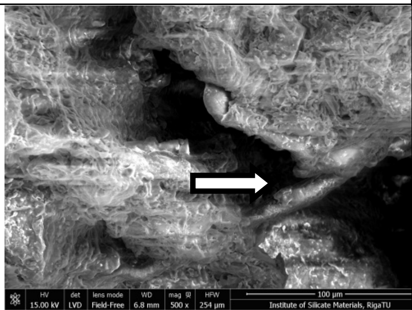
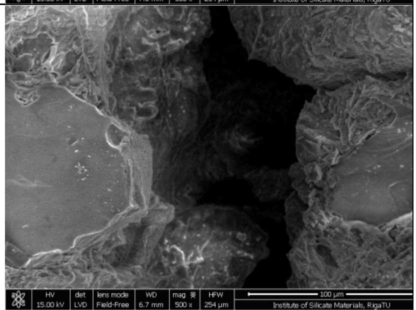
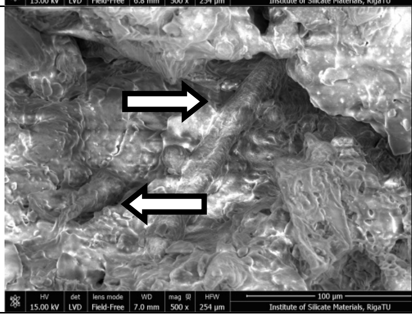
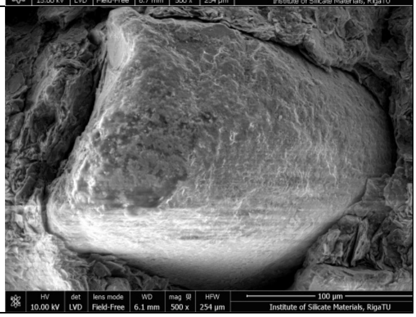
Fig. 6. Drying mechanism in clay when the surfactant is used [46].

A highly effective method for crack reduction is material reinforcement. The largest glass fibre manufacturer in Latvia – JSC "Valmieras stikla šķiedra" – has a high amount of manufacturing waste in the form of short fibreglass. The estimated amount of inorganic waste in Europe exceeds 1,500 million tones. Traditionally, non-hazardous inorganic waste is landfilled and is often disposed of directly in ecosystems without appropriate treatment [47], [48]. The production of bricks and ceramic tiles worldwide requires a huge amount of natural raw materials. The composition of these natural raw materials can be further changed by other raw materials, allowing different types of waste to be included in the internal structure of the bricks as part of their matrix. So, glass fibre waste can be utilized in clay bricks. It is a good way to substitute for natural raw materials and reduce their use. In collaboration with glass fibre manufacturer JSC “Valmieras stikla šķiedra”, manufacturing waste – short glass fibres – were used in waste recycling. In research presented in **Paper V** the main objective was to study the effect of adding glass fibre waste on the properties of clay bricks after drying and firing. Previous research done by other authors has shown positive effect on the properties of clay materials if glass waste was used [31], [47], [49]–[51]. Addition of glass to the ceramic matrix accelerates the densification process due to the fusion of the silica and alumina components and improves material properties such as compressive strength, water absorption, and others, even with small amounts of additive (below 10 wt%) [31], [49].

Using clay mass of given granulometry, samples with 0 %, 5 %, and 10 % glass fibre waste were made. In **Paper V** table 1 crack amount is expressed as the average crack area as a percentage of the sample area. Table 2 presents crack images and illustrates how cracks were measured. The results show that by increasing the glass fibre amount the crack area of the dried sample decreases consistent also with the results of shrinkage during the drying. Glass fibre in the sample matrix reduces the shrinkage during the drying, which is characteristic of clay materials. Thus cracking is avoided [47]. The results are opposite with fired samples. Both shrinkage during the firing and the crack area increases when the amount of the glass fiber additive is increased. That could be caused by silica phase transition at around 573 ºC and increasing its volume during the heating while shrinking during the cooling process [49]. Sand and glass fibre have a high amount of silica. Also, the rapid increase of heat can increase the evaporation rate of the absorbed water and cause cracking during the first stages of the firing process. Table 4 presents SEM images of all three mix samples fired at 1000 ºC. Different size quartz grains are found in the clay matrix. In samples without glass fibre, microcracks have formed around large quartz particles which are also the reason for the formation of larger cracks as discussed above due to the quartz volumetric changes during the heating and cooling processes.

Table 4

SEM Images of Mix_7 Without Fibreglass Additive, Mix_7_5 % with 5 % Fibreglass Additive and Mix_7_10 % with 10 % Fibreglass Additive [52]

Sample type	Glass fibre strands found in sample	Crack formation around sand grains
Mix_7 without fibreglass additive		
Mix_7_5 % with 5 % fibreglass additive		
Mix_7_10 % with 10 % fibreglass additive		

An increase in the amount of glass fibre additive increases the sintering shrinkage, as was discussed above, leading to a densification of the material. Glass fibre additions change the recrystallisation processes, leading to the formation of dense clay-glass agglomerates [49]. Clay-glass agglomerates affect the compressive strength. Sample density, after firing, increases with increased amount of the glass fibre additive. With increasing density of clay bricks, the compressive strength also increases. Thus, the compressive strength increases with increased amount of glass fibre additive. As discussed above, fibreglass has good adhesion with clay structures, and that works like a reinforcement, which strengthens the structure. Our sample's compressive strength is very similar to that published by C. N. Djangang et al. who obtained the compressive strength of 10.49 MPa for a 10 % glass powder addition to kaolinite clay [49], [53]. With a higher compressive strength, other properties like flexure and resistance to abrasion are also improved. While other properties are relatively difficult to evaluate, the compressive strength is easy to determine [31].

By adding glass fibre waste to clay mixtures, the desired effect in the final product was not achieved and the amount of cracks after the firing increased, but we obtained several advantages. First, the amount of waste was reduced and, second, properties of clay bricks were improved. The addition of glass fibre waste (size range 20 to 2 μm) showed good influence on the mechanical characteristics of clay bricks in that the compressive strength and density increased by increasing the amount of fibreglass in the mixture. Fibreglass worked as reinforcement for the clay matrix.

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3. Cement-based facade materials

Unlike ceramic materials, cement-based materials do not have quality problems during the production process, but rather during their service life. Concrete is a widely used material in buildings starting from load-bearing structures to decorative elements. Different types of visual and structural defects can appear in concrete structures during a long-term service. Defects, like cracks, can appear due to plastic shrinkage, strengths reduction of concrete, etc. and result in the material strength reduction over a more extended time period. It is caused by the exposure of concrete structures to aggressive agents such as chlorides, sulphides, carbonates; by water penetration through pores and small cracks due to absorption or due to hydrostatic pressure; and by freeze-thaw cycles [54]. Defects on facades are mainly from mechanical impacts, environmental exposure, or due to inappropriate maintenance. To reduce these risks, proper maintenance of concrete structures is necessary [55]. De Rooij [56] stated that in the UK 45 % of the annual construction costs are spent on the maintenance of concrete structures. To decrease these costs, novel materials and techniques are to be designed. The choice to use in construction concrete with the self-healing ability is like an insurance for the given construction for a long life and reduced repair and maintenance costs. As discussed in **Paper VI** self-healing materials have many long-term advantages: fewer man-hours spent on inspection, monitoring, and repair work on site and thus increased money savings. Concrete with the self-healing ability does not need human interaction to start the healing process. The self-healing abilities of concrete [54], stainless steel reinforced concrete [57], high-performance concrete [58], and high-performance fibre reinforced concrete (HPFRC) [54], [58] have been researched by many authors. The self-healing can be divided into two mechanisms:

- Autogenic healing: natural closure of cracks due to un-hydrated cement in the concrete with no specific self-healing admixtures. This type of self-healing is not predictable and cannot be controlled [59]–[61].
- Autonomic healing: engineered closure of cracks using additives which are not part of the ordinary cement [58]–[60], [62], [63]:
    - passive – no need for human involvement, the self-healing starts due to external promoters [57], [64];
    - active – human involvement required to activate and complete the self-healing mechanisms [57], [64].

Numerous researchers have investigated autonomic self-healing mechanisms with fibre reinforcement [54], [58], bacteria [65]–[68], crystalline additives [54], [58], [63], [69], [70], absorbent polymers [73], absorbent clay materials [62], encapsulated healing agents [59], [62], [74], expansion agents like sulfo-aluminate [62], [69], [70], and others.

The crystalline admixtures available on the market consist of cement, sand, and a crystalline admixture or "active chemical" which is the producer's proprietary information [71], [75]. A crystalline admixture only starts to react when a high enough level of moisture is reached. It reacts with $Ca(OH)_2$ and other products that appear during the cement hydration. Elsalamawy [72] has concluded that by increasing w/c ratio, the efficiency of crystalline admixtures increases. But if the concrete is made with low w/c ratio, no significant difference in water absorption and crack closure between samples with/without crystalline admixtures has been observed.

3.1. Glass fiber reinforced concrete (GRC)

It is hard to control crack size during cracking due to the brittleness of concrete. To increase the tensile strength and control, the crack size different fibres, like organic, polymer, basalt, steel, and glass fibres can be incorporated in the matrix.

Glass fibre reinforced concrete (GRC) is a composite which consists of concrete and glass fibre. Glass fibre has low water absorption, high modulus of elasticity, and high tensile strength [76]. Fenu et al. have even concluded that basalt fibre reinforcement shows lower performance than glass fibre reinforcement when used under dynamic conditions [77]. GRC has been used for non-structural parts of buildings like facade panels for over 30 years. GRC is known for its high tensile and impact strength which is due to glass fibres. These properties allow making GRC panels 12 mm thin [78]–[80]. GRC can be produced by two methods. The first is a premix method, where concrete is mixed with chopped glass fibre and then cast in a formwork. The fibre length is usually around 12 mm, and it is added to form 3 % of the mortar mass. If the fibre length and amount is increased, then the workability of the mix is reduced. Barluenga et al. have noted that by using short fibres in the premix the cracked areas of concrete surfaces can be reduced. If a crack appears perpendicular to a fibre, it limits the size of the crack, but if the crack appears parallel to a fibre, then the reinforcement does not work and the crack size grows freely [81]. The second method is the spray technique, and for it the fibre length can be varied, but usually the fibres are 25–40 mm in length and 5 % by mortar weight. During the spraying process a uniform and well mixed composite is achieved compared to the premix method. This type of material has high durability and high impact and tensile strengths [79], [82].

As discussed in **Paper III** the glass fibre used in the material must be alkali-resistant (AR) due to the high alkalinity of concrete. In the past, ordinary glass fibre, like E glass fibre, has been used, but over time it has shown low durability. Fibres lose their durability due to the hydration reaction in the concrete. Portlandite ($Ca(OH)_2$) is produced during the hydration process. It increases alkalinity in the cementitious matrix to high alkalinity (pH 12). Hydroxyl ions (OH^-), which are at a high concentration in the cementitious matrix due to the alkaline solution in pores, break the Si-O-Si bonds in glass fibres (Eq. (3)) and causes weight and diameter losses making fibres fragile [80], [82]–[84].

$$- \underset{|}{\overset{|}{Si}} - O - \underset{|}{\overset{|}{Si}} - + OH^{-} \rightarrow \underset{|}{\overset{|}{Si}} - OH + SiO^{-} \quad (3)$$

To improve glass fibre chemical stability in concrete, ZrO_2 is added. According to standard EN 15422, AR glass fibre must contain a minimum of 16 % of ZrO_2 for it to be used in concrete [85]. But the addition of ZrO_2 does not solve this problem completely, as research has shown a decrease in the flexural properties of aged GRC compared to fresh GRC [86]–[88]. There exist different methods to reduce fibre embrittlement. Admixtures like fume, metakaolin, and nano-silica can be added to a concrete mixture. This permits pozzolanic reactions and transforms portlandite to C-S-H, thus decreasing the alkalinity of concrete [76], [80], [84]. The use of low alkalinity cement, like sulphoaluminate cement, could solve the problem because Portlandite ($Ca(OH)_2$) is not produced during the hydration process, but pH value in the pore solution still remains high at around pH 10. Matrix polymers like PVA, AC, and others can be used to densify the interface between fibres and cement. This technique reduces the diffusion of lime into fibres [84], [89].

Due to the thickness of samples and its standard application the tensile test is used to evaluate GRC properties. The level of proportionality (LOP) and the modulus of rupture (MOR) are used to describe the tensile strength of a given sample.

The LOP value describes the maximal linear elastic deformation of a given sample, which means that any applied load lower than LOP will not harm the material, no cracks will be seen on the surface, and the material will return to its present state. A sample in the elastic region, below LOP, can be described by Hooke's law whereby stress is proportional to the strain and the constant of proportionality is the Young's modulus. The LOP value demonstrates the maximum flexural strength of a matrix. After the load is increased above the LOP value, the material exhibits plastic behaviour, and it deforms irreversibly. At this moment, multiple cracks can appear on the GRC surface, but the material still has load-bearing properties. Increasing the load increases the elongation of the GRC sample due to the connection of fibres in the concrete matrix until a crack is too large and, at the fracture point, the fibres pull out or brake. The MOR value describes the destructive strength of a given sample [76], [82], [87]. The LOP and MOR values are influenced by many factors such as:

- fibre amount,
- fibre length,
- if sprayed GRC or premix is used,
- compaction.

In this research impact of the amount of glass fibre in a composition and of the length of fibre on the properties of sprayed GRC has been evaluated.

GRC samples were made using the spray method. The research was done to determine the fibre length and the resulting influence on GRC properties. Basic GRC components are cement CEM I 52,5R, fine quartz sand, superplasticiser, and acrylic polymer. The AR-glass fibre roving was used. The fibre length between 6 mm and 41 mm was used for different samples and cut during the spraying process. The fibre amount was varied in the range from 0 to 7 %. Sample flexural tests were performed according to standard BS EN 1170 [90]. Samples were cut from a test board in size 275 x 50 mm and tested with a 4-point bending test. The AR glass fibre was investigated using Phenom ProX Desktop SEM. Further analysis is done in **Paper III**.

The addition of AR glass fibre to concrete matrix has a significant impact on the ultimate load-bearing capacity of a given sample. As shown in Fig. 7, if 41 mm long fibres are used, the MOR values increase with the increase of fibre amount in the concrete, meanwhile the LOP values do not change significantly. The LOP values are more affected by the strength of the concrete matrix than by fibre addition [76].

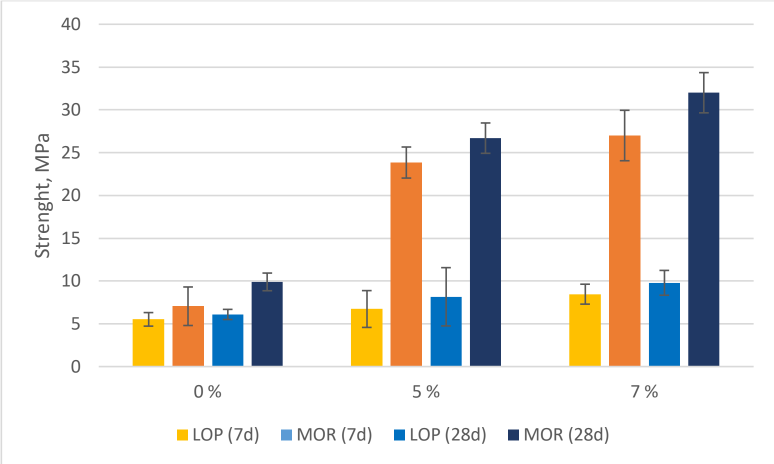


Fig. 7. Influence of fibre amount on LOP/MOR values [85].

Samples without fibres have very low plasticity and a low brittle fracture point. When the amount of fibres is increased from 0 % to 5 % and further to 7 %, the elasticity of the material increases. By increasing the fibre amount from 5 % to 7 %, the load-bearing capacity of the material increases, but as the fibre length has not changed the deflection of the material does not change.

In further experiments, the fibre length was changed from 6 mm to 41 mm, while the fibre amount remained fixed at 5 %. Figure 8 shows the LOP/MOR dependence on fibre length. The fibre length does not considerably change the LOP values. As we concluded before, the LOP values are more influenced by the properties of the cement matrix. The MOR values increase as the fibre length is increased from 6 mm to 41 mm.

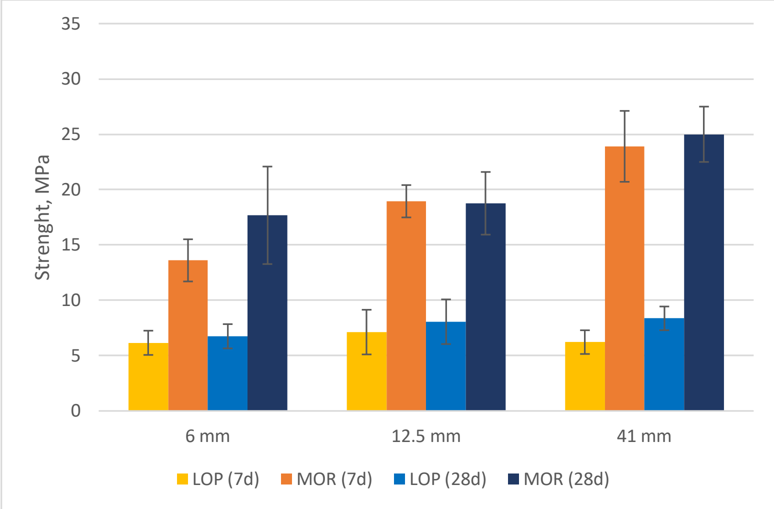


Fig. 8. Influence of fibre length on LOP/MOR values [85].

As shown in Fig. 9, by adding longer fibres, the plastic behaviour of the material is improved. H. Kasagani et al. have also noted that samples with longer fibres have higher deformation capacity compared to samples with shorter fibres [91]. Longer fibres have a higher chance to deform in the bundle structure improving post crack deformation of GRC. When analysing collapse of samples it can be seen that when longer fibres are used, the deflection stays constant for higher ΔForce. The filaments slowly pull out of the bundle instead of fracturing with the applied load and the crack size (deflection) does not change.

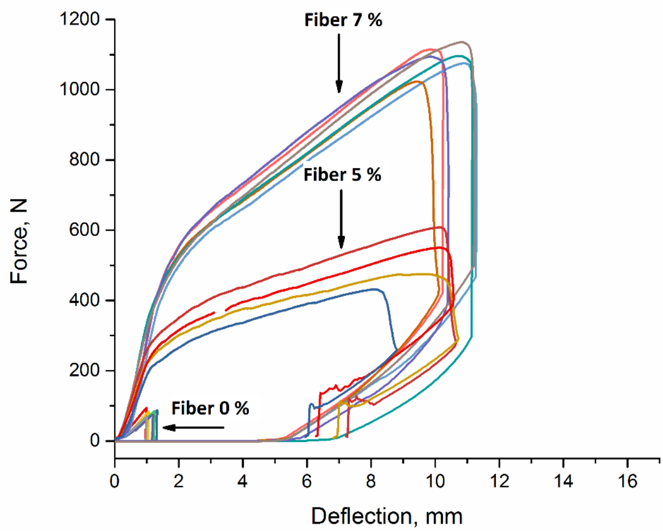


Fig. 9. Influence of fibre amount on deflection [85].

3.1.1. GRC properties with added crystalline admixtures

For **Paper VII** GRC samples were made using the same ingredients as before using spraying method. An acrylic polymer was added to the GRC mixture to produce polymer GRC (PGRC) samples. AR glass fibre constituted 5 % of the mortar for all of the samples.

Crystalline admixtures were used in this research to achieve GRC and PGRC self-healing after a crack appears on the concrete surface. In **Publication VII,** different crystalline admixtures from different suppliers were analysed by using FTIR and XRD.

Difference between flexural strength values depending on the GRC type and the admixture are presented in Fig. 10. The MOR values of the GRC samples are increased by adding admixtures, but the LOP values are not influenced and stay at the same level as of the reference sample. The PGRC samples have higher LOP and MOR values compared to the GRC samples. That can be explained by polymer addition which makes the brittle concrete more flexible and receptive to deformations.

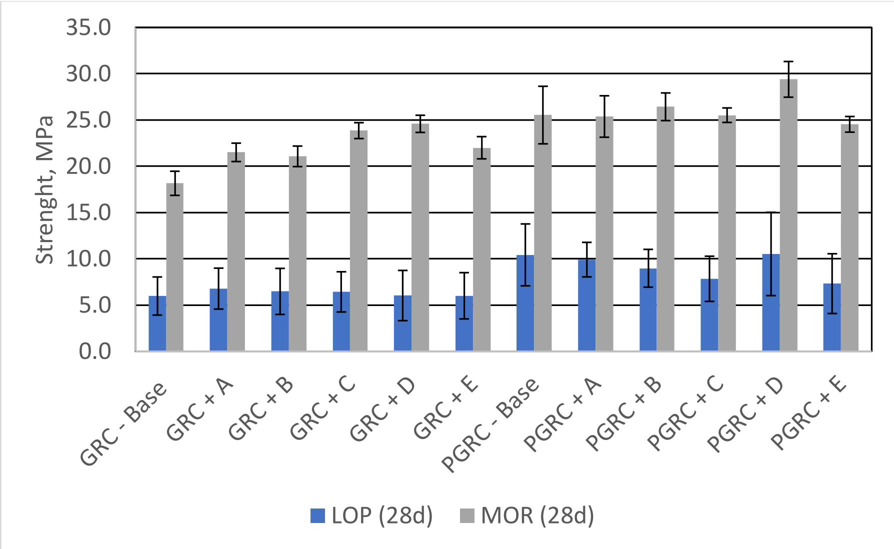


Fig. 10. Flexural strength depending on the GRC type and admixture [92].

The GRC and PGRC sample volumetric weight, water absorption (Wm%), and open porosity (Wvol%) are compared in Fig. 11. The PGRC samples have a lower volumetric weight compared to the GRC samples, but the open porosity and the water absorption are at the same level regardless of the type of reinforced concrete. Difference between the GRC and PGRC volumetric weights is about 5 %, so we can conclude that the added polymer has made the concrete structure lighter. Still, it has not reduced open porosity and water absorption.

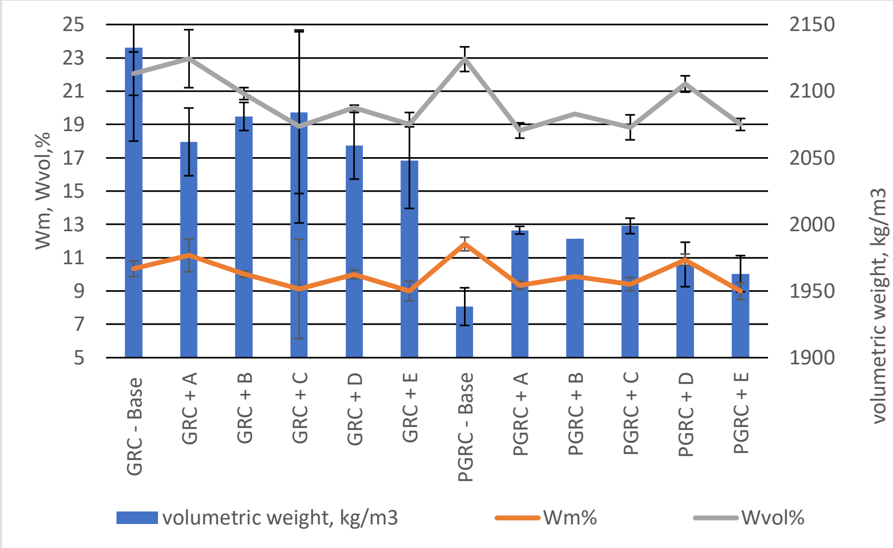


Fig. 11. The volumetric weight, water absorption (Wm%) and open porosity (Wvol%) of the GRC and PGRC samples with and without crystalline admixtures [92].

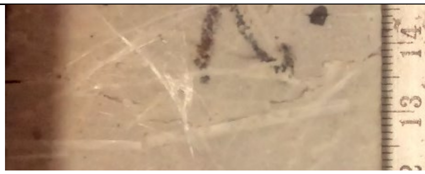
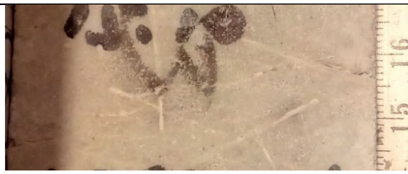
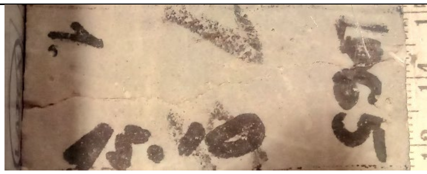
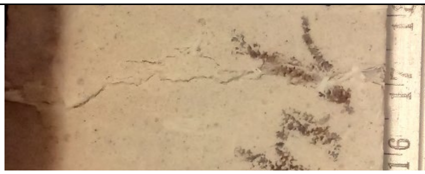
3.1.2. GRC and PGRC self-healing with and without crystalline admixture

In the research described in **Paper VII** 28-day old GRC and PGRC samples were pre-cracked and entirely immersed in water. After 14 weeks, the sample self-healing was evaluated visually, and the healed crack size measured with a microscope. It can be seen in **Paper VII** table 1 that the PGRC samples have better self-healing results than the GRC samples. Maximal healed crack size is from 134.7±0.06 to 308.7±0.06 µm. Other researchers have reported similar healed crack sizes [14], [17], [23].

The samples with the most promising admixtures were tested to evaluate the self-healing dynamic during an 8-week period. Results are shown in Table 5. Comparing the results after the self-healing of GRC and PGRC samples it can be concluded that the PGRC samples with and without crystalline admixtures start to heal after a more extended time period than the GRC samples because a crystalline admixture and an acrylic polymer are added during the mixing process and the crystalline admixture is "encapsulated" with a thin layer of acrylic polymer. Due to these differences, the average measured healing rate differs from 7±0.06 to 19±0.06 µm per week.

Table 5

Analysis of GRC and PGRC Sample Delf-healing Dynamic (Sample Width 4 cm) [92]

Sample type	**Start of the test**	**After 8 weeks**	*Evaluation of crack healing*	*Average healing rate in a week, μm*
GRC - Base			±	*16*
GRC + A			±	*12*
GRC + C			±	*7*
PGRC - Base			-	*17*
PGRC + A			-	*15*
PGRC + C			-	*19*

In Table 6, SEM images are presented. Most of the crystals formed during the healing process are $CaCO_3$ and CSH gel. $CaCO_3$ crystals are brittle, so the healed crack can easily reopen, but the CSH gel which is found in healed cracks is one of the basic concrete constituents that determinate its strength and durability.

Table 6

SEM Images of GRC and PGRC Samples after Self-healing [92]

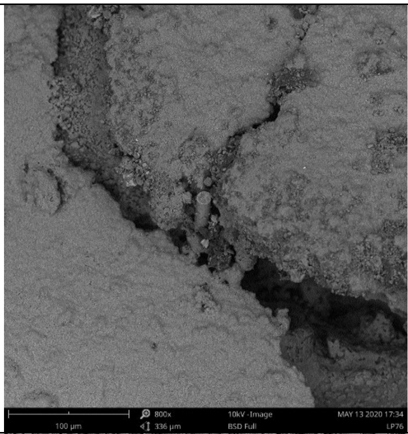
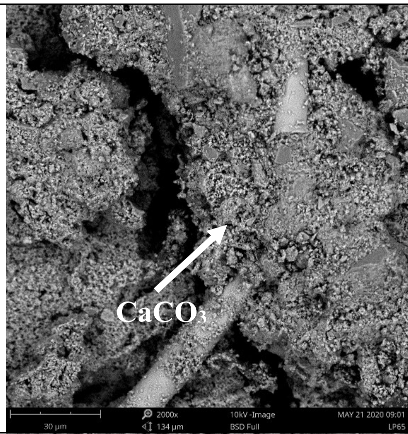
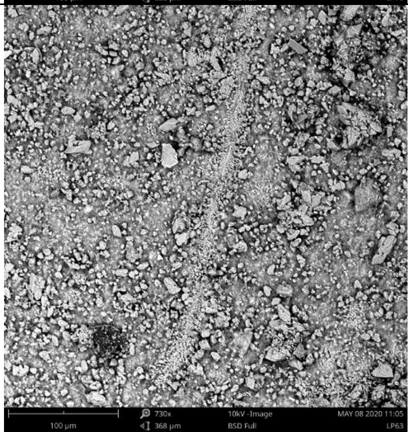
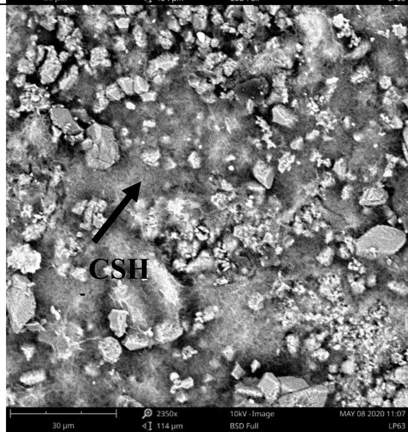
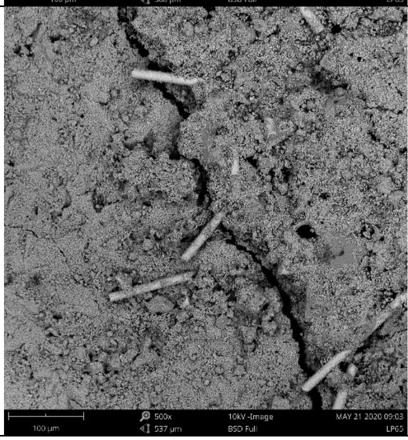
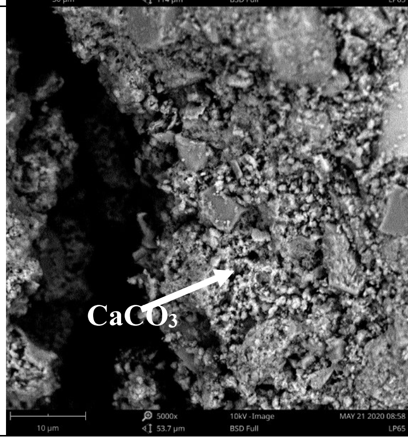
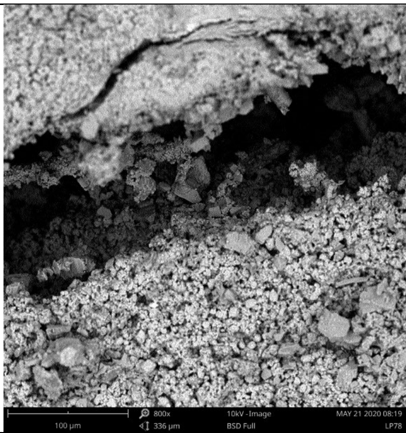
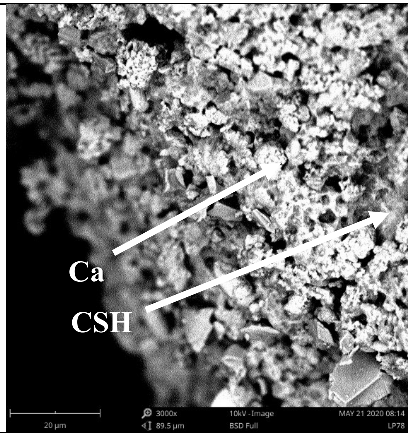
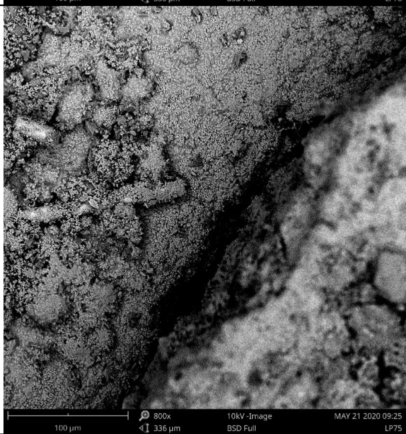
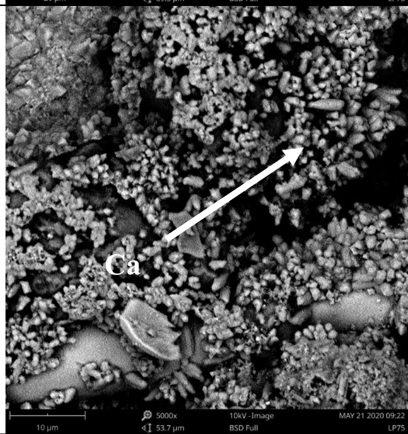
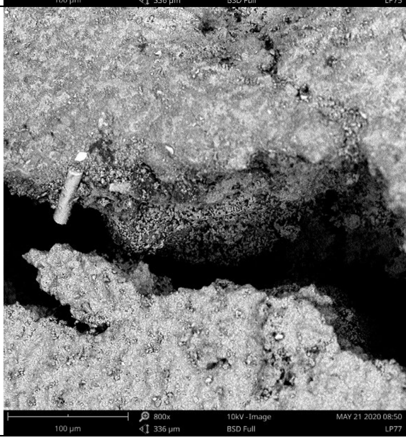
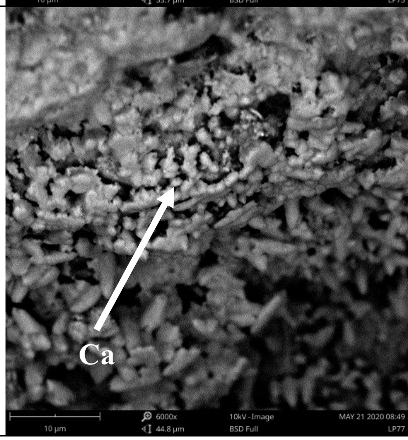
Sample type		
GRC - Base		
GRC + A		
GRC + C		

Table 6 continued

Sample type		
PGRC – Base		
PGRC + A		
PGRC + C		

Conclusions

1. The formation of cracks in solid bricks during drying is reduced by 30 % if the surfactant Triton X-100 is added close to the critical micelle concentration (CMC), as the surfactant helps water molecules to move easier from the brick centre to surface.
2. The addition of glass fibre waste (size ranges from 20 to 2 µm) to the clay mass shows an increase in compressive strength by 27 % and an increase in density by 4 % with increasing amount of glass fibre in the mixture.
3. By increasing sand size particle amount in the sample mix, an average crack area of the dried sample area decreases by 75 %. Increasing the amount of sand-sized particles proportionally reduces the amount of clay-sized particles that absorb water molecules, thus reducing the stresses caused during drying and reducing the amount of cracks.
4. After evaluation of the 8-week crack healing dynamics, the GRC samples with and without crystalline admixture have partially healed but the PGRC samples show poor signs of self-healing. It can be concluded that in the PGRC samples the start of the healing is delayed because a crystalline admixture and an acrylic polymer are added during the mixing. The crystalline admixture is "encapsulated" in a thin film of the acrylic polymer and does not have the water needed for crystal growth.
5. The main self-healing products of GRC and PGRC with and without crystalline admixtures after 8 weeks are $CaCO_3$ and C-S-H gel.
6. Crack growth using commercially available crystalline admixtures with bond inherent bond oscillations of C-H, Si-O, and $-SO_4^{2-}$ shows the best crack self-healing results. These elements indicate the presence of gypsum and organic matter in crystalline additives; as well it participates in the formation of C-S-H gel, which primarily provides the mechanical properties of concrete.
7. For ceramic materials the most effective method for reducing cracks compared to other methods used in the presented research is the optimization of the grading in the clay mix, i.e., increasing the amount of sand size particles. For cement-based facade materials the best self-healing results were achieved with the crystalline admixture that contains C-H, Si-O and $-SO_4^{2-}$ chemical compounds.

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APPENDICES

Full texts of publications

Paper I

S. Guzlēna, G. Šakale, S. Čertoks. Clayey Material Analysis for Assessment to be Used in Ceramic Building Materials. *Procedia Eng.*, **2017**, Vol.172, pp 333.-337. https://doi:10.1016/j.proeng.2017.02.031

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Modern Building Materials, Structures and Techniques, MBMST 2016

Clayey material analysis for assessment to be used in ceramic building materials

S.Guzlēna[a], G.Šakale[a], S.Čertoks[b]*

[a]*Institute of Technical Physics, Facultiy of Material science and Applied Chemistry, Riga Technical University. P.Valdena 7, Riga, Latvia*
[b]*LODE SIA, Lode street 1, Priekuli country, Latvia*

Abstract

Ceramic building materials, like bricks and blocks, are a modern choice. However, the production of ceramic materials is complex process, therefore, principal element, clay, research is very important. Clay from Latvian biggest quarry was used in this study. Grain size distribution was determinate using the pipette method. Quarry soil is available in various grain size compositions from clay and clay alerolites to sandstone. Mineralogical composition was determined by XRD. Mixing very plastic clay with sand may be very important to obtain optimal proportion of clay, silt and sand size particles for qualitative ceramic building material manufacturing.

Peer-review under responsibility of the organizing committee of MBMST 2016

*Keywords:*brick; clay; grain size distribution; XRD; pipette methode.

1. Introduction

Ceramic building materials are modern and environmentally-friendly solution for buildings. Materials, such as bricks and blocks, have high strength, durability and the ability to control humidity, creating a healthy indoor microclimate. The main raw material for the creation of ceramic building materials is all period sediments occurring in clayey soils. In Latvia these are the Devonian and Quaternary, and to a lesser extent Triassic and Jurassic clayey soils. The most important quarry in Latvia consists of Devonian clayey terrain. Field covers a whole different set of

* Corresponding author. Tel.: +371 29923504.
*E-mail address:*Sandra.Guzlena@gmail.com

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soil - from clay and clay alerolites to sandstone, but mostly they are different shades of red, no carbonate clayey aleirolites[1,2].

The grain size distribution in quarry varies within a wide range. It is very important to determinate appropriate grain size distribution, because it influences considerably the behavior of ceramic bodies during the technological process, like behavior of the material during the shaping and drying processes. It has effect on the microstructure and the mechanical properties of fired materials as well [3]. Raw materials with high percentages of clay size particles experience difficulties in the drying process as consequence the high moisture required in the molding one. However, the firing temperature can be decreased due to the fact that the vitrification process, which initiates on the specimen surface, can occur more easily [4].

Based on German experience, suggested grain size distribution limits for solid bricks, perforated bricks and tiles are illustrated in triangular diagram (Fig. 1). It includes three main clayey soil grain distribution – clay (<2μm), silt (2-20μm) and sand (>20μm)[5]. Diagram is divided into four parts: unsuitable – all type of products liable to shrinkage during firing; roofing tiles; perforated bricks; solid bricks only.

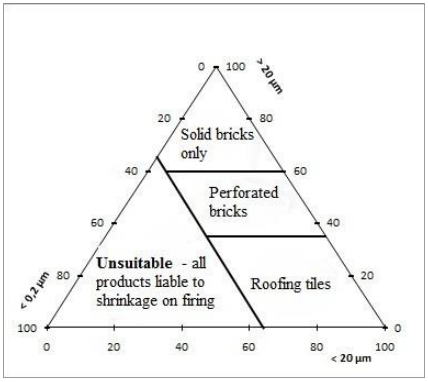

Fig. 1. Suggested grain size distribution for bricks an roofing tiles [5].

As the goal is to have solid bricks it is important to investigate granulometry of available quarry clay and to made proportion of soil grain distribution that confirm "Solid bricks only" part. It consists0 – 35% of clay (<2μm), 65 – 100 % of silt (2-20μm) and 60-100% of sand (>20μm). Very small percentage of coarser fraction up to 10 mm can be included to improve mixing and aesthetic reasons. Most common used is grog (pre-fired clay). Particles in the size range 50μm to 1, 2 mm is used as filler fraction, usually sand-size quartz. High sand-size particle content may be used also for aesthetic reason to obtain attractive texture, ore to balance very plastic clay. Silt-size and clay-size particles is used as plastic fraction for good forming properties [5]. However general requirements for brick making materials are:

- Plasticity of brick making mass: Mass should be plastic enough to extrude, shape or press it into a mould;
- Shrinkage and cracking must be limited: Formed materials shrinkage and cracking must be controlled during drying and firing process;
- Sufficient dry material strength: Dry material strength must be enough to during technological process quarry firing it will not be damaged and will have adequate strength after firing [5].

2. Materials and methods

Four basic types of clayey soil from quarry were used – Grey, Red, Composite and Sand. Composite clay is colorful mottled, no carbonate clayey aleirolites and in them occurring like intermediate state - light gray, refractory clays.

The mineralogical analysis is carried out by X-ray diffraction (XRD) using Analytical X'PertPro difractometer whit Cu Kα1 radiation, scanned from 2° 2θ to 29° 2θ using 0,1º 2θ scanning step. Four textured clay powder samples was made - untreated, heat treated (400ºC and 550ºC) and ethylene glycol treated.

Particle size distribution was determined by granulometry analysis done by the pipette method. Raw material was dried at 105ºC sieved till <2mmfraction. Oxidation of organic matter was done by hydrogen peroxide (H_2O_2). Samples were dipped in distilled water whit dispersant ((Na_2PO_3))+Na_2CO_3 for 24 h. The aim of pretreatment is to promote dispersion of the primary particles, and to get more accurate clay size particle amount in the sample. Suspension was wet sieved on 63 µm sieve. Fraction >63 µm that left on sieve was dried at 105ºC and dry sieved. Fraction that passed 63 µm sieve was analyzed by sedimentation method taking samples at appropriate intervals [6].

3. Results and discussions

3.1. Grain size of clays

The particle size range in clayey soils should include an even mixture of clay, silt and sand to use in producing building ceramic. But, as shown in Fig. 2 after grain size distribution analysis it can be concluded, in grey clay dominates clay fraction (<2 µm) and has least sand fraction (>20µm). Uniform distribution over the fractions has red clay. Composite clay due to its aleirolites inclusions has high contents of coarse silt and sand. Sand has shown that 95% of it has sand fraction but only less than 1% clay fraction.

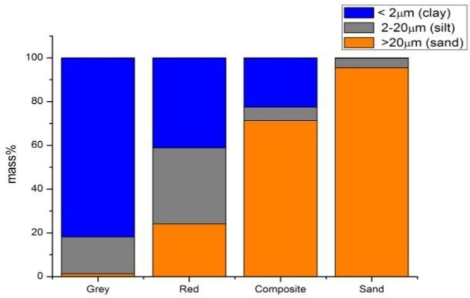

Fig. 2. Grain size distribution on grey, red and composite clay and sand soil.

After determination each fraction in every raw clay sample was inserted in diagram which was described in introduction (fig. 3). This diagram is separated in four parts as it was shown in fig.1: solid bricks only, perforated bricks, roofing tiles, and unsuitable-all products liable to shrinkage on firing.

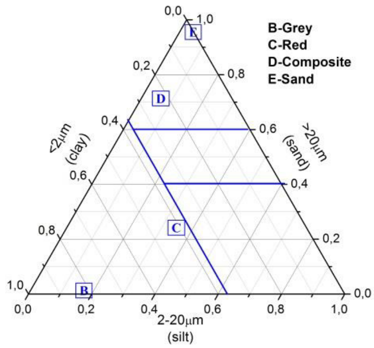

Fig. 3. Grey (B), red (C), composite (D) and sand (E) location in diagram.

Grey (B) and red (C) clay as only components in mix is unsuitable for ceramic building materials as it shown in diagram. Lack of sand sized particles in grey and red clay, cause considerable shrinkage during drying and firing process. Particles in sand size range function are to control shrinkage, lamination and to provide gas drainage paths during firing. Using only sand for solid brick manufacturing is possible as shown in diagram. But its mechanic properties would not be satisfactory it can weaken the brick. Cracks forms if the cooling process is inadequately executed, due to quartz volume decrease of about 0,8% at 573 ^{o}C which is caused by its crystallographic modification of the quartz[7]. Abrasiveness of sand show destructive effect in primary production line, especially on the mechanic parts of extruder. More appropriate for ceramic building material production is composite clay composition due to its balance between all fractions[5,8].

3.2. Mineralogy of clays

Results of mineralogical analyses are shown in Fig. 4 for clay samples. Illite, kaolinite and quartz are detected in the X-ray diffraction pattern of the studied samples.

Illite is essentially a group name for non-expanding, clay-sized, dioctahedral minerals. Members of the illite group are characterized by intense d=~10Å (~ 8.1 2θ) and d=~5Å (~17.42θ) peaks that remain unaltered by ethylene glycol and heating to 400^{o}C and 550^{o}C [9]. No evidence of illite polytypes and mixed layer clays was noticed. Illite is mineral that impart materials good plasticity, but tend to vitrify at low temperatures and does it over a narrow temperature range. It means that kiln temperature has to be tightly controlled. Too low temperature will result in the product with weak mechanical properties due to insufficient sintering (underfiring). On the opposite, too high temperature will result in deformed final product as fusion accelerates (overfiring) [5].

The kaolinite group includes the dioctahedral minerals kaolinite, dickite, nacrite, and halloysite, and the trioctahedral minerals antigorite, chamosite, chrysotile, and cronstedite. The primary structural unit of this group is a layer composed of one octahedral sheet condensed with one tetrahedral sheet. Kaolinite diffraction peaks can be noticed at d=~7.18Å (12.1 2θ). Peaks remain unaltered by ethylene glycol, but disappear by heating to 400^{o}C and 550 ^{o}C, because they become amorphous to x-rays [9]. Well ordered kaolinites tend to be relatively coarse grained, non plastic and to have a high melting point. This makes them useful as refractory raw material but less suitable for brick making [5].

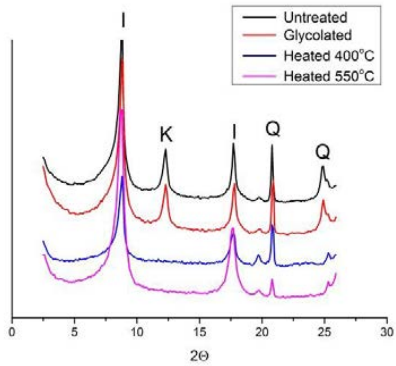

Fig. 4. XRD pattern for untreated, glycolated and heat treated clay sample.

4. Conclusion

Four basic types of clayey soil from Latvia biggest quarry were used – grey, red, composite and sand.Using raw material from quarry without additives considering only to grain size distribution requirements most appropriate is composite claydue to its balance between all fractions.To use grey and red clay they must be stabilized with sand size particles to avoid unnecessary shrinkage during firing. Sand found on quarry would be suitable stabilizing additive for plastic clay. Illite and Kaolinite are the only clay minerals found in the samples. Suitable brick making material should have suitable proportion of illite, kaolinite and quartz. Illite could provide plasticity, kaolinite promotes fired hardness and sand grains act as a stabilizer and filler material.

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Paper II

S. Guzlena, G. Sakale, S. Certoks, L. Grase. Sand size particle amount influence on the full brick quality and technical properties. *Constr Build Mater*. **2019**, Vol.220, pp 102-109. https://doi.org/10.1016/j.conbuildmat.2019.05.170

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Sand size particle amount influence on the full brick quality and technical properties

S. Guzlena [a,*], G. Sakale [a], S. Certoks [b], L. Grase [c]

[a] Institute of Technical Physics, Faculty of Material Science and Applied Chemistry, Riga Technical University, P.Valdena 7, Riga, Latvia
[b] LODE SIA, Lode Street 1, Priekuli Country, Latvia
[c] Institute of Silicate Materials, Faculty of Material Science and Applied Chemistry, Riga Technical University, P.Valdena 7, Riga, Latvia

HIGHLIGHTS

- Sand addition reduces a crack formation during the drying process.
- Phase change of quartz creates stress and cause cracking during firing process.
- Proper clay mass and proper extruder must be used to get the good quality bricks.

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ABSTRACT

In this study was used three different clay types and sand from the local quarry in Latvia. Clay mass mixes were made by adding sand in each mix and remaining the same amount of clay proportion. Sand size particle amount was increased from 33% to 59%. Samples were extruded with laboratory extruded as full brick samples. After formation samples were dried at 105 °C and fired at 1000 °C for 1 h. Water absorption, shrinkage, density, compressive strength were measured after the firing process. The crack amount was determinate by the digital camera after drying process and after firing process. Fired sample cracks start/end points were analysed with SEM-EDX.

1. Introduction

In many quarries clay granulometry variates in the wide range from very plastic clay with high clay size particle amount till sandy clay with high silt and sand-size particle amount. It is very important to control and adjust it with additives for production, to minimize varieties in granulometric because that can affect product quality and properties after firing [1]. Quality control must be done by analyzing granulometric data, clay mass moisture content during forming, drying shrinkage, firing shrinkage, water absorption, and other properties. In production, a quality control process is important that ensures that a good product reaches the customer and meets his needs, like thermal conductivity, strength, home environment and visual aspect [2,3]. Quality of the brick must be controlled through all manufacturing process, starting with material testing, formed green brick, dried brick and fired brick. One of the quality tests is visual inspection – brick cracking. First cracks can be visually observed after drying. Drying rate must be set right to not cause too fast temperature increase. In the manufacturing process drying usually happens in the drying tunnel. Brick temperature is increased 30–80 °C to evaporate free water from bricks. A drying tunnel is split into 3 parts. In the first part of the tunnel, a brick air temperature is about 50 °C and relative humidity is around 80%. In the second part of the tunnel, drying air temperature is kept constant and relative humidity is starting to fall due to a brick moisture decrease and tunnel fans that suck out wet air from the drying zone. In the third part is a temperature of brick is around 80 °C and moisture content is around 3% [4].

Secondly, cracks can appear after firing. Fracture and cracking mostly are caused by volume changes during the firing process. Usually, it happens with SiO_2 which modifies from alpha to beta phase and changes its volume. During the firing process, water, and volatiles have been loosened. At temperature 1000 °C and higher starts vitrification of clay-size particles and fluxes. This process is also called sintering [5]. As Kadir et al. have noted firing

* Corresponding author.
E-mail address: sandra.guzlena@rtu.lv (S. Guzlena).

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process is essential for brick quality. They have changed heating rates during firing process form 0,7 till 10 °C min^{-1} and concluded that brick properties start to decrease if heat rete is 2 °C min^{-1} or more [6].

Orhan and Demir in their research have found that river sand addition to clay mix decrease fractures occurring during the production process. Creation of cracks is reduced due to clay mass plasticity decrease. In mixture 10–15% of clean-river sand was used. River sand is quite clean, and it contains fewer organic impurities comparing to hill sand or crushed rocks. It can be concluded that the addition of 15% of river sand had reduced the crack formation during the drying and firing procedures [5].

Azzi et al. [7] have concluded that high plasticity clays can be forced dried at 200 °C and 300 °C to reduce their plasticity. After drying clays are not able to fully absorb the amount of water what they could before force drying. These force dried clays can be used as additive to the mixture to decrease plasticity of clay mass and reduce shrinkage.

As Mcnally wrote in his work that suggested particle size distribution for full bricks is determined as follows: particles >20 µm should be 60% or more, particles 2–20 µm should <40% and particles <2 µm should be <35% [8].

In the literature are many investigations about clays in new and existing quarries and their granulometry, different technologic properties to be used for brick production [9–11], but are a very low paper amount in which are discussed clay ceramic material cracking, crack measurements cracking, detection, and elimination potential. One of the ways is to define the composition of clay mass with the basic materials, clay and sand, whose boundaries are possible to obtain a qualitative, free cracks product. Based on literature review, it was decided to test sand size particle amount increase influence on technical parameters and crack formation for clay bricks. Sand size particle amount was varied from 33% to 59%, by granulometric data.

2. Material and methods

From a quarry in Latvia was obtained sand and three types of clay – grey, red and composite clay. All ingredients were mixed changing sand ratio and water was added to obtain plastic clay mass. After mixing samples were extruded and its green size was 12.0 × 3.4 × 7.0 cm (length × high × width). Samples were dried at 105 °C. The firing process was done at 1000 °C for 1 h. Five different mixes were made with a sand-size particle amount increased from 33% to 59% due to granulometric data.

Clay mixture granulometry was determined using the pipette method. A raw clay mix sample was dried at 105 °C. The dried sample was crushed and sieved till <2 mm fraction. Hydrogen peroxide (H_2O_2) was used to oxidize organic matter. The sieved sample was immersed in distilled water and dispersion (Na_2PO_3) + Na_2CO_3 were added. The material passed in this solution for 24 h. Dispersing in this solution is carried out to obtain a more accurate amount of clay particles in the sample. The suspension was sieved using a 63 µm mesh and mimic method. Particles that did not pass through a 63 µm sieve were dried at 105 °C. Particles that were sieved through a 63 µm sieve were analysed by a sedimentation method by taking samples at appropriate time intervals [12].

Sample moisture content was determinate in two ways. First to control moisture content during mixing quick analysis by moisture analyser weights, KERN DAB 100-3, was made. After samples was formed clay mass was dried at 105 °C and measured weight changes before and after drying.

Sample drying shrinkage measurements was done controlling sample length, width and high before and after drying process.

Fired sample water absorption was determinate by immersing samples in water tank at room temperature for 24 h.

Cracks were analysed after firing and drying of samples. Bricks were sawed in 2–3 mm width pieces. Crack measurements and analysis were made by Software Motic Images Plus 2.0 and a digital camera (Moticam 2000).

Differential thermal analysis (DTA) and thermogravimetry (TG) was determinate using derivatograph "SETSYS Evolution TGA-DTA/TMA SETARAM". Dried powder samples were analysed. Derivatogramms were taken from 25 till 1100 °C with heating rate 10 °C/min.

The sample crack start/end point was analysed by SEM-EDX Hitachi S-900.

3. Results and discussion

3.1. Clay and sand analysis

As mentioned all three types of clay and sand were gained from the same quarry in Latvia. The granulometric data of all sample differ. Grey clay is very plastic with high clay size particle content as shown in Fig. 1. Red clay consists of evenly distributed clay, silt and sand-size particles, but composite clay mostly consists of sand size particles and in less manner silt and clay size particles. Sand which was used as an additive and individual clay sample granulometry and mineralogy are analysed in our previous work [13].

3.2. TG and DTA

To identify process during firing, differential thermal analysis (DTA) and thermogravimetry (TG) was made. Samples were heated till 1100 °C. All three types of clay were analysed.

During firing clays proceed many processes as volume and phase change and crystallization process (Fig. 2a). After samples are heated till 100–150 °C water evaporates from pores and voids and there can be seen this endothermic reaction. Next process happens between 200 and 600 °C when organic matter oxidises. Between 600 and 900 °C hydroxyl water evaporates from the sample. Evaporation rate is influenced by clay type, mix proportions and other factors [14].

General conclusions from DTA (Fig. 2b) analysis of clays from a quarry may be summarized and described below. The endothermic peak below 200 °C indicates that illite clay is used. It has been approved in our previous work by X-ray diffraction (XRD) analysis [13,14]. Illite clays provide the plasticity of the clay mass, therefore its increased amount can increase unwanted shrinkage and cause cracking during the drying process and the firing process. Shrinkage is related to absorbed water evaporations and weight loss (about 0.3–0.4%) [15,16]. The endothermic peak between 200 and 300 °C points at a dehydroxylation of iron or alumina oxyhydroxides. Organic material presence can be seen in wide exothermic reactions till 600 °C [11,14,15]. Clays with high organic material content must be fired slow and steady to ensure complete carbon oxidation [14].

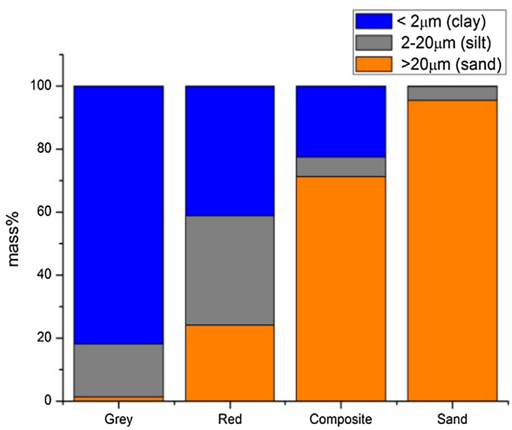


Fig. 1. The granulometric proportion for grey, red, composite clay and sand samples.

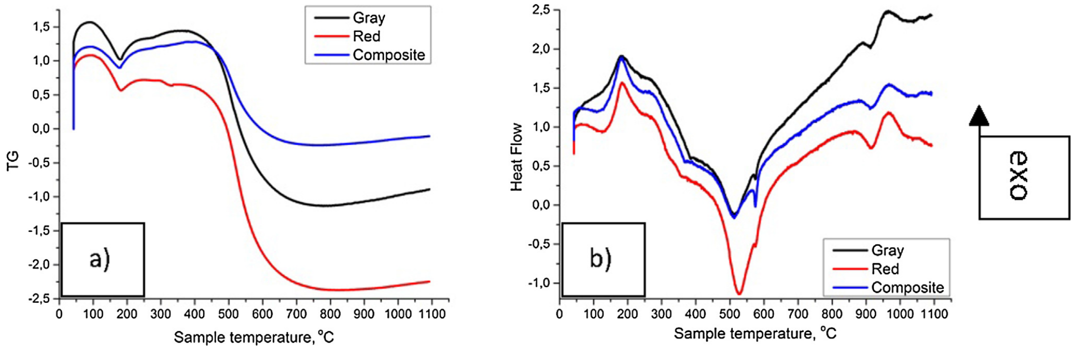


Fig.2. a) TG and b) DTA analysis of Grey, red and composite clay.

Kaolinite mineral presence in clays represents by an endothermic peak around 600 °C and exothermic peak around 975 °C. Once more illite mineral presence in clay approves by low-intensity peak around 500 °C and endothermic reaction at 900 °C. Presence of illite indicates that it is not refractory clays and vitrification will happen in a small diapason [14]. An endothermic peak at 572–575 °C is typical of quartz phase transformation [14,15]. Calcite decompose between 700 °C and 800 °C [17,18], but such isn't absorbed due to low carbonate content in quarry. Feldspar melting, and liquid silicate appearance causes an endothermic reaction around 800-1000C. But the exothermic reaction is due to spinel or mullite phase formation [15].

3.3. Brick sample granulometry

To discuss how the increasing amount of sand-size particles have affected sample properties is important to determinate its granulometry. As [8] suggests particle size distribution range for brick and tile clays are illustrated in Fig. 3a). For brick manufacturing, sand-size particles (>20 μm) should be minimum 60%, silt-size particles (2–20 μm) should be maximum 40% and clay-size particles (<2μm) should not >35%.

Five clay mixes were made. In diagram Fig. 3(b) is illustrated by sample granulometric data. Most appropriate for solid bricks was Mix_5, where sand size particle amount was increased till 59%, silt-size particle amount decrease till 9% and clay-size particle amount was 32%, but it is only the boundary of tiled and faced bricks. So due to this diagram sand size particle amount can be even higher to achieve particle size distribution range for solid brick.

In Fig. 3(c) are shown all mix granulometric composition. As shown, the sand-size particle amount was increased by each mix, but the silt and clay amount were decreased. During drying process quartz sand reduce drying shrinkage and decreases risk of crack formation [1]. During the firing, the sand ensures the formation of gaseous channels, lamination and shrinkage control, which also contributes to the reduction of crack appearance. At the end of firing quartz make a glassy phase which sinters the clay particles and provides dense structure and improved mechanical strength. Using a large amount of sand must be careful during the cooling process, quartz volume decreases at 573 °C and cracks can appear due to volume change. The decrease of about 0,8% is caused by SiO_2 alpha to beta phase transformation [1,19].

The properties of silts are more sensitive to the amount of water in a clayey material than are those of coarse-grained clayey soils. With sufficient moisture content, capillary forces can reduce friction between particles and easily change the state of silt from solid to liquid [20].

3.4. Dried sample analysis

Clay was taken from a local quarry and used for brick sample preparation. Therefore, in Table 1 is shown that average sample moisture content during forming changes from 14,4 to 20,8%. But as shown in third column average drying shrinkage do not depend only on sample humidity. Average drying shrinkage correlates with a sample clay-size particle amount. As known, clay has a layered structure. Between these layers are water molecules, during drying water molecules evaporate and layers move closer to each other which results in sample shrinkage [9].

3.5. Fired sample analysis

Water absorption of fired clay brick samples as shown in Table 2 is highest of the samples where the sand-size particle amount is the lowest. Comparing to Temga et al. work our water absorption is similar, around 14% [1]. But it is twice bigger than regulated water absorption in Latvia which is <7%. Mix 3, Mix 4 and Mix 5 water absorption is lower because sand size particles are >50% in the mixture. Sand size particle water absorption is less than clay size and silt size particle water absorption. Since the durability of bricks is closely related to water absorption, this parameter can be used to characterize the quality of bricks [21]. Our sample density is in the same range that have noted Temga et al. in their work [1].

In Table 3 is shown sample compressive strength. From results we can conclude that if higher is sand size particle amount in clay mix, higher compressive strength can be achieved.

3.6. Crack analysis

In Table 4 are shown that by increasing sand size particle amount in the sample mix, an average crack area of the dried sample area decreases. That can be explained as before by decreasing clay-size particles which absorb water molecules and increasing sand-size particles, sample drying-induced stresses decrease and cracks do not form [9,22]. An important influence on cracking during the drying process has the drying rate. If the drying process is fast, brick drying tends to be more irregular, contributing to the presence of more flaws, which led to the generation of more fractures. Therefore, drying must be done evenly [23]. In the laboratory, drying happens in 105 °C. In a manufacturing process, it is a

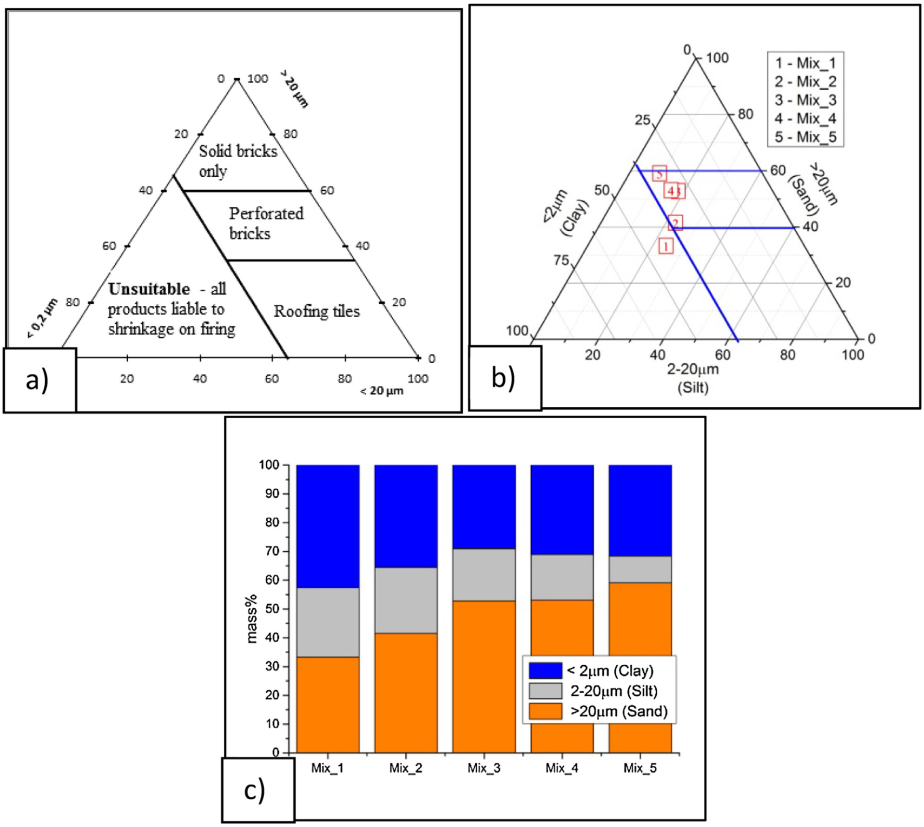


Fig.3. a) Suggested particle size range for brick and tile clays [8]; b) Mix location in a diagram; c) Mix granulometric composition.

Table 1
Brick sample moisture content during forming, drying shrinkage and granulometry.

Mix number	Average sample moisture content during forming	Average drying shrinkage	Granulometry, % (>20 μm/<20 μm/<2 μm)
Mix_3	14,4%	5,0%	53/18/29
Mix_5	18,4%	5,2%	59/9/32
Mix_4	15,4%	6,6%	53/16/31
Mix_1	18,9%	6,7%	33/24/43
Mix_2	20,8%	7,5%	42/23/35

Table 2
Fired brick sample water absorption, weight loss after heating and granulometry.

Mix number	Average water absorption of a fired sample (1000°C), %	Average sample density, g/cm^3	Average heating weight loss, %	Granulometry (>20 μm/<20 μm/<2 μm)
Mix_3	11,86	1,79	6,27	53/18/29
Mix_4	13,41	1,71	6,58	59/9/32
Mix_5	13,84	1,60	2,10	53/16/31
Mix_2	14,08	1,82	2,39	33/24/43
Mix_1	14,72	1,79	4,05	42/23/35

Table 3
Fired brick sample compressive strength.

Mix number	Compressive strength, MPa	Granulometry (>20 μm/<20 μm/<2 μm)
Mix_2	16,11	33/24/43
Mix_3	14,96	53/18/29
Mix_1	14,70	42/23/35
Mix_4	12,50	59/9/32
Mix_5	12,31	53/16/31

three-stage process where the brick is evenly heated, and humidity decreased. The brick temperature at the start of drying is around 30 °C and moisture content 25%, but at the end of drying brick temperature is 80 °C and moisture content 3% [4].

The fired sample average crack area does not show any relation to granulometry or an average crack area of dried samples. But an average crack area can be related to an average heating weight loss if comparing Tables 2 and 4. Samples with the lower average crack area have lower heating weight loss. It can be explained by to a rapid temperature increase in two sensitive zones. In the first temperature zone of 100–150 °C, where water evaporates from pores. The second zone from 500 to 900 °C, where hydroxyl water evaporates, and quartz transforms from alfa to beta phase around 573 °C.

An analysis of the number of cracks depending on the crack length was performed in Fig. 4. Cracks with a length of 1–5 mm dominate in the samples. MIX1 is dominated by cracks with a length of 5 mm–1 cm. MIX2 have cracks with length 1 mm–2 cm.

MIX3 have large number of cracks with length 1 mm–1 cm. MIX4 have cracks 1 mm–2 cm. MIX5 have mostly cracks with length 1 mm–1 cm.

Brick sample visual crack measurements are shown in Table 5. Similar measurement technique has used Chaduvula et al. in their work following parameters were measured: average crack opening width, spacing between cracks, a total cracked area of expansive clay samples with/without fibre reinforcement [24]. Samples with higher average crack area, like Mix_1 have higher amount of cracks longer than 5 mm.

Differences between, dried and fired samples are significant. In every mix that was made, crack amount increases after the firing process. After firing highest crack amount is for compositions were sand dominate, Mix_3 and Mix_4, excluding composition Mix_5, which by clay material granulometric triangular diagram [13] is the closest to ideal solid brick composition. But composition Mix_5 shows unfortunately lowest compressive strength result. Optimum compositions by taking into account amount of cracks and compressive strength data are Mix_1 and Mix_2.

Table 4
Dried and fired sample average crack area due to sample granulometry.

Mix number	The average crack area of the dried sample	The average crack area of the fired sample	Granulometry (>20 μm/<20 μm/<2 μm)
Mix_1	1,00%	1,34%	42/23/35
Mix_2	0,16%	0,86%	33/24/43
Mix_3	0,37%	4,65%	53/18/29
Mix_4	0,37%	3,36%	53/16/31
Mix_5	0,26%	0,80%	59/9/32

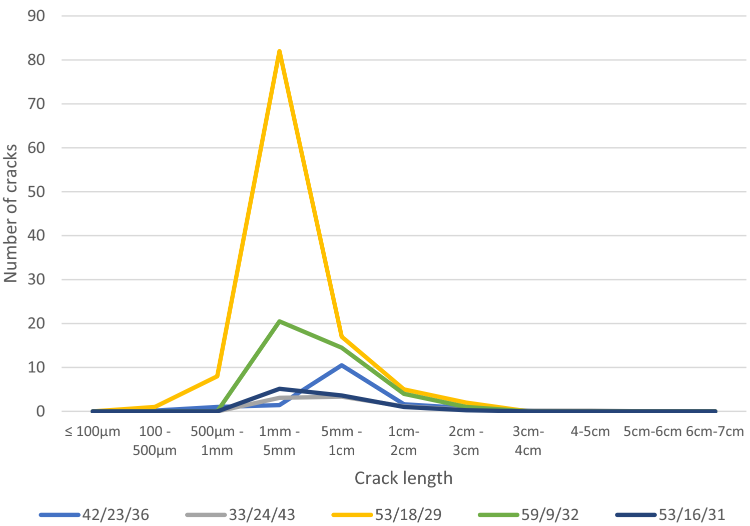


Fig.4. Crack length and number of cracks difference between material mixes.

Round shaped racks that appear in almost every sample at centre comes from extruder auger. This type of crack is a combination of flow lamination and rotational movement of clay mass. Also, hallow space created by an auger hub in the centre of the emerging clay mass which has not entirely re-joined as a result of mass characteristics or geometry of pressure head and die [25]. These hallow spaces in a sample which do not occur in dried samples are visible in fired samples. There are two reasons for this type of cracks. First quartz nature to transform from alfa to beta phase around 573 °C, the second – firing rate [26]. Second, due to Arsenovic et al. research [9], firing temperature must be increased slowly (2–5 °C/min) during the first part of the process. Slow warming should be carried out till 600 °C in order to minimize the possibility of cracks occurring during quartz phase transformation from alpha to beta phase at 573 °C. At the second part heating rate can be increased to 5–10 °C/min. The firing rate that is used in our research is 5 °C/min from room temperature to 1000 °C. In this case, probably the warming rate in the first part is too rapid and cause cracking in samples. As Ukwatta et al. have concluded heating rate influences also other properties as firing shrinkage, compressive strength and water absorption. As heating rate increase compressive strength of brick decreases as a result of low vitrification time which bonds clay particles leaving them more porous and brittle [1,27].

After firing samples were analysed using SEM (see Table 6). Almost in every sample were found sand grains in size 150 μm and more. Area around grains are cracked, due to quartz phase change. In many works [26,28,29] cracks around quartz grains have been seen. Allegretta et al. in their work [29] also have encountered problems with cracking around quartz grains and by increasing the firing temperature from 750 °C to 1000 °C detachment zone and cracks size increases.

EDAX analyses of all samples were similar. In Table 7 is shown typical results from all samples. Brick samples generally consist from Si which in the sample is 46% from a detected area and O is 27% from a detected area. And other elements like Al, K, Fe, and Mg are the main elements in illitic clays which in a smaller amount, respective 14%, 5%, 4% and 2%, is found on a detected sample area.

Table 5
Dried and fired sample crack measurements.

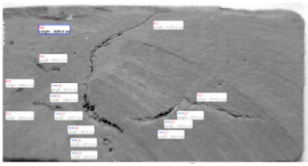
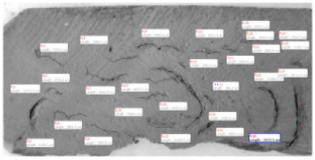
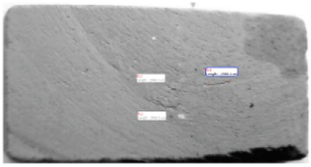
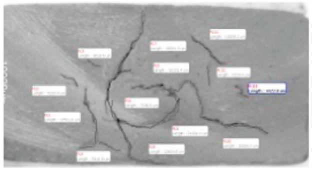
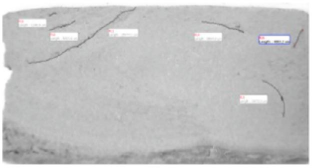
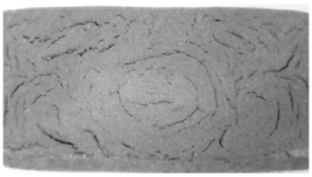
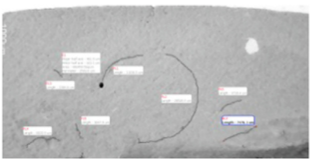
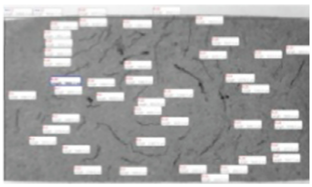
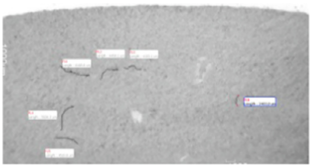
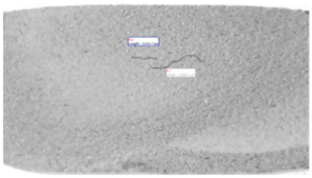
Mix number	Dried sample	Fired sample
Mix_1		
Mix_2		
Mix_3		
Mix_4		
Mix_5		

Table 6
SEM images of fired samples.

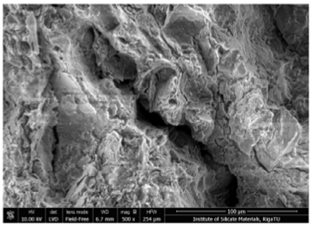
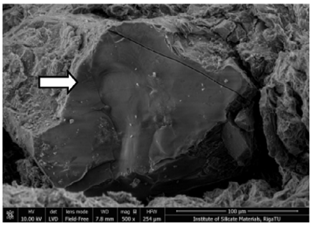
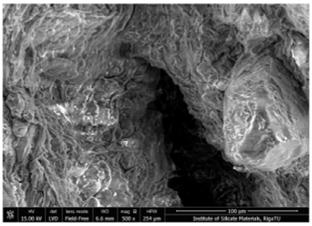
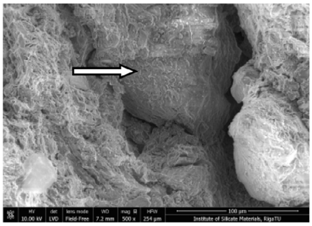
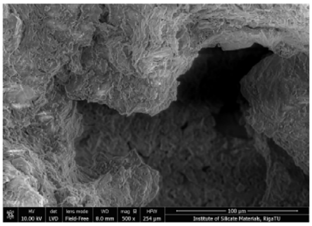
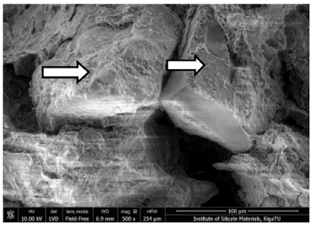
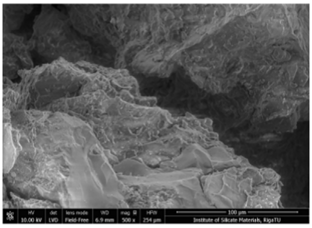
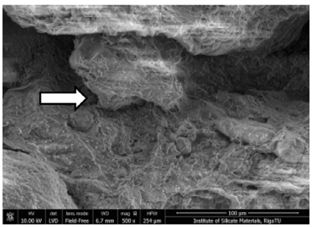
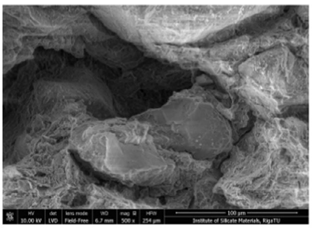
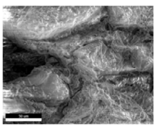
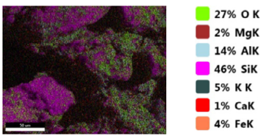
Mix number	Images of crack and big sand grains	Images of crack
Mix_1	Sand grains were not observed	
Mix_2		
Mix_3		
Mix_4		
Mix_5		

Table 7
EDX images of a fired Mix_5 sample.

Mix_5		

4. Conclusion

In this work brick mass granulometry was changed by increasing sand size particles (>20 μm) and decreasing silt-size (20–2 μm) and clay-size (<20 μm) particle amount. Samples were analysed after firing and drying. Sand addition reduces a crack formation during the drying process, but mainly increase crack amount after firing Sand, which consists of quartz change phase from alpha to beta around 573 °C. This phase change creates unnecessary stresses in the clay during the firing process, therefore temperature increase during the firing process must be slow to avoid cracking. In our work used heating rate 5 °C/min was too rapid. The second reason for major cracking was large sand grains observed in SEM. Round shaped lamination observed before and after firing has created an auger hub in the centre of an emerging clay mass which has not entirely re-joined because of mass characteristics or geometry of pressure head and die. These laminations or cracking increases after firing. So very important is not only proper clay mass but also proper extruder to get the best results during material testing and later production.

Declaration of Competing Interest

None.

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Paper III

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ALKALI RESISTANT (AR) GLASS FIBRE INFLUENCE ON GLASS FIBRE REINFORCED CONCRETE (GRC) FLEXURAL PROPERTIES

Guzlena S. [(1)] * **and Sakale G.** [(1)]

[(1)] Institute of Technical Physics, Faculty of Material science and Applied Chemistry, Riga Technical University. P.Valdena 7, Riga, Latvia

* Corresponding author: sandra.guzlena@rtu.lv

ABSTRACT

Glass fibre reinforced concrete (GRC) is lightweight material mostly used for façade panels and decorative elements. GRC can be made using two methods – spraying and premixing. Glass fibre in both cases has main influence on material flexural properties and ductility. Historically ordinary E type glass fibre has been used, but during concrete aging and alkaline medium fibres become fragile (weight and diameter loses). New type, alkali resistant (AR) glass fibres have been developed. In this research AR glass fibre amount and length influence on GRC flexural properties is investigated. Fibre length was changed from 6 mm till 41 mm for different samples and cut during spraying process. Fibre amount was changed from 0 – 7%. Samples were analysed using SEM-EDX to evaluate AR glass fibre and concrete matrix bond. GRC mechanical properties was evaluated using four-point bending tests and characterised by level of proportionality (LOP) and modulus of rupture (MOR).

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KEYWORDS: GRC, AR glass fibre, flexural strength

1. INTRODUCTION

Concrete is most widely used engineering material in construction all over the world due to its strength, durability and low cost as compared to other construction materials. The major drawback of concrete is its low tensile strength. Crack appearance due to stress during loading reduces concrete strength, durability and makes concrete more vulnerable to deleterious outside environment [1]. To increase tensile strength different fibres, like organic, polymer, basalt, steel and glass fibres can be incorporated in matrix.

Glass fibre reinforced concrete (GRC) is a composite which consist of concrete and glass fibres. Glass fibre has low water absorption, high modulus of elasticity and tensile strength [2]. Fenu et.al. have even concluded that basalt fibre reinforcement was shown to be less performing than glass-fibre reinforcement when used under dynamic conditions [3]. This material is used for non-structural parts of buildings like facade panels for over than 30 years. GRC is known for its high tensile and impact strength which is due to glass fibres. These properties allow make GRC panels 12 mm thin [4]–[6]. There are two types of techniques used to produce GRC elements. First of them is premixing, when concrete is mixed with chopped glass fibres and then casted in formwork. Fibre length usually is around 12 mm and it is added 3% of mortar mass. If the fibre length and amount is increased workability of the mix is reduced. Barluenga et.al. have noted that using short fibres in premix the cracked area of concrete surface can be reduced. If crack appears perpendicular to fibre it limits the size of the crack, but if the crack appears parallel to fibre, reinforcement does not work and crack size grows freely [7]. For spray technique fibre length can be changed, but usually it is 25-40 mm and 5% by mortar weight. During spraying process uniform well, mixed composite is achieved comparing to premix method. This type of material is with high durability, impact and tensile strength [5], [8].

Glass fibres used in material must be alkali resistant due to concretes high alkalinity. In the past, ordinary glass fibres, like E glass fibres were used, but during the time they showed low durability. Fibres lose their durability due to hydration reaction in concrete. Portlandite ($Ca(OH)_2$) is produced during hydration process and increases alkalinity in cementitious matrix to high alkalinity (pH 12). Hydroxyl ions (OH^-), which is at high concentration in cementitious matrix due to alkaline pore solution, brakes Si-O-Si bonds in glass fibres Eq.1 [9], [10] and causes weight and diameter loses making fibres fragile [6], [8], [9], [11].

$$- \overset{|}{\underset{|}{Si}} - O - \overset{|}{\underset{|}{Si}} - + OH^- \qquad \overset{|}{\underset{|}{Si}} - OH + SiO^- \text{ in solution (1)}$$

To improve glass fibre chemical stability in concrete ZrO_2 have been added. In the EN 15422:2008 is mentioned AR glass fibre must contain minimum 16% of ZrO_2, to use it in concrete. But ZrO_2 addition do not solve his problem completely, as researches showed decrease in aged GRC flexural properties comparing to young GRC [12]–[14]. There exist different methods to reduce fibre embrittlement. Admixtures like fume, metakaolin, nano silica can be added to a concrete mixture. This permits pozzolanic reactions and transforms portlandite to C-S-H in this way decreasing alkalinity of the concrete [2], [6], [9]. Use of low alkalinity cement, like sulphoaluminate cement could solve the problem, because Portlandite ($Ca(OH)_2$) is not produced during hydration process, but pH value in pore solution still is high, around pH 10. To densify the interface between fibers and cement matrix with polymers, like PVA, AC and others. This technique reduces lime diffusion into fibres [9], [10].

Due to thickness and common application of GRC tensile test is used to evaluate GRC properties. Level of proportionality (LOP) and modulus of rupture (MOR) is used to describe sample tensile strength.

LOP value describes sample maximal linear elastic deformation, which means every load that is put below LOP will not harm material, no cracks will be seen on surface and material will return to its present state. Sample elastic region, below LOP, can be described by Hookes law stress is proportional to the strain and the slope is Young's modulus. Basically, LOP value demonstrates matrix maximum flexural strength. After load is increased over LOP value material exhibits plastic behaviour, it deforms irreversibly. In this moment multiple cracks can appear on GRC surface, but material still have load bearing properties. Increasing load elongation increases of GRC sample due to fibres connection in concrete matrix until crack is too large and fibres pull out or brake at fracture point. MOR values describe samples ultimate strength before failure [2], [8], [13]. LOP and MOR values are influenced by many factors as:

- fibre amount,
- fibre length,
- sprayed GRC or premix is used,
- compaction.

In this research impact of amount of glass fibres in composition, length of fibre on sprayed GRC properties have been evaluated.

2. MATERIAL AND METHODS

GRC samples were made using spraying method. Basic components of GRC was cement CEM I 52,5R, fine quartz sand, superplasticizer and acrylic polymer. AR-glass fibre roving was used. Fibre length from 6 mm till 41 mm was changed for different samples and cut during spraying process. Fibre amount was changed from 0 – 7% by weight of GRC material (concrete matrix) in the uncured, green state. Sample flexural test was carried according to BS EN 1170 [15]. Samples were cut from test board in size 275 x 50 mm and tested with 4-point bending test after 28 days. GRC samples and AR glass fibres were investigated using Phenom ProX Desktop SEM.

3. RESULTS

3.1. SEM – EDX analysis

For all tests have been used AR glass fibre roving which was cut during manufacturing process. In SEM image (fig.1) can be seen bundle of AR glass fibres. Fibre surface is smooth, and the shape is regular. EDX analysis was done on glass fibre fracture zone, marked in figure 1. In table 1 is shown element analysis. Base oxides in fibre is Si, Na and Zr. With EDX analysis is not possible to obtain precise amount of ZrO_2 amount in fibre, but this method can be used to indicate Zr in fibre and to know that this is AR glass fibre.

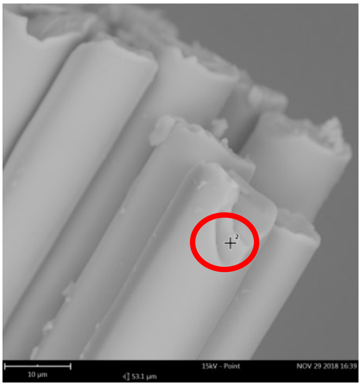

Figure 1. SEM image of AR-glass fibre used in GRC samples. EDX analysis was done at marked location.

Table 1. AR-glass fibre element analysis

Element Symbol	Weight Concentration, %
O	46.92
Si	25.32
Na	12.72
Zr	9.38
Ti	2.91
C	2.75

In figure 2 calcium carbonate crystals [16] are grown on surface of AR glass fibre which means that glass fibre surface has good bonding abilities with concrete matrix. Fenu et.al. have also noted that fibres in fracture are almost completely covered by a thin surface layer formed by the products of reaction between the outer alkali resistant glass of the fibre and the cementitious matrix [3] .

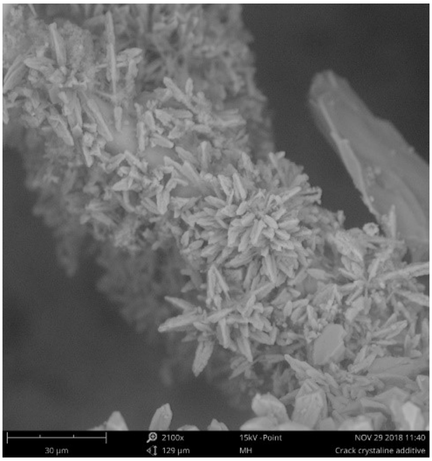

Figure 2. AR-glass fibre in GRC sample

In figure 3 is shown fibre bundles incorporated in GRC matrix. GRC performance in strength depends on mortar bonding with fibre filaments, fibre bundles and concrete matrix strength [8]. As can be concluded from the SEM image fibre bundles have good incorporation in matrix, no air voids or cracks can be seen around interface.

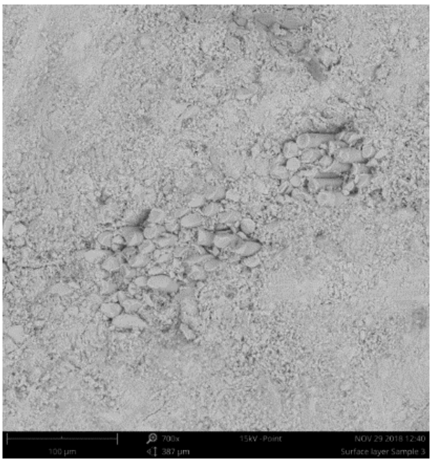

Figure 3. AR-glass fibre bundles in GRC sample cross section

3.2. Mechanical properties

Concrete as itself is brittle material in tensile strength. Fibre addition increase material elastoplastic behaviour. In this section GRC property dependence from fibre amount and length are presented.

AR glass fibre addition to concrete matrix has significant impact on sample ultimate load bearing capacity. As shown in figure 4, if 41 mm long fibres are used MOR values are increased with increase of fibre amount in the concrete, meanwhile LOP values do not change significantly. LOP value is more affected by the strength of concrete matrix than fibre addition [2].

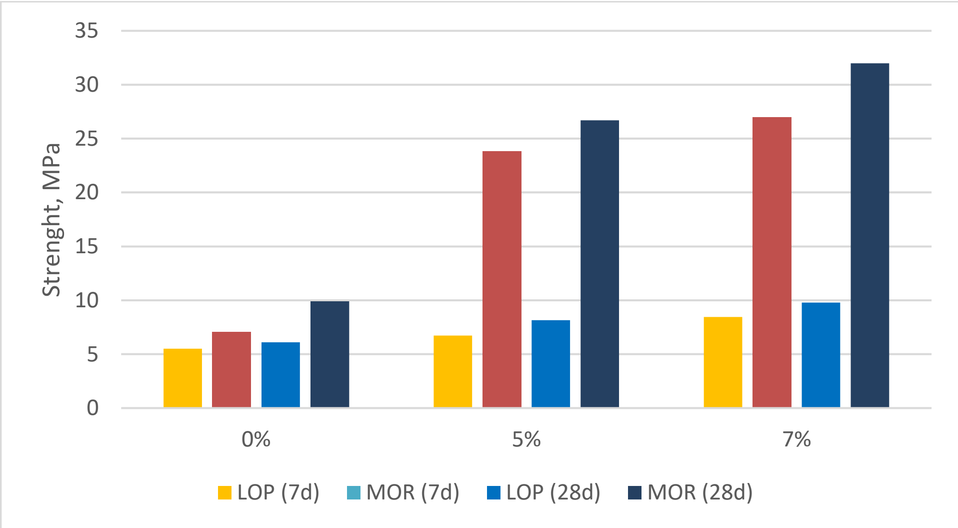

Figure 4. Influence of fibre amount on LOP/MOR values

Concrete as material is nonelastic, brittle material, as shown in figure 4, LOP and MOR values are almost the same if no AR glass fibres are added to concrete matrix. In figure 5 is shown that test samples without fibres are with very low plasticity and brittle fracture point. When the amount of fibres is increased from 0% to 5% and 7%, material elasticity is increased. By increasing fibre amount from 5% to 7% increases the load-bearing capacity of the material, but as the fibre length has not change the deflection of material does not change.

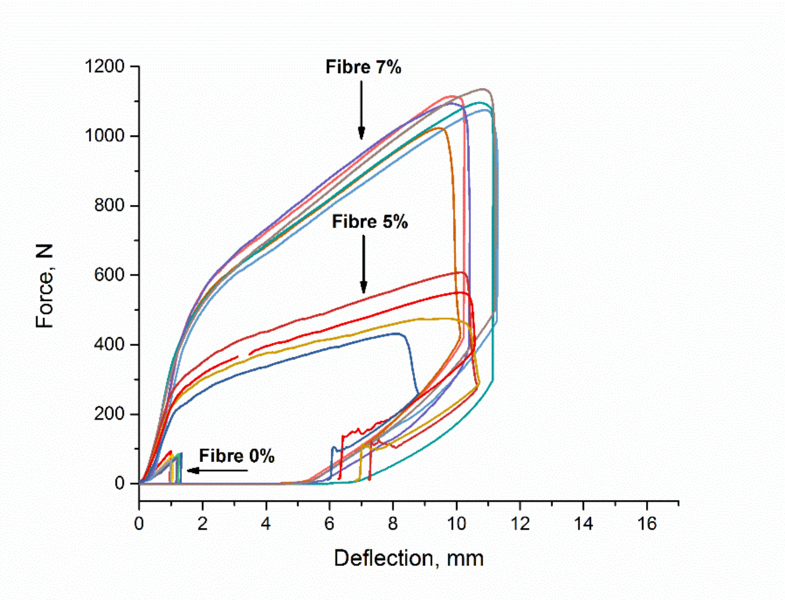

Figure 5. Influence of fibre amount on deflection

Fibre length was changed from 6 mm to 41 mm, but amount during these experiments was 5%. In Figure 6 is shown LOP/MOR dependence from fibre length. Fibre length do not considerably change LOP values. As we concluded before the LOP values are more dependent on the matrix properties of the concrete. MOR values increases by increasing fibre length from 6mm to 41 mm.

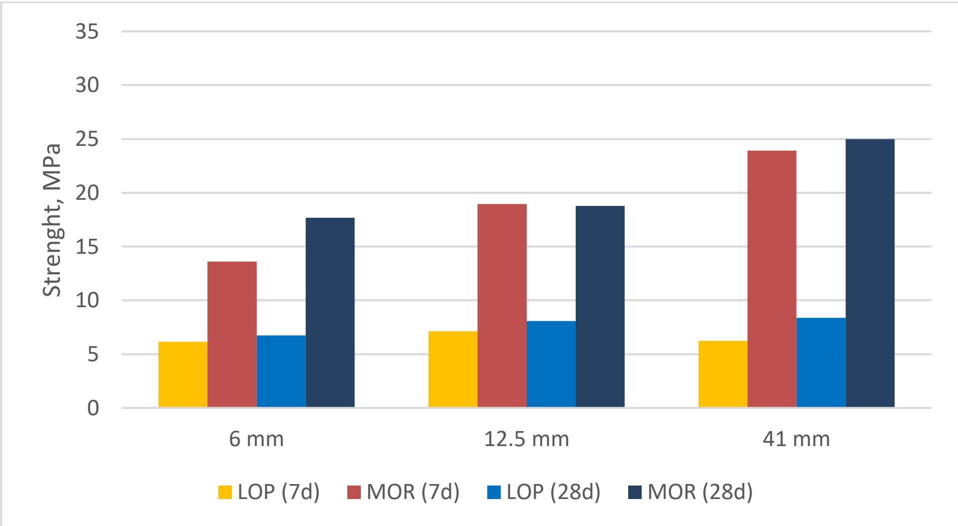

Figure 6. Influence of fibre length on LOP/MOR values

As shown in Figure 7 by adding longer fibres plastic behaviour of material increases. H. Kasagani et. al. have also noted that samples with longer fibres have higher deformation capacity comparing to smaples with shorter fibres [17]. Longer fibres have higher chance to defom in the bundle structure improving post crack deformation and still keep increment of force applied to GRC. Analysing collapse of samples can be seen that using longer fibres deflection stays constant for higher ΔForce. The filaments slowly pulls out of the bundle, not brittle when applied under load, crack size (deflection) do not change.

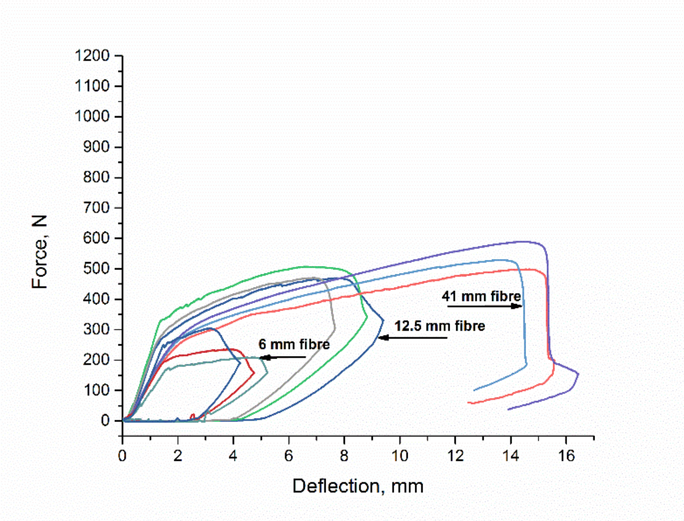

Figure 7. Influence of fibre length on deflection of material

4. CONCLUSIONS

Using AR-glass fibres is possible to change GRC flexural properties:

- by increasing fibre amount from 5% to 7% increase the load-bearing capacity of the material, but as the fibre length of all samples have been the same (41 mm) the deflection of material does not change.
- LOP value, which describes material elastic deformation, is dominated by concrete matrix flexural strength, addition of fibres has inessential effect.
- by increasing fibre length from 10 mm till 41 mm ,MOR values increases considerably due to composite plastic behaviour.

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Paper IV

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Crack Reduction during Drying Process by Using Surfactant

Sandra Guzlena[1], Gita Sakale[1], Sergejs Certoks[2] and Arnita Spule[1]

[1]Institute of Technical Physics, Faculty of Material science and Applied Chemistry, Riga Technical University, Paula Valdena 3, LV-1048, Riga, Latvia
[2]LODE SIA, Lode Street 1, Priekuļi country, Latvia

Abstract. In this research clay brick samples were made from illitic clays. Surfactant was added to clay mass during formation process to reduce crack generation during drying process. Surfactant amount was changed below critical micelle concentration (CMC). Nonionic surfactant Triton X-100 was used. Samples were prepared using extruder. Clay mass samples were analysed by granulometry, surfactant by contact angle and surface tension measurements and brick samples by microscope to determine crack amount. Changing surfactant amount closer to CMC is possible to reduce crack amount in sample during drying stage.

1 Introduction

Clay products have been used as building material already from ancient times, but it is still popular material in current building solutions. Nevertheless, there are still challenges to improve brick making technology, as an example, to decrease drying time, by using higher temperatures or lower moisture. But that can result in brick cracking. Brick cracking can cause mechanical strength decrease and big amount of defective bricks [1]. There are different ways to avoid this problem – glass fibre addition to clay mass [2], submicron particle addition (PVA submicron particles), temperature and moisture precise regulation during drying and firing process [3], and also addition of surfactants during material mixing before brick forming process [1], [4], [5].

Drying process is often one of the most complex operations in a manufacturing process. Drying does not only refer to the removal as much as possible moisture but also to the protection of the physical structure and appearance as an important part of the drying process [6].

Drying process can be split in three parts. At the beginning of stage I, the wet bricks and tiles have a moisture content 20–30%. In this stage heat is added. When product heats up until it reaches a certain temperature. Moisture diffuses by capillary action and first evaporates from the region near the solid surface. As some of the surface moisture evaporate, more moisture is transported from interior of the solid to its surface. For optimal drying, this process should be done slowly, because if the heat transfer takes place quickly, it becomes impossible for the moisture inside to escape and because of this some cracking may occur. At the second stage moisture content is crucial due to the water level in the clay reaching the critical point. The critical brick moisture content is an important parameter for determining the coefficient of sensitivity of clays to drying. It means the moisture of a solid material must be determined experimentally. At the third stage, the drying rate can be increased by providing the maximum amount of heat to reach the minimum moisture content (approximately 3–10%). Sometimes the damage done during drying process can be observed only after firing [6].

Mostly cracks appear during first part of drying process, which can be explained with tension between dry brick surface and wet brick core. This is due to nonlinear moisture and/or temperature changes in material. If surfactants are used tension between surface tension and material pour walls can be controlled, to improve moisture transport from brick core to surface [1]. Positive influence is also on clay particles to achieve maximal electrokinetically potential increase and wetting. This influences in good way electrostatic rejection between the particles by changing layered structure in fibrous state with high surface roughness [7], [8]. Clay surface properties changes with added surfactant amount. As Kowalski et. al. [1] have investigated that if dedocilsulfate and fluoric surfactants are used at very low concentrations (0,001%) surfactant must have been used to reduce crack formation. Clays with tendency to swell, which can cause cracking in contact with amido-amin bas cationic surfactants decrease this risk and reduce cracking [4].

Triton X-100 surfactant which was used in this study was chosen, due to M. J. Sánchez-Martín et.al. [9] ilitic clays have better adsorption to surface than octadecyltrimethylammonium bromide (ODTMA) and sodium dodecyl sulphate (SDS). Most of the previous investigations restrained the concentration range below critical micelle concentration (CMC) or near CMC, for which the maximum of adsorption was reached [1], [10]. CMC is concentrations where molecules of surfactants arrange into micelles and all additional surfactants added to the system go to micelles:

• Before reaching the CMC, the surface tension changes strongly with the concentration of the surfactant.

• After reaching the CMC, the surface tension remains relatively constant or changes with a lower slope.

• For the improvement of moisture transport the surfactants concentration should be below CMC [1].

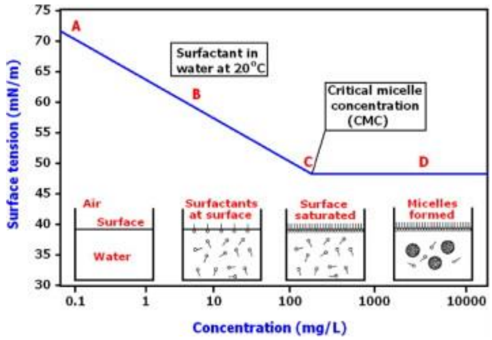


Figure 1. Micelle formation process [11].

Gluck (2010) study revealed that the CMC, an important property of surfactants, does not act as a threshold in the soil behaviour. If it did, soil properties would not change for concentrations higher than the CMC. But, as shown from experiments, increasing concentrations above the CMC had an effect on plasticity limit and particle compaction. [5]. So, in our research we use concentrations above CMC concentration.

2 Materials and methods

To form bricks four types of components were used - grey, red and composite clay and sand from the biggest quarry in Latvia (Figure 2). Clay samples were analysed and discussed in our previous work [12]. Using raw material from quarry without additives considering only to grain size distribution requirements most appropriate is composite clay due to its balance between all fractions. To use grey and red clay, mixes must be stabilized with sand size particles to avoid unnecessary shrinkage during firing. Sand found on quarry would be suitable stabilizing additive for plastic clay. Illite and Kaolinite are the only clay minerals found in the samples. Suitable brick making material should have suitable proportion of illite, kaolinite and quartz. Illite could provide plasticity, kaolinite promotes fired hardness and sand grains act as a stabilizer and filler material.

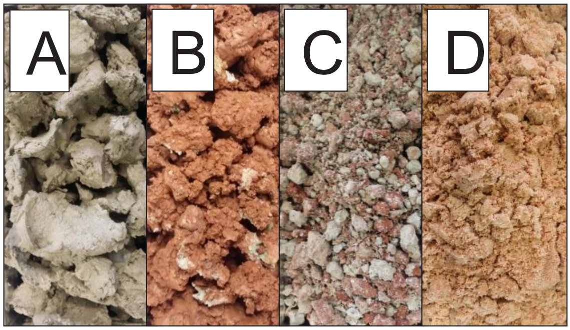


Figure 2. A - Grey clay; B – Red clay; C – Composite clay; D – Sand.

As surfactant used for sample making is Triton X-100 from sigma Aldrich. Triton X-100 or polyethylene glycol *tert*-octylphenyl ether is anionic surfactant (Figure 3).

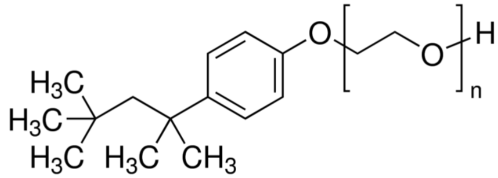


Figure 3. Structural formula of Triton X-100 [13].

Contact angle was measured using Triton X-100/water contact angle and drop test on glass plate. Contact angle was measured automatically for 30 seconds recording readings. Test was done three times for each concentration. The water contact angle measurements were made by the drop method by an optical tensiometer Theta Attension (Finland).

Surface tension was measured by plate method. Measurements were made by Kruss 100 M tensiometer. Surface tension was detected at 25°C with Plate method (20x10x0,2 mm). Depth of emersion of plate 1mm.

Granulometry was made by the pipette method. Raw material was dried at 105°C sieved till<2mm fraction. Oxidation of organic matter was done by hydrogen peroxide (H_2O_2). Samples were dipped in distilled water whit dispersant (Na_2PO_3) +Na_2CO_3 for 24 h. The aim of pre-treatment is to promote dispersion of the primary particles, and to get more accurate clay size particle amount in the sample. Suspension was wet sieved on 63 µm sieve. Fraction >63 µm that left on sieve was dried at 105°C and dry sieved. Fraction that passed 63 µm sieve was analysed by sedimentation method taking samples at appropriate intervals [14].

Cracks were analysed using digital camera (Moticam 2000) and software Motic Images Plus 2.0.

3 Results

3.1. Contact angle measurements and surface tension

In table 1 is shown contact angle measurements between glass plate and different concentration droplet of Triton X-100. Wettability increases or contact angle decreases by increasing concentration of Triton X-100. Using Triton X-100 concentration of 0,00001% contact angle is even higher than for water. Which means that to improve wettability lowest concentration that can be used is 0,0001%.

Surface tension values are shown in table 1. It can be concluded surface tension increases if surfactant concentration is decreased.

Table 1. Results of contact angle and surface tension measurements of different Triton X-100 concentrations.

Triton X-100 molar concentration	H_2O	0,1	0,01	0,001	0,0001	0,00001
Contact angle °	20,9	12,9	13,8	16,7	17,3	26,2
Surface tension mN/m	71,8	31,9	31,8	31,6	37,6	55,7

In Figure 4 are shown CMC determination. CMC can be determinate easily at 0,001%. Below this concentration surface tension values changes very slowly are almost constant, but above CMC surface tension changes very fast. All samples are made with surfactant concentration above CMC to achieve improvement of moisture transport and less cracking during drying.

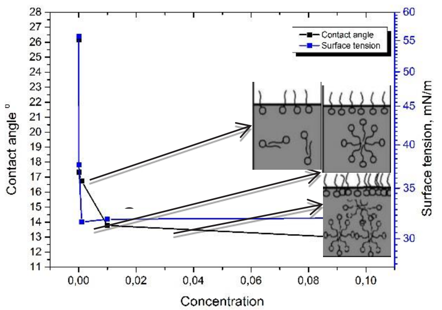


Figure 4. Contact angle and surface tension dependence on concentration. CMC detection.

3.2 Granulometry

Granulometry was carried out using pipette method for all samples. Granulometry in all three samples, VAV0,01 with Triton X-100 concentration 0,01%, VAV0,001 with Triton X-100 concentration 0,001% and VAV0,0001 with Triton X-100 concentration 0,0001%, are almost the same. Sand size particles >20µm are about 45%, silt size particles 2-20 µm are about 25% and clay size particles are about 30%. Due to Figure 4 B), sample granulometry is right in place for perforated bricks, but as we have extruded full bricks more sand size particles should be added to improve workability and drainage paths.

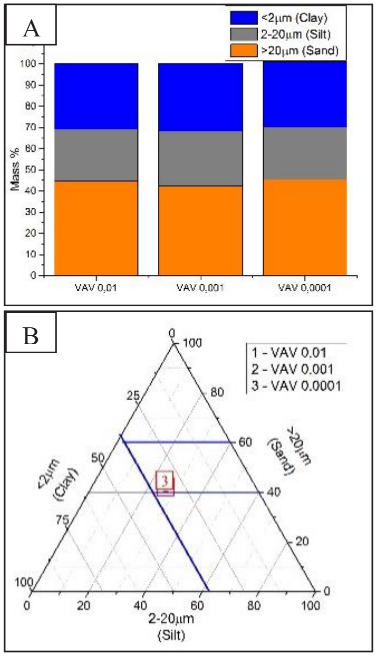


Figure 5. A) Particle size distribution of VAV0,01 with Triton X-100 concentration 0,01%, VAV0,001 with Triton X-100 concentration 0,001% and VAV0,0001 with Triton X-100 concentration 0,0001%; B) VAV0,01, VAV0,001, VAV0,0001 location in Winklers diagram.

3.3 Crack amount

In table 2 is shown crack amount as average crack area of dried sample. In different publications are different opinions of results below or above CMC [1]. The best quality of the dried samples was achieved for clay saturated with water containing rather low concentration of surfactants. But R. Guéganet. al. [10] have come to conclusions that nonionic surfactant (C10E3) concentration range above CMC or near CMC, gives the maximum adsorption to montmorillonite clay surface.

Our suggested drying mechanisms with surfactants are shown in Figure 6. Hydrophilic head of surfactant molecule adsorbs to clay structure and hydrophobic tail stays in pours. Water molecules during drying process are rolling through hydrophobic tails and does not adsorb to other particles or do not interact in structure. This mechanism helps water molecules to move easier from brick centre to surface.

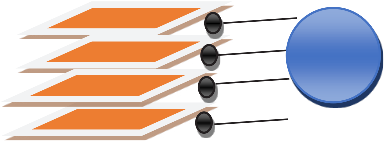


Figure 6. Drying mechanism in clay when surfactant is used.

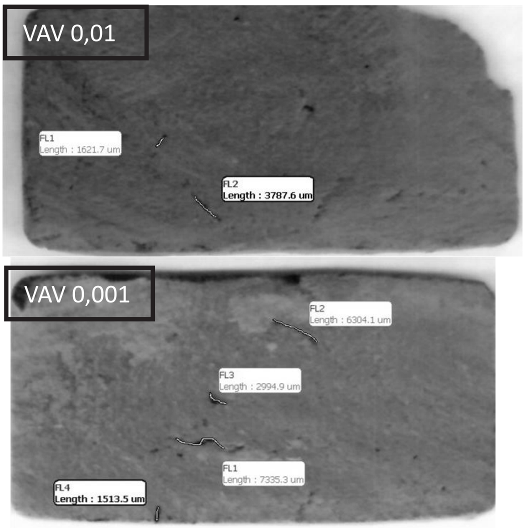
3.4 Evaluation of crack

Cracks were measured after drying. Average crack area on sample area was measured. Results are shown in table 2. Better results are achieved if higher Triton X-100 concentration is used and it is closer to CMC.

Table 2. Crack amount of samples with different Triton X-100 concentrations.

Triton X-100 molar concentration	0,01	0,001	0,0001
Crack amount, %	0,07	0,09	0,10

In Figure 7 are shown sample crack measurements. Crack in samples are mostly concentrated in the middle and they are round shaped. Round shaped racks come from extruder auger. This type of crack is combination of flow lamination and rotational movement of clay mass. Also hallow space created by auger hub in the canter of emerging clay mass which have not entirely joined as a result of mass characteristics or geometry of pressure head and die [8]. So, these cracks are not from drying process and must be eliminated during forming process.



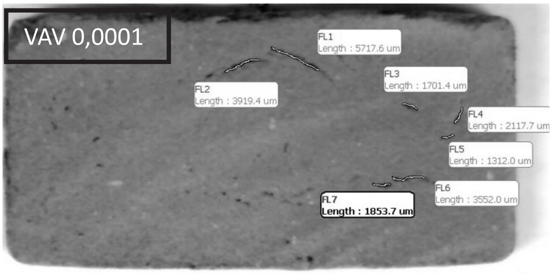


Figure 7. Sample crack measurements.

4. Summary

Due to information available in literature, surfactant addition to clay materials to develop drying performance is important theme, because cracking even in drying process is a large problem and needs to be controlled and eliminated. In our research crack development was measured after drying process for samples with added surfactant Triton X-100 near and below CMC. Crack amount decreases from 0,10% till 0,07% if Triton X-100 concentration is increased closer to CMC. Crack amount decreases, because surfactant molecules help water molecules to move easier from brick centre to surface reducing tension between dry brick surface and wet brick core. Our results show that to achieve lower crack amount during drying process surfactant must be added near CMC.

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Paper V

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Effect of the addition of fibreglass waste on the properties of dried and fired clay bricks

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Effect of the addition of fibreglass waste on the properties of dried and fired clay bricks

S Guzlena[1], G Sakale[1], S Certoks[2] and L Grase[3]

[1]Institute of Technical Physics, Facultiy of Material science and Applied Chemistry, Riga Technical University. P.Valdena 7, Riga, Latvia
[2]LODE SIA, Lode street 1, Priekuli country, Latvia
[3]Institute of Silicate materials, Facultiy of Material science and Applied Chemistry, Riga Technical University. P.Valdena 7, Riga, Latvia
E-mail:Sandra.Guzlena@gmail.com

Abstract. The main objective of this study was to investigate the effect of the addition of fibreglass waste on the properties of the dried and fired clay bricks. Different amounts of waste glass (0 – 10 wt %) were added to the original brick clay and fired at 1000 °C. The effects on the technological properties of the bricks such as compressive strength, water absorption and density after firing were investigated. Also cracks and fibreglass influence in dried and fired samples were analysed by digital camera and SEM-EDX analysis.

1. Introduction

Fired clay bricks are construction materials which have been used since ancient times and still are used in nowadays. However, demand for higher quality fired bricks are growing in modern construction. Bricks have been designed to become more homogenous and porous, with higher compressive strength due to the ceramic bond from the fusion phase of silica and alumina constituents in clay. The firing process sinters the particles of clay together to form a bond, which gives the bricks its characteristic strength and durability. The sintering process is achieved by heating silicon dioxide or quartz (SiO_2) which occurs naturally in clay and shale to high temperatures, causing it to melt. Sand which consist of quartz function also are to control shrinkage, lamination and to provide gas drainage paths during firing. Upon cooling, the quartz forms a bond between adjacent clay or shale particles at the points of contact [1].

The world production of bricks and ceramic tiles requires massive amount of natural raw materials. These natural raw materials can tolerate further compositional variations and raw material changes, allowing different types of wastes to be incorporated into the internal structure of bricks as part of their own matrix. The amount of inorganic wastes in Europe is estimated to be more than 1,500 million tonnes. Traditionally, non-hazardous inorganic wastes have been disposed in landfills and often dumped directly into ecosystems without adequate treatment [2], [3]. So fibreglass waste can be utilised making clay bricks by substituting natural raw materials and reducing its use rates.

Many studies have shown that the use of waste glass in bricks produces positive effects on the materials properties [1], [2], [4]–[6]. In general glass addition to ceramic matrix accelerates the densification process and mechanical and physical properties are improved already with small amounts of additive, below 10 wt%. Glass rigid nature induces local constrains in the matrix reducing sintering shrinkage and changing the physical properties as compressive strength, water absorption and others [1], [4].

2. Materials and methods

Three basic types of clayey soil from quarry were used – Grey, Red and Composite. Sand used as non-plastic material was obtained from the same quarry. All obtained soils are shown in figure 1. Composite clay is colorful mottled, no carbonate clayey aleirolites and in them occurring like intermediate state - light gray, refractory clays [7]. Analysis and more detail information of clays and sand used in this research can be found in our previous work [8]. All types of clay and sand were homogenized and mixed with water to obtain plastic brick clay mass. Different amounts of waste fibreglass (0 – 10 wt %) were added to this mass. Due to other studies mechanical and physical properties are improved already with small amounts of additive, below 10 wt% [1], [2], [4]–[6]. Samples was extruded and its green size was (L x H x W) 12,0 x 3,4 x 7,0 cm. Samples after forming was dried at 105°C and fired at 1000 °C for 1h. Three mixes were made with 0 wt%, 5 wt% and 10 wt% of fibreglass. Following the particle size distribution and SEM data can conclude that most of fibreglass is dust size - from 20 to 2 μm.

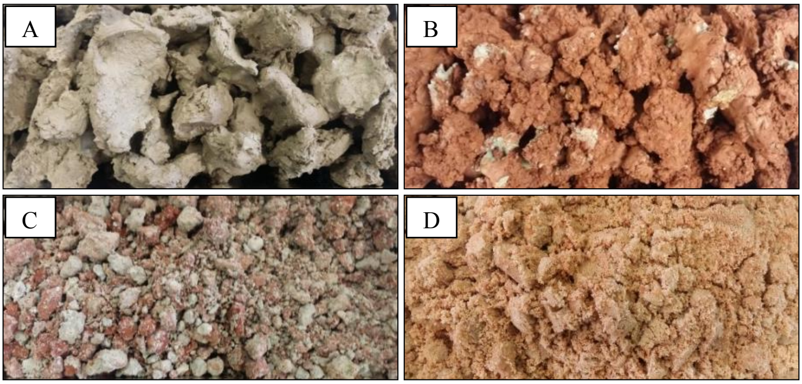


Figure 1. Soil from quarry: A – Grey clay; B – Red clay; C- Composite clay; D – Sand.

Particle size distribution was determined by granulometry analysis done by the pipette method. Raw material was dried at 105°C sieved till <2mm fraction. Oxidation of organic matter was done by hydrogen peroxide (H_2O_2). Samples were dipped in distilled water whit dispersant (Na_2PO_3) +Na_2CO_3 for 24 h. The aim of pretreatment is to promote dispersion of the primary particles, and to get more accurate clay size particle amount in the sample. Suspension was wet sieved on 63 μm sieve. Fraction >63 μm that left on sieve was dried at 105°C and dry sieved. Fraction that passed 63 μm sieve was analyzed by sedimentation method taking samples at appropriate intervals [9].

Crack detection was made of fired and dried samples which were cut in 8-10 pieces. Detection and measurements was made by digital camera (Moticam 2000) and software Motic Images Plus 2.0. The crack amount is expressed as a percentage of the average crack area on the sample area.

For samples morphology and chemical analysis Scanning electron microscope with integrated energy dispersive x-ray spectrometer (SEM-EDX) Hitachi S-900 was used.

3. Results and analysis

3.1. Granulometry

Granulometry was made for all mixes: Mix_7 without fiberglass additive, Mix_7_5% with 5% fiberglass additive and Mix_7_10% with 10% fiberglass additive. Our goal according to Winklers diagram for solid bricks is to make composition with >60% sand size particles, <60% silt size particles and <35% clay size particles (red triangle in figure.2.B)) [10]. As shown in figure.2.B) base composition Mix_7 is close to this location in diagram. By increasing fiberglass additive proportion, clay size particles increase, but sand size particles decreases (Fig.2.A). So fiberglass additive during mixing crushes due to its fragile nature and mostly are silt size - from 20 to 2 µm.

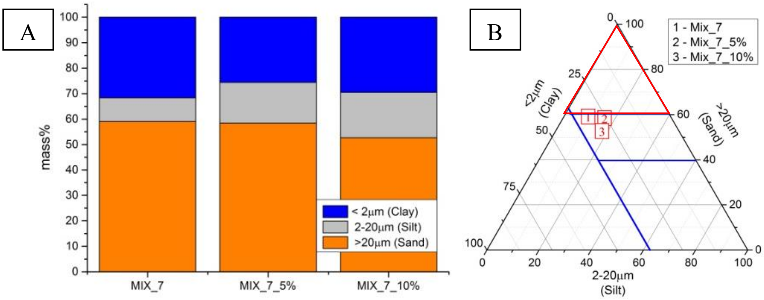


Figure 2. A) Particle size distribution of Mix_7 without fiberglass additive, Mix_7_5% with 5% fiberglass additive and Mix_7_10% with 10% fiberglass additive. B) Mix_7, Mix_7_5% and Mix_7_10% location in Winklers diagram.

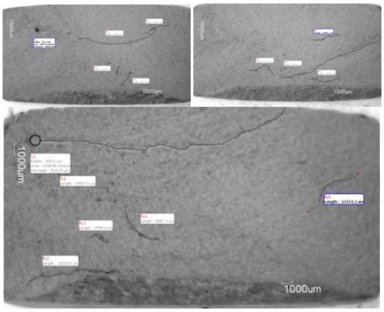
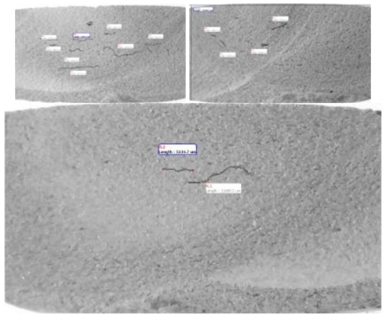
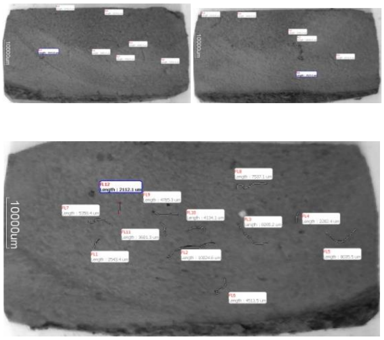
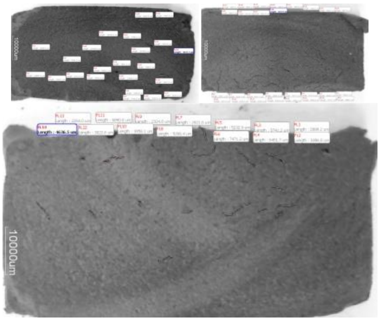
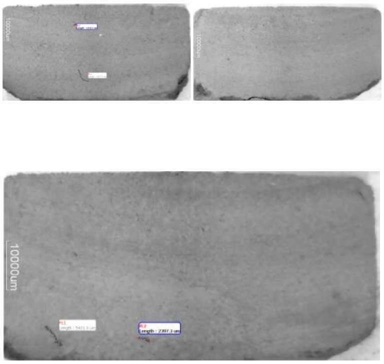
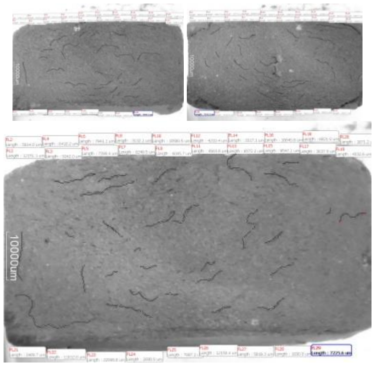
3.2. Cracks

Cracks in solid bricks were measured after drying and firing. The crack amount in table 1 is expressed as a percentage of the average crack area on the sample area. In table 2 are illustrated images of cracks and how they were measured. In both tables are expressed that by increasing fiberglass additive amount, percentage of cracks decreases in dried samples that are also consistent with the drying shrinkage results. Opposite from clays, that have plastic behavior and shrink after water evaporation, the fiberglass in the sample matrix reduces the shrinkage during drying thus avoiding cracking [2]. Opposite results are with fired samples. Firing shrinkage increases and crack amount increases by increasing fiberglass additive. That may be explained by silicon nature to melt around 573°C during heating and shrinking during cooling process [4]. Grate amount of silicon is found in added sand and fiberglass additive. Heating process during firing may be too rapid as well, which can increase absorbed water evaporation rate and cause cracking.

Table 1. Crack amount in dried and fired samples.

Sample	Granulometry (>20µm/ 2-20µm/ <2µm)	Average drying shrinkage	Crack amount of dried sample	Average firing shrinkage	Crack amount of fired sample
Mix_7	59,09/ 9,3/ 31,61	5,2%	0,26%	-1,14%	0,99%
Mix_7_5%	58,41/ 16,00/ 25,59	3,6%	0,37%	0,15%	1,62%
Mix_7_10%	52,72/ 17,85/ 29,43	2,7%	0,04%	1,27%	2,07%

Table 2. Crack images of dried and fired samples of Mix_7 without fiberglass additive, Mix_7_5% with 5% fiberglass additive and Mix_7_10% with 10% fiberglass additive.

	Dried samples	**Fired samples**
Mix_7 without fiberglass additive		
Mix_7_5% with 5% fiberglass additive		
Mix_7_10% with 10% fiberglass additive		

3.3. SEM EDX

SEM images were taken of 1000°C fired all three mix samples. As shown in table 3 large quartz grains are embedded within the clay matrix, but the finer fraction of quartz is also distributed around. In samples without fiberglass additive micro-cracks has formed around large quartz particles which also are larger crack formation reason as discussed above due to its nature to dilute during heating and

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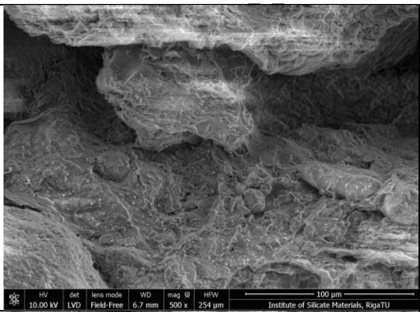
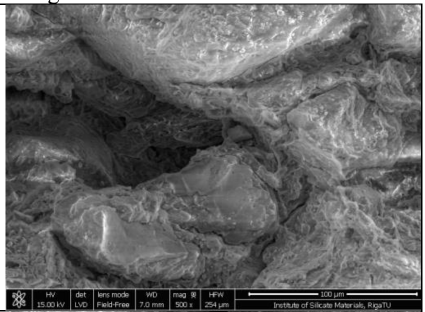
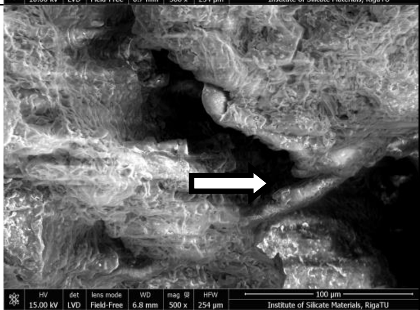
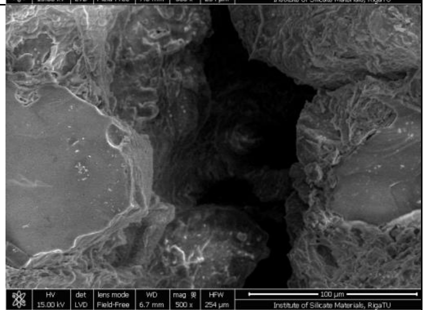
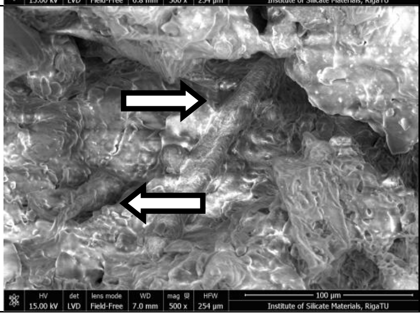
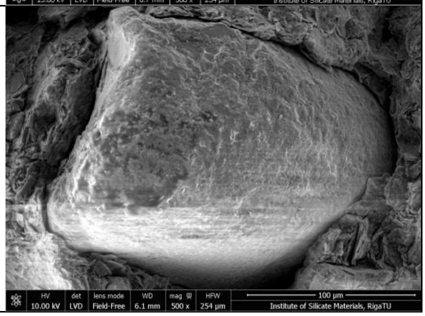
shrink during cooling process. In samples with fiberglass additive, which is indicated by arrow, clay like structures has grown around them during firing process and additive work like reinforcement.

Table 3. SEM images of Mix_7 without fiberglass additive, Mix_7_5% with 5% fiberglass additive and Mix_7_10% with 10% fiberglass additive.

Mix_7 without fiberglass additive		
Mix_7_5% with 5% fiberglass additive		
Mix_7_10% with 10% fiberglass additive		

Of all three mixes EDX analysis has been made and all typical clay elements have been found like Si, Al, O, K as shown in figure 3. According on our previous research [8] these elements has been found in illite, kaolinite and quartz structures which were detected by XRD analysis. Most common elements found is Si, which is 46% of all scanned area and O (27%). These elements can be found in illite $KAl_2Si_3AlO_{10}(OH)_2$, kaolinite $Al_2Si_2O_5(OH)_4$ and quartz Si_2O_2 structures [11], [12].

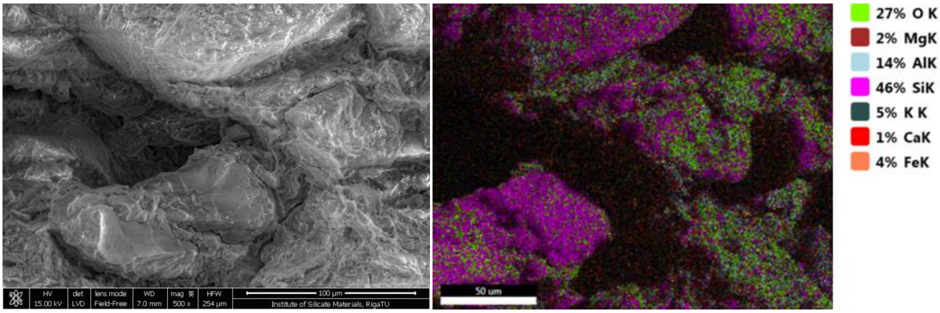


Figure 3. Image of SEM and EDX analysis.

3.4. Technological data

Some technical data are summarized in table 4. Forming moisture for all three mixes is around 18%, which is suitable moisture for clay mixture forming. Increase of fiberglass additive to clay increases the sintering shrinkage, as it was discussed above, leading to a densification of material. Fiberglass additions change the recrystallization processes, leading to the formation of dense clay-glass agglomerates distributed within the three dimensional quartz network [4]. Clay-glass agglomerates affect compressive strength. Sample density, after firing, increases by increasing fiberglass additive amount. As the density of a clay brick increases, its strength also increases.

Water absorption was measured after firing by immersing samples in water for 24 h. As shown in table 4 it is high, but comparable with N. Phonphuak et al. results which were in the range of 14.78 - 18.66%, depending on firing temperature [1]. High water absorption is related with samples open porosity and cracks that accrued after firing process. Thus the pores of fired clay brick are affected of glassy phase formation during firing process.

The compressive strength of clay brick is the most important engineering-quality index for building materials. Compressive strength increases by increasing fiberglass additive amount. As discussed above fiberglass has good adhesion with clay structures and that works like reinforcement, which strengthens the structure. Our sample compressive strength is very similar with C. N. Djangang et al obtained compressive strength with 10% glass powder addition to kaolinite clay which is 10,49 MPa. [4], [13]. With a higher compressive strength, other properties like flexure, resistance to abrasion are also improved. While other properties are relatively difficult to evaluate, the compressive strength is easy to determine [1].

Table 4. Technical data of Mix_7 without fiberglass additive, Mix_7_5% with 5% fiberglass additive and Mix_7_10% with 10% fiberglass additive.

	Forming moisture	Density, g/cm^3	Water absorption	Compressive strength, MPa
Mix_7	18,40%	1,60	13,84%	7,86
Mix_7_5%	18,00%	1,62	16,42%	5,77
Mix_7_10%	17,90%	1,67	14,66%	10,04

4. Conclusion

By addition of fiberglass waste in clay mixtures, we gain several benefits, firstly, the amount of waste is reduced and secondly, properties of clay bricks are improved. Addition of fiberglass waste (size range 20 to 2 µm) show good influence on mechanical characteristics of clay bricks that is compressive strength and density increases by increasing fiberglass amount in mixture. Fiberglass works as reinforcement for clay matrix. However, in future it is planned to use submicron reinforcing materials to gain higher improvement in crack reduction of clay materials.

Acknowledgments

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Paper VI

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Self-healing concrete with crystalline admixture – a review

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Self-healing concrete with crystalline admixture – a review

S Guzlena[1] and G Sakale[1]

[1]Institute of Technical Physics, Faculty of Material science and Applied Chemistry, Riga Technical University. P.Valdena 7, Riga, Latvia

Abstract. Concrete is the most used construction material for years. However long term serviceability is an issue, when we focus on sustainable building and use of materials. In long term service different types of visual and structural defects can appear in concrete structures. Defects in material enhances due to aggressive factors in outdoor (chlorides, sulphides and carbonates, freeze-thaw) which reduces material properties over a longer period. Mainly defects on visible surfaces of structures are from mechanical impacts, environmental exposure or due to impropriate maintenance. There are methods to reduce the risks and improve service life. One of them is self-healing materials, like bacteria, crystalline additives, in-capsulated healing agents and other. Use of self-healing material has preference, like lower maintenance costs for inspection, monitoring and complicated repair. Crystalline admixtures have several advantages comparing to other self-healing techniques, like improved concrete water tightness, no need for encapsulation before addition to concrete mix, which reduces sample preparation time.

1. Introduction

There are several chemical and physical factors, which affect aesthetic look and shorten service life of concrete structures. That is concrete structure exposure to aggressive agents as chlorides, sulphides, carbonates; water penetration through pours and small cracks due to absorption or hydrostatic pressure; freeze-thaw cycles [1]. As a result pores and small cracks like hairline cracking can be observed in concrete structures. To prolong service life self-healing materials can be used. Self-healing materials have many advantages: less people work and money saving due to inspection, monitoring and repair. Self-healing of concrete is triggered on its own. Self-healing ability of concrete [1], stainless steel reinforced concrete [2], high-performance concrete [3] and high-performance fibre reinforced concrete (HPFRC) [1], [3] have been researched. Due to healing mechanisms it can be divide in two parts:

- autogenic healing: natural closure of cracks due to un-hydrated cement in concrete and no specific self-healing admixtures are added. But this type of self-healing is not predictable and cannot be controlled [4], [5].
- autonomic healing: engineered closure of cracks with additives which is not a part of a ordinary cement [3]–[7]:
  - Passive – no need for human involving, self-healing starts from external promoters [2], [8].
  - Active – human involved to activate and complete the self-healing mechanisms [2], [8].

A lot of researchers have been investigated autonomic self-healing mechanisms with fibre reinforcement [1], [3], bacteria [9]–[12], crystalline additives [1], [3], [7], [13], [14], absorbent polymers [15], absorbent clay materials [6], incapsulated healing agents [4], [6], [16], expansion agents like sulfo-aluminate [6], [13], [14] and others.

In this review most of interest is focused on chemical additives – crystalline additives (CA), because of their dual positive influence – crack healing and concrete permeability reduction ability. In moist environment CA react with cement and forms calcium silicate hydrates. These additives help to reduce concrete porosity and water permeability as well [4]. Difference between CA (permeability reducing admixtures) and hydrophobic additives are - the first ones have hydrophilic nature apart prom CA [7]. Due to Belie et. al. [17] CA performance is largely influenced by mix design. Better effect can be noticed with HPFRC. But still healing process for 0,2 mm wide cracks is relatively time demanding from few weeks to several months.

2. Autogenous crack healing

Autogenous crack healing mechanisms has been studied almost for forty years. Main conclusions are that crack healing mechanisms changes by concrete age. In case of young concrete main mechanism is hydration of un-hydrated cement particles in concrete in presence of high moisture [1], [3], [4], [18], [19]. In case of concrete with low water:cement ratio, <0,3, even 20-30% of all cement particles can be un-hydrated and participate in self-healing process [5], [20]–[22]. For older concrete calcium hydroxide ($Ca(OH)_2$) converts to crystalized calcium carbonate ($CaCO_3$) as a result of exposure to atmosphere with carbon dioxide (CO_2) [1], [4], [7], [18], [19], [23]. This phenomena first was discovered in 1836 by the French Academy [8]. Crack size which can be healed by this mechanism's variates from 0,05 mm till 0,3 mm, but it can be unpredictable. Much more presence of water, not moisture, is needed [6], [14], [24]. Different additives such as fly ash, blast furnace slag, silica fume, limestone powder promote autogenous crack-healing in later stage in concrete due to un-hydrated binders, which have slower pozzolanic reactions [1], [3], [4], [7], [18], [23], [25]. These pozzolan materials in presence of water reacts with portlandite and produce binding products [25]. Van Tittelboom et al. have done research adding to Portland cement fly ash or blast furnace slag as pozzolanic additives. It was found that concrete crack self-healing rate is 0,015 mm/day, and are influenced by environmental conditions [26]. K. Tomczak et.al. has challenged autogenous healing by making samples with very low w:c ratio 0.28 and 0.23. Pre-cracked samples with crack width 0-0,8 mm was healed in water for 28 days. Cracks below 0,46 was completely healed after 2 months, but wider cracks have healed only for 15-30%. Self-healing speed also changes during time, but most influencing factor is crack width, if cracks are wider healing considerably slows down [27].

3. Autonomous crack healing using crystalline additives

Crystalline additives are mostly water soluble additives which improve concrete properties at fresh and hardened state [24]. As American concrete institute (ACI) writes on their report crystalline additives are one type of permeability reducing admixture. ACI divide permeability reducing additives in two parts. That reduces water permeability at no-hydrostatic conditions and at hydrostatic conditions. In category that reduces water permeability at no-hydrostatic conditions falls typical hydrophobic products. Admixtures that can function under hydrostatic conditions include crystalline admixtures. Some of these additives have a lot of benefits like reducing drying shrinkage, minimize chloride ion penetration to surface, improved frost resistance and autogenous healing properties [28].

Crack width that can be healed with CA variates till 300 μm. [4], [24] but if the techniques are combined, like crystalline additives with expansive admixtures, healed crack width can be increased till 400 μm [7].

Moisture is very important if chemical agents (crystalline additives) is added in concrete to promote deposition and fill the crack. Usually in outside environment it is not an issue [4], [7]. After crack is formed, in presence of moisture tricalcium silicate (C_3S) reacts and Calcium Silicate Hydrate (CSH) is deposited on crack walls and it fill the crack[7]. Quality of concrete self-healing ability can be affected by mix proportion, temperature, environmental humidity and stress in crack region. If stress is too high, at already healed crack area new cracks can appear [4]. Calcium carbonate as an deposit in self-healed crack can appear in different morphologies – calcite, aragonite, vaterite, monohydrocalcite, ikaite and amorphous $CaCO_3$. By-product which appeared after concrete self-healing with CA mostly is the same that can be seen after autogenous healing – calcite, brucite and aragonite.

It must be mentioned that self-healing using crystalline additives show better results when samples are immersed in water or subjected to wet-dry cycles [2].

Authors mostly use commercially available crystalline additives, as WT- 250 commercialized by Sika [3] Admix C-1000NF by Xypex [6] Penetron Admix by Penetron [5] or do not specify what crystalline admixture is used. Esscffres et. al. has mentioned higher Mg content in admixture which can lead to higher amount of healing by-product aragonite [3]. Other authors [1], [5], [6] have mentioned as one of compounds reactive silica or micro silica.

Results that are achieved differs, because of many different parameters used in each research. Starting from mixture: cement type, sand:cement proportion, water:cement proportion, different additives, dosage and type (superplasticizers, viscosity agents). There is a luck of standardized testing method, methods of testing differs almost in each paper. Therefore, it is hard to compare the results. In fallowing text research by different authors will be discussed one by one and summarized in table 1. This problem have mentioned also other authors and standardizations is needed [29]–[31].

Ferrara et. al. [1] used crystalline admixture which consists from cement, sand and micro silica. Admixture were investigated by EDS analyses and compared with Portland cement specimen. Only difference was slightly higher peak of sulphur. Normal strength concrete (NSC) and HPFRCC samples were pre-cracked after 2 months and healed by immersing in water and exposed to an open air. Crack width were 0,2 mm for NSC and deflection till 2mm for HPFRCC. Normal strength concrete and HPFRCC with crystalline admixture showed good healing capacity after 1 month. In both cases better results was achieved if samples were immersed in water. Sample testing continued and showed improvement even after 12 months [1].

Escoffres et. al. [3] used commercially available crystalline admixture from SIKA WT-200. EDS analysis was made, and high amount of Mg were found in this admixture. After 28 days sample prisms of HPC, HPFRC and HPFRC-CA have been exposed to three-point bending test to pre-crack samples with crack width close to 0,2 mm. Part of each material was immersed in water and one part left in air. After 56 days all prisms were tested again with three-point bending test. HPFRC-CA showed by 10-20% better results than HPFRC in air exposure. HPC, HPFRC and HPFRC-CA self-healing ability is discussed during constant loading similar to service conditions [3].

M. Roig-Flores and her team [7] has investigated crack-healing ability of concrete samples with and without crystalline admixture. Pre-cracked 2 days old samples (crack width bellow 0,3 mm) were investigated. Different conditions – water immersion, water contact, humidity chamber and air exposure were used to accelerate crack-healing. Conclusions are that crack healing abilities increase for samples with crystalline admixture that have been immersed in water. Cracks have been healed around 95%, but samples without admixture only 75%. Samples with and without crystalline admixture that have been exposed in humidity chamber and in air have very low crack healing ability. All samples have been compared after 0 and 42 days [7].

Wang et. al. [6] used porous expanded clay aggregates as sodium carbonate container, commercially available crystalline additive from Xypex Admix C-100NF and sulfoaluminate based expansive additive Denka CSA#20. Crystalline additive mainly consists from crystalline catalysts and reactive silica. Samples were pre-cracked after 7 days and healed in water for 28 days. Best self-healing results showed samples with mix proportions 10-12% of Na_2CO_3, 7-10% of sulfoaluminate based expansive agent, 4,5% of crystalline additive [6].

Sisomphon et.al. [14] used in his research crystalline additive and expansive additive. Crystalline additive consits from reactive silica and crystalline catalysts (hydrophilic water-proofing materials). As an expansive additive calcium sulfoaluminate based commercial product was used with mineral composition of Hauyne, anhydrite and free lime. Samples were pre-cracked after 28 days. Pre-cracked sample crack size was 0,1-0,4 mm. Samples were immersed in water for 28 days. Surface cracks up to 0,15mm can be closed in 28 days [14].

Cuenca et. al. [5] has done research of self-healing of fibre reinforced concrete after several self-healing - pre-cracking cycles. To inhance self-healing crystaline admixture was used. Samples were pre-cracked using Double Edge Wedge Splitting test and crack width was controled with crack mouth opening displacements (CMOD). Samples were cured after pre-cracking in water, at open air and wet-dry cycles. Samples at air represented low crack-healing ability. Completly crack healed only for samples with crack width less than 0,3 mm, which were immersed in water, dosen't matter perminantly or periodicaly. Ever more showed good repeated crack healing ability over an 1 year after repeated pre-cracking tests.

Coppola et.al. [32] has investaigated novel method of self -sealing concrete by adding in concrete mixture fumaric acid-vased admixture (WP). It was found that by using WP is posible to parcipitate denser crystals compearing with comercially available products. Cracks till 0,4mm is compleatly filled with white crystal products. No improvement on waterproofing was seen if doage of WP increased form 1 to 2 % versus cement content. Cracks till 0,4mm was healed alreay after 7 days after sample immersion in water.

Roig-Flores et.al. [2] has investigated healing in erly age concrete, that is, samples were pre-cracked at age 2 days. Crystalline admixture influcence on crack healing at different conditions were evaluated: water immersion at 15 °C and 30 °C and wet/dry cycles. Exposrue time 42 days. Best results were obtained for samples immersed in water and held at 30°C temperature. Samples subjected to these conditions gained 0.99 average healing ratio with smallest standard deviation in results. Cracks in width to 0.4mm were healed.

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Table 1. Summary of concrete types, mix design, used crystalline additives, pre-cracking methods and healing.

Reference	Concrete type	Cement type	W:C	Additive	Method to crack	Time till Pre-cracking	Crack width	Healing - days	Conditions
[1]	normal strength concrete	II 42.5	0,63	blend of cement, sand and microsilica	3-point bending test with COD-controller	35-42 days	130 and 250 µm	1, 3, 6 and 12 months	immersed in water or exposed to open air
[6]	normal strength concrete	Not specified	0,25	calcium sulfoaluminate based expansive additive (CSA, Denka CSA#20) and crystalline additive (CA, Xypex Admix C-1000NF)	3-point bending test. notch in the middle of sample	7 days	0–200 µm	28 days	healing in still water
[23]	normal strength concrete (NSC)	P II 52.5	0,45	calcium sulphoaluminate expansive agent and natural metakaolin with a small amount of additives	compressive strength tests	7 d	Not specified	28 d	water
[2]	fiber-reinforced concrete	II/A-L 42.5 R	0,6	not Specified	splitting test	2 days	100 and 400 µm	42 days	water immersion at 15 °C, water immersion at 30 °C, and wet/dry cycles
[5]	fiber reinforced concrete	II 42.5	0,5	crystalline admixture (Penetron Admix)	residual flexural tensile strength with CMOD	3, 7; 28 and 56 days and 4 months	below 300 µm	Till 6 months	submitted to air exposure
[7]	fiber-reinforced concrete	II/A-L 42.5 R	0,45	not specified	splitting test	2 days	below 300 µm	42 days	water immersion, water contact, humidity chamber and air exposure at laboratory conditions
[14]	fiber reinforced concrete	I 42.5 N	0,25	crystalline additive and expansive additive	splitting tensile strength test	28 days	100–400 µm	4, 7, 14- and 28-days	water
[3]	high- performance fiber reinforced concrete	Not specified	0,43	crystalline additive (WT- 250 by Sika)	three points bending with RCOD	28 days	200 µm	56 days	immersed in water submitted to air exposure
[1]	high performance fiber reinforced cementitious composites	I 52.5	0,18	blend of cement, sand and microsilica	4-point bending with COD-controller	2 months	130 and 250 µm	up to six months	immersed in water

4. Conclusion

Overlooking literature research on self-healing concrete has been made for different type of concretes, cements and w:c ratio in recent years. Various pre-cracking time has been investigated which can affect healing rate and results. If pre-cracking has been made at early stage of concrete hardening, autonomic healing goes together with autogenic healing and it is hard to differ these healing processes. Applied healing time differs from healing conditions. One can be concluded, if crystalline additives are used in cementitious materials more important is water then moisture in surrounding. Samples immersed in water gain faster crack healing and quality of healed crack is better, even cracks after repeated pre-cracking can be healed. This limits the use of the crystalline additive in materials for facades, panel constructions and other objects in the outdoor and indoor environment which are not in direct contact with water.

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Paper VII

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Self-healing of glass fibre reinforced concrete (GRC) and polymer glass fibre reinforced concrete (PGRC) using crystalline admixtures

S. Guzlena *, G. Sakale

Institute of Technical Physics, Faculty of Material Science and Applied Chemistry, Riga Technical University, P.Valdena 7, Riga, Latvia

HIGHLIGHTS

- PGRC have higher LOP and MOR values comparing to GRC samples. Added polymer to PGRC samples makes brittle concrete more flexible and prone to deformations.
- For all samples with admixtures and reference samples maximal healed crack width is from 134,7–308,7 µm.
- Healing rate during first 8 weeks after pre-cracking for GRC and PGRC is from 7 till 19 µm per week.
- Main self-heling products of GRC and PGRC with and without crystalline admixtures are $CaCO_3$ and CSH gel.

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ABSTRACT

In this study glass fibre reinforced concrete (GRC) with and without polymer was used as base material to evaluate concrete self-healing enhanced by crystalline admixture. GRC was used because of fibre reinforcement which allow to control width of crack during sample pre-cracking. Properties, like flexural strength, volumetric weight, water absorption and open porosity of GRC and PGRC were evaluated. XRD and FTIR were used to investigate crystalline admixtures produced by different manufacturers. Crystalline admixtures were added 1% from cement. Samples were pre-cracked after 28 days. After pre-cracking samples were fully immersed in water during healing process. Analysis of GRC and PGRC sample self-healing was made. Crack healing rate was monitored for 8 weeks for samples with crystalline admixture, which achieved the best results in crack healing. Results showed difference in self-heling start between GRC and PGRC samples.

1. Introduction

Concrete is most widely used construction material in building industry. Different types of concrete have been developed: water-tight concrete, freeze–thaw resistant concrete, light-weight concrete, self-compacting concrete, architectural concrete, fast/slow setting concrete, cement based composite materials, reinforced (with metallic, polymer, natural or inorganic fibres) concrete. Glass fibres are lightweight, cost-effective and has high tensile strength [1]. Glass fibres in concrete are used from late 1950′s [2]. After first applications were built, it was detected that the GRC properties were strongly affected as time passed. Conclusions of strength retention was that using regular glass fibres in concrete, over time, due to concretes high alkalinity, fibre filament diameter decreased leading to reduced strength of composite material. Loss of diameter is based on Si-O-Si bond brake by OH^- which are present in high amounts in cement matrix [3–5]. At early 1980′s alkali resistant (AR) glass fibre, with ZrO_2, became available commercially and used as reinforcement in concrete [2]. AR glass fibres significantly improved the long-term performance of GRC.

Glass fibre reinforced concrete (GRC) has been used from 1950′s in the construction industry in Europe, especially in facade panels [2,6]. One of the most popular and oldest buildings with GRC façade is the Nanjing Youth Olympic Centre in London, UK. Building was completed in 1974 and covered in total with 110 000 m^2 (12000 psc.) of GRC panels [2]. Use of GRC significantly improves architectural design of concrete building and offers endless possibilities of design and is more cost effective comparing to alternatives. Two basic material, concrete matrix, and glass fibre, excellent interaction ensures remarkable mechanical properties of this material. Fibres improve ductility, tensile and flexural properties, but the stiffness and compressive properties are provided by concrete matrix [7]. GRC elements can be made only 10 mm thick

* Corresponding author.
E-mail address: sandra.guzlena@rtu.lv (S. Guzlena).

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in almost any shape allowing architects and structural engineers to create lightweight elements with incredible facades. GRC can be made with and without acrylic polymer. Acrylic polymer is used to eliminate the required 7 day wet cure to achieve GRC 28 day strength and increases long-term durability [8]. GRC without polymer must be cured at approximately 20 °C and 95% RH. For both polymer and non-polymer GRC, sudden and rapid drying, high temperature changes must be avoided to reach de-moulding strength.

As cracks appear in concrete it can reduce concrete service life. Many techniques to repair are available, but it is time consuming and expensive [9,10]. To prevent maintenance work and expenses concretes self-healing techniques can be used. Self-healing divides in two types autonomic and autogenic. Autogenic self-healing, which happens due to hydration reaction of un-hydrated cement, no special self-healing enhancing admixtures are not used. This phenomenon have mentioned and proposed as self-healing by Beville in 1981 [11]. GRC usually are produced using 0,3 water:cement ratio and high amount of cement [3]. High cement amount and low w:c ratio in GRC is very favorable to autogenic self-heling. Autonomic self-healing happens due to admixture which is not part of the ordinary concrete. Most popular autonomic self-healing techniques are using bacteria [9,12,13], crystalline admixture [14–18], encapsulated healing agents [19], vascular systems [19], and others. Crystalline admixtures are used in this research. To achieve good healing results by applying crystalline admixture as healing substance to concrete, presence of water is very essential, to promote deposition and filling of cracks. As Roig-Flores has concluded concrete samples with and without crystalline admixture did not heal when exposed to moist conditions [15]. In this research self-healing of GRC and first time GRC with polymer (PGRC) was evaluated. Samples with 5 different crystalline admixtures to enhance self-heling and without admixtures was made. Microstructural analysis of the healing products was analyzed.

Table 1
Mix proportions of GRC and PGRC.

	GRC	PGRC
Cement:Sand	1:1	1:1
Water: cement	0,30–0,34	0,30–0,34
Polymer solids Content	-	4–7%
Superplasticizer	Adjusted	Adjusted
AR glass fiber content, by cement weight	5%	5%

2. Material and methods

GRC samples were made using spraying method. The GRC material is sprayed and built up in thin layers until the 10–12 mm. Simple hand rollers are used to compact the material between layers. Basic components of GRC mixture was cement CEM I 52,5R and fine quartz sand. Acrylic polymer was used in polymer GRC (PGRC) samples and added into to the mixture the same time as water.

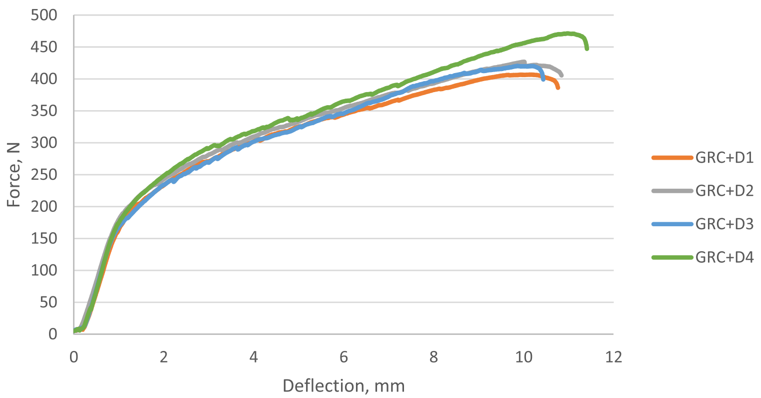


Fig. 1. Four-point bending test to pre-crack GRC + D samples.

Table 2
Analysis of GRC and PGRC sample self-healing.

Sample type	Number of samples	Self-healing - immersed in water visual evaluation	Average crack width, µm	Average healed crack width, µm	Max healed crack width, µm
GRC - Base	8	±	158,7	151,3	292,0
GRC + A	8	±	104,3	134,2	204,3
GRC + B	8	–	105,3	121,7	277,5
GRC + C	8	±	43,7	52,8	134,7
GRC + D	8	–	265,7	204,8	308,7
GRC + E	8	–	163,2	174,4	268,6
PGRC - Base	8	±	176,8	163,5	301,5
PGRC + A	8	±	143,3	151,2	259,4
PGRC + B	8	±	178,7	161,7	295,4
PGRC + C	8	±	199,4	215,3	305,9
PGRC + D	8	–	124,3	118,5	286,4
PGRC + E	8	–	146,9	150,4	247,3

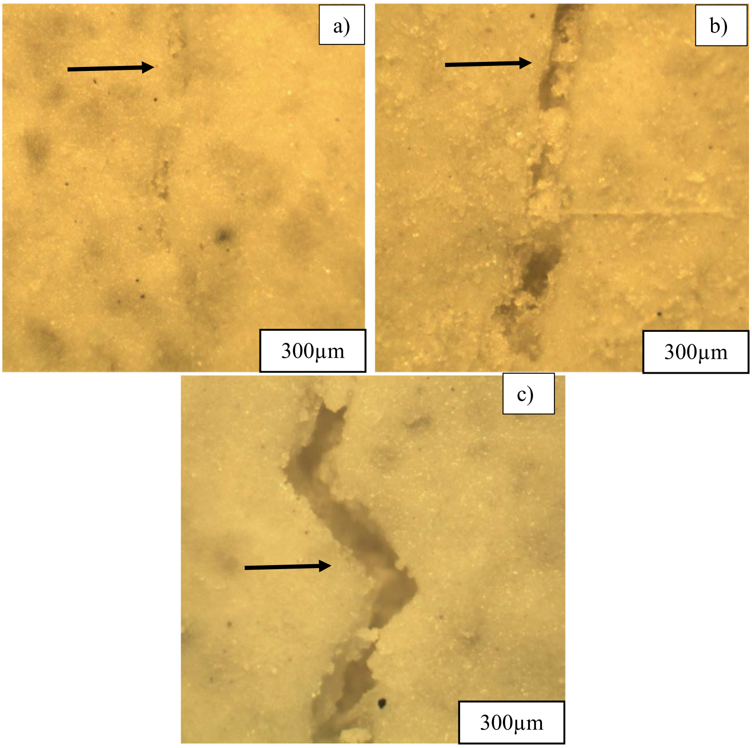


Fig. 2. Crack self-healing steps a) Healed crack; b) partly healed crack; c) unhealed crack.

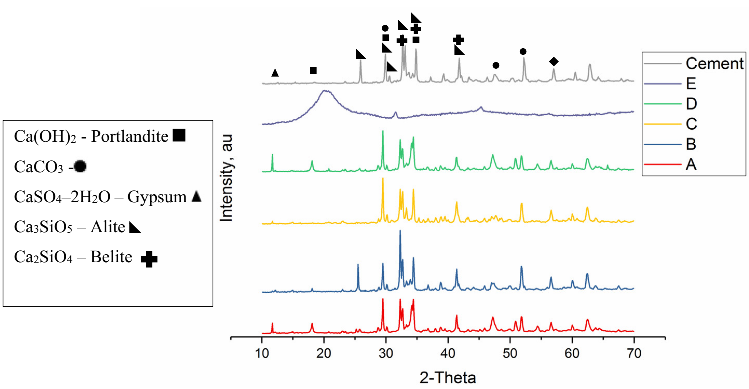


Fig. 3. XRD of crystalline admixtures and white cement.

Flowability and spray ability of mixtures with crystalline admixtures were regulated using superplasticizer and/or water. Mix proportion of GRC and PGRC are represented in Table 1. AR-glass fibre roving, which was cut during spraying process, was used.

Sample flexural test was carried according to EN 1170-5 [20]. Samples were cut from test board in size 275 × 50 mm and tested with 4-point bending test. Level of proportionality (LOP) which presents plastic deformation and modulus of rapture (MOR) which presents samples destructive strength was calculated according to EN 1170–5 [20] and evaluated.

Volumetric weight, porosity and water absorption was measured.

Two types of GRC samples for self-healing have been made with acrylic polymer and without. Samples with five different manufacturers (A; B; C; D; E) crystalline admixtures were made. Crystalline admixtures were investigated using X-ray diffraction (XRD) using Analytical X'PertPro diffractometer with Cu Kα1 radiation and Fourier Transform Infrared (FTIR). Crystalline admixtures were added 1% from cement. Reference sample was made for GRC and PGRC samples. Samples after 28 days were cracked using

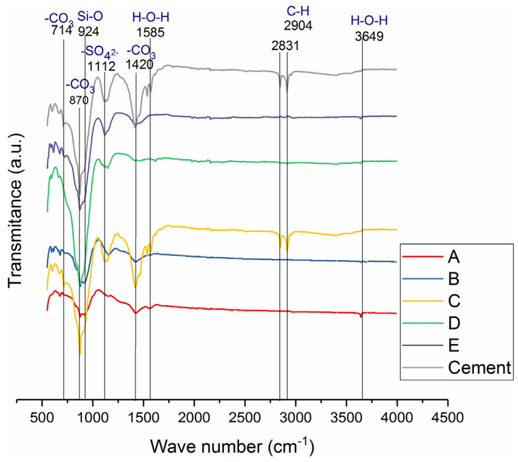


Fig. 4. FTIR of crystalline admixtures and white cement.

four-point bending test measuring applied force and deflection. Test speed 3 mm/min. Test was stopped after sample was cracked – applied force reduced by 5% from maximal force. In Fig. 1 example of tested pre-cracked GRC + D samples. As the concrete is reinforced with glass fibre sample do not rapture as brittle material and crack can be controlled. After pre-cracking crack width was measured using optical microscope. Results are represented in Table 2 in the results section. To initiate self-healing samples were completely immersed in water. Samples investigated after 14 weeks by optical microscope and scanning electron microscope (SEM).

To evaluate self-healing of samples the following rules were made. The state of crack during self-healing can be divided into 3 stages:

- Healed crack (Fig. 2a)
- Partly healed crack (Fig. 2b)
- Unhealed crack (Fig. 2c)

Crack self-healing was evaluated by two factors: sample amount, as with the same recipe pre-cracked sample amount variates and sample self- healing stage, described above.

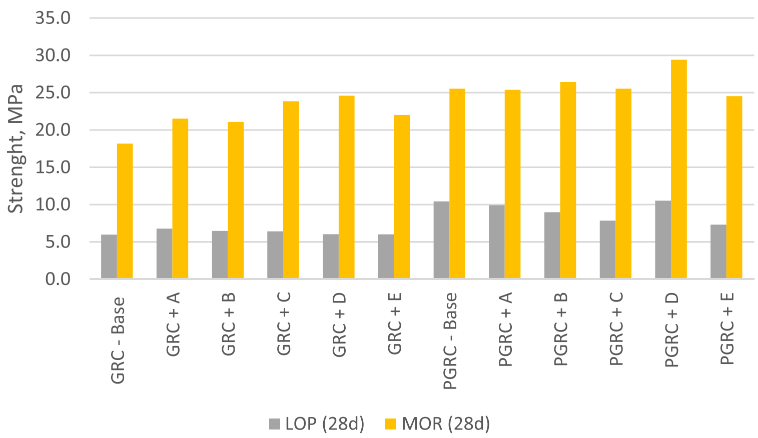


Fig. 5. Flexural strength depending on GRC type and crystalline admixture.

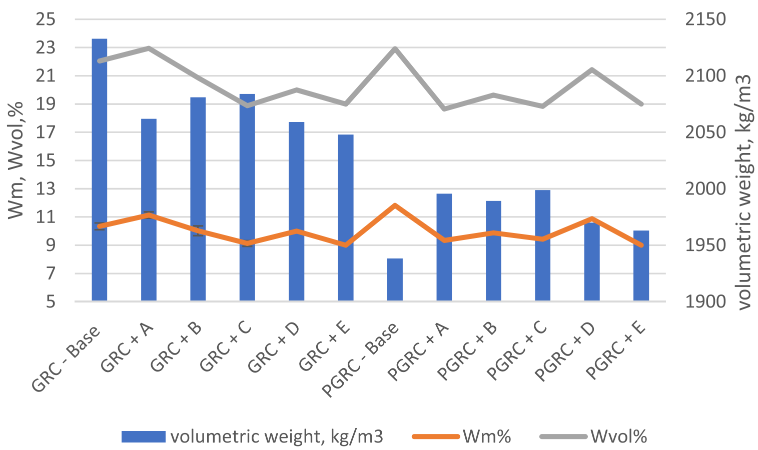


Fig. 6. Volumetric weight, water absorption and open porosity of the GRC and PGRC samples with and without crystalline admixtures.

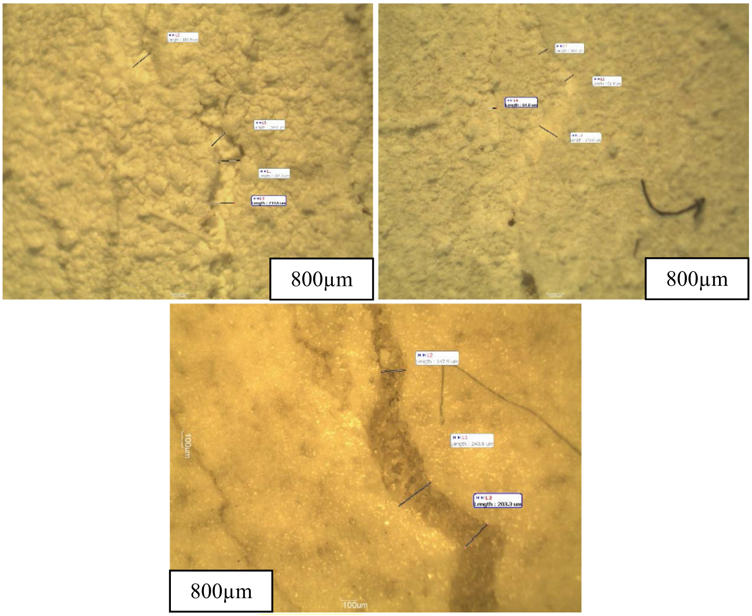


Fig. 7. Examples of healed GRC cracks.

- If < 10% of all samples showed some self-healing it is evaluated as "- "
- If > 90% of all samples have healed self-healing it is evaluated as "+ "
- If 11–89% of all samples showed some self-healing it is evaluated as "± "

Mixes with the best healing results were evaluated to determinate crack healing dynamic. Untested samples from the same batch were pre-cracked as mentioned before. Samples were fully immersed in water and monitored using optical microscope once per week for 8 weeks in total.

3. Results

3.1. Crystalline admixture investigation by XRD

Crystalline admixtures and white cement were investigated using X-ray diffraction (XRD) and patterns are shown in Fig. 3. Comparing white Portland cement with crystalline admixtures most similar pattern have admixture B. Admixtures A and D have almost the same pattern comparing them. These admixtures are only ones who have spikes with gypsum and portlandite 10 and 20 2Θ. For crystalline admixture E was not possible to make XRD measurements – it is x-ray amorphous powder [21].

3.2. FTIR of admixture

The performed FTIR spectra analysis graphs are presented in Fig. 4. Unlike concluded in XRD analysis, FTIR data crystalline admixture C and white Portland cement have the same FTIR spectrums. Admixtures A and D have almost the same pattern comparing them at XRD but looking on FTIR spectres crystalline admixture D have a lot intensive transmittance of $–CO_3$, do not have Si-O bonds and have more intensive $-SO_4^{2-}$ bond stretching [21,22].

3.3. Properties of GRC with crystalline admixtures

Difference between flexural strength values depending on GRC type and admixture are represented in Fig. 5. MOR values of GRC are more influenced by admixtures than LOP values. Admixture addition increase MOR values of GRC samples. PGRC have higher LOP and MOR values comparing to GRC samples. That can be explained by polymer addition which makes brittle concrete more flexible and prone to deformations. It is assumed that the polymer prevents the formation of zones of stress concentrations in the matrix by distributing strains more uniformly.

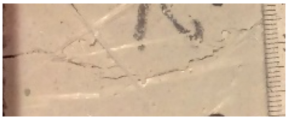
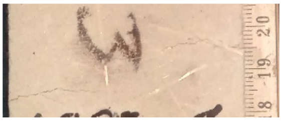
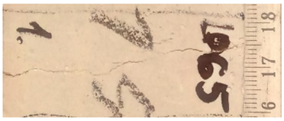
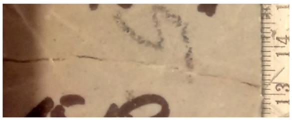
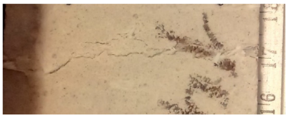
GRC and PGRC sample volumetric weight, water absorption (Wm%) and open porosity (Wvol%) have been tested and results are represented in Fig. 6. Volumetric weight values are higher for all GRC samples comparing to PGRC, but water absorption and open porosity is in the same level of both types of reinforced concrete. Difference in volumetric weight between GRC and PGRC is about 5%, so we can conclude that polymer concrete structure has made lighter, but open porosity and water absorption has stayed the same level as GRC samples. As the GRC is usually used to produce façade materials where one of the important factors is element weight the polymer addition to the mixture positively improves it.

3.4. GRC and PGRC self-healing with and without crystalline admixture

Two types of GRC samples for self-healing have been made with acrylic polymer and without. Samples after 28 days were cracked using four-point bending test. After pre-cracking samples were completely immersed in water. Samples were investigated after 14 weeks. In Table 2 can be seen that PGRC have better self-healing results than GRC samples. Eight samples of each type were evaluated visually, and healed crack width measured with microscope.

Visually evaluating admixtures D and E have poor self-healing results for both GRC and PGRC samples. For all samples with

Table 3
Analysis of GRC and PGRC sample self-healing dynamic.

Sample type	Start of the test	After 8 weeks	Evaluation of crack healing	Average healing rate in week, μm
GRC - Base			±	16
GRC + A			±	12
GRC + C			±	7
PGRC - Base			–	17
PGRC + A			–	15
PGRC + C			–	19

admixtures and reference samples maximum healed crack width is from 134,7–308,7 μm, illustrated in Fig. 7. Closure of crack similar width have noted also other researchers [14,17,23].

3.5. Dynamic of GRC and PGRC self-healing with and without crystalline admixture

Concluding from previous results GRC and PGRC base samples and samples with A and C admixture was evaluated for healing dynamic. Crack healing dynamic have been monitored once per week for 8 weeks. Results are represented in Table 3. GRC samples with and without crystalline admixtures have started to heal during 8-week period. PGRC samples with and without crystalline admixtures signs of self-healing appeared later compared to GRC. PGRC samples start to heal after longer time because crystalline admixture and acrylic polymer is added during mixing process and its crystalline admixture is like "encapsulated" with thin layer of acrylic polymer. As good healing results was visible after 14 weeks it can be concluded that polymer film deteriorates during time, and then crystalline admixture have enough water to grow crystals and heal the crack. Due to differences of heling rate average measured heling rate differs from 7 till 19 μm per week. The crack self-healing rate within the crack is dependent on the crack width.

3.6. SEM images of GRC and PGRC self-healing with and without crystalline admixture

In Table 4 is represented GRC and PGRC sample SEM images with and without crystalline admixtures after 8 weeks of self-healing. All samples have formed $CaCO_3$ in the induced crack [24]. Between $CaCO_3$ crystals can be seen calcium silicate hydrate (CSH) gel formation in samples PGRC - Base and best healed sample GRC + A surface and in crack. CSH gel is main product of Portland cement hydration [9]. CSH gel is primarily responsible for the strength in cement-based materials.

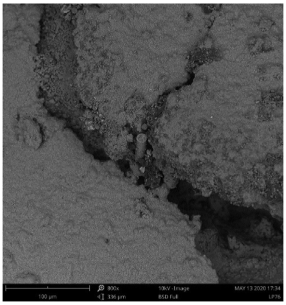
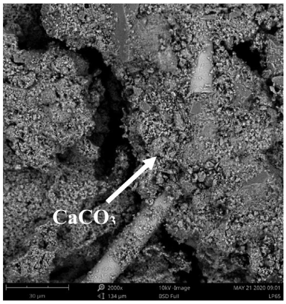
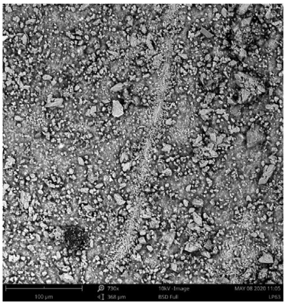
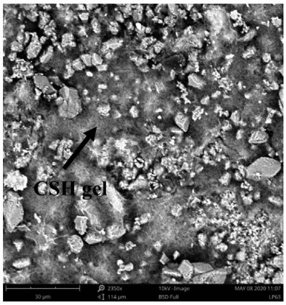
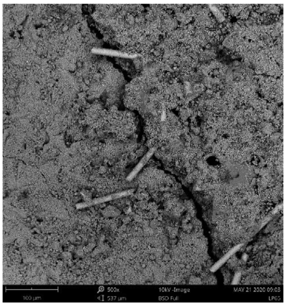
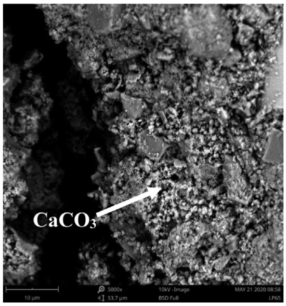
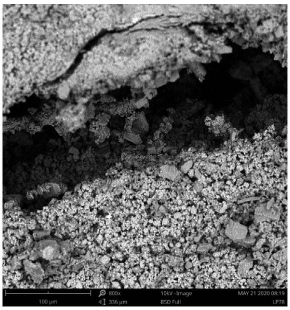
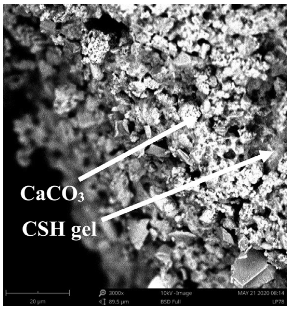
Analyzing XRD, FTIR, SEM data and results gained from sample self-healing in this process mainly participates C–H, Si-O and SO_4^{2-}, because admixtures containing these chemical compounds have the best healing results. As crystalline admixture C and white Portland cement have the same FTIR and XRD patterns the results of self-healing are achieved similar. Interesting that admixtures A and D have similar FTIR and XRD patterns too, but self-healing results strongly differ.

4. Conclusions

GRC and PGRC samples with and without crystalline admixture were made. Samples mechanical properties were evaluated.

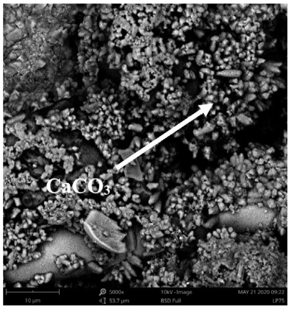
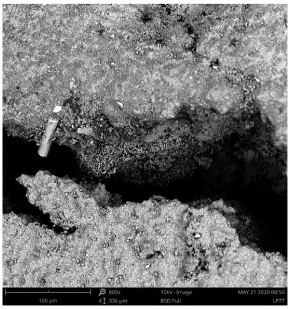
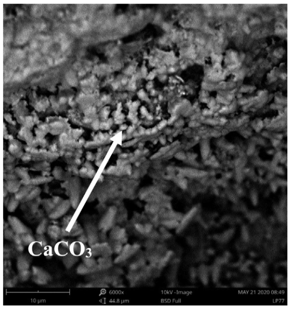
1. PGRC have higher LOP and MOR values comparing to GRC samples. Added polymer to PGRC samples makes brittle concrete more flexible and prone to deformations. It is assumed that the polymer prevents the formation of zones of stress concentrations in the matrix by distributing strains more uniformly.
2. Added polymer also reduces volumetric weight of sample, but open porosity and water absorption comparing to GRC samples are the same.

Table 4
SEM images of GRC and PGRC samples after self-healing.

Sample type		
GRC - Base		
GRC + A		
GRC + C		
PGRC - Base		

(*continued on next page*)

Table 4 (*continued*)

Sample type		
PGRC + A		
PGRC + C		

Samples were pre-cracked after 28 days and then fully immersed in water to evaluate crack self-healing of GRC and PGRC with and without admixtures:

3. Crystalline admixtures D and E have poor self-healing results for both GRC and PGRC samples.
4. For all samples with admixtures and reference samples maximal healed crack width is from 134,7–308,7 μm.
5. After evaluating crack healing dynamic after 8 weeks GRC samples with and without admixture have partly healed, but PGRC samples have poor signs of self-healing. It can be concluded that PGRC samples start to heal after longer period of time because crystalline admixture and acrylic polymer is added during mixing process and its crystalline admixture is like "encapsulated" in thin acrylic polymer film and have no access to water, which is needed for crystal growth.
6. Crack self-healing rate within the crack is dependent on the crack width after pre-cracking. Healing rate during first 8 weeks after pre-cracking for GRC and PGRC is from 7 till 19 μm per week.
7. Main self-heling products of GRC and PGRC with and without crystalline admixtures after 8 weeks are $CaCO_3$ and CSH gel.

CRediT authorship contribution statement

S. Guzlena: Investigation, Writing - original draft. **G. Sakale:** Writing - review & editing, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Sandra Guzlēna was born in 1991 in Jelgava. She obtained a Bachelor's degree in Materials Science (2013) and a Master's degree in Materials Nanotechnology (2015) from Riga Technical University. She worked at the Institute of Technical Physics of Faculty of Materials Science and Applied Chemistry of Riga Technical University, where since 2013, she has been a senior laboratory assistant and from 2016 a scientific assistant. In 2015, she was a ceramic technologist at LODE Ltd. She worked at Skonto Concrete Cladding Ltd where since 2017, she has been a manager of research and development department.