Determination of surface properties of treated cement pastes by acoustic methods and scratch test - pilot experiments

Libor Topolář[1,*], *Lukáš* Kalina[1], *Dalibor* Kocáb[1], *Valeriia* Iliushchenko[1], *Petr* Bílý[2], and *Josef* Fládr[2]

[1]Brno University of Technology, Antonínská 548/1, 602 00 Brno, Czech Republic
[2]Czech Technical University in Prague, Jugoslávských partyzánů 1580/3, 160 00 Prague, Czech Republic

Abstract. The concrete has been and at the same time will be the most used construction material worldwide. It is exposed to various physical and chemical degradation processes that deteriorate its properties and shorten its service life in most applications. As most of the detrimental effects on concrete come from the ambient environment, the quality of concrete surface plays a key role in the overall concrete performance. It should be resistant to abrasion, free of microcracks and open pores to prevent ingress of water, aggressive solutions, and gases. To enhance the properties of the concrete surface, various approaches can be used. The treatment via silicate-based sealers is becoming increasingly popular in concrete technology, especially in preventing deterioration when exposed to highly aggressive environments. This contribution focuses on applying unconventional test methods (e.g. combined scratch/acoustic emission method) to detect the fundamental properties of the treated cementitious surfaces will also provide a new perspective approach of material testing, which may be in the future advantageously used in technical practice. The present combined scratch/acoustic emission test evaluation will provide an excellent insight into lithium silicate sealers' physical behaviour on fine-grained cement-based materials during this test.

1 Introduction

The ever-increasing requirements applied to cementitious composites with optimized physical properties are the main driving force of the current building and material engineering. The surface of a cementitious composite interacts with the environment around it, which may include various physical or chemical degradation factors. These subsequently adversely impact the properties of cementitious materials and therefore shorten their service life. The use of surface layers and coatings is a logical solution to this situation.

The surface of cementitious composites should be resistant to abrasion and free of microcracks and open pores to prevent ingress of water, aggressive solutions and gases. Various approaches can be employed to improve the surface properties of cementitious composites. For example, the standard EN 1504-2 distinguishes between hydrophobic impregnation (a water-repellent surface without the pore-filling effect), impregnation (a partial or complete pore-filling effect) and coating (a continuous protective film on the surface of concrete), nevertheless, multifunctional surface treatments are also known [1].

The pilot experiments described below involve alkali-silicate sealers applied to the surface of cementitious composites. The generally accepted mechanism of action of alkali-silicate sealers or hardeners lies in their reaction with calcium ions in the surface layer of a cementitious composite and a subsequent formation of a calcium-silicate-hydrate (CSH) gel, which blocks the pores and therefore increases the hardness and impermeability of the cement surface [1,2]. The main source of calcium ions is portlandite, yet this probably also applies to other hydration products, such as ettringite monosulfate resp. phase AFm, which may lead to the absorption of Al and the formation of the C-A-S-H gel [3]. In most cases, scholarly publications define sodium silicate as an inexpensive and widely used sealer. Its use for surface treatment is reported to result in a reduction in water absorption or permeability [4-7], a reduction in chloride permeability [5,7,8] a reduction in the carbonation depth of cementitious composites [4,8,9] and at the same time in a significant increase of the abrasion resistance [4,10,11] and a slight improvement of frost resistance[8].

On the other hand, there is only a marginal direct effect on the compressive strength of the cementitious composite since the surface treatment acts only on the surface of the composite and not through its entire volume. However, improved mechanical properties have been found after applications of alkali-silicate sealers onto the surface of a cement sample [5, 12].

One of the methods used for surface layer testing of specimens is nanoindentation. Nanoindentation belongs among basic tests used in thin film metrology to investigate elastic and plastic properties. The test involves penetration of a test sample with a diamond tip of precisely defined dimensions and geometry under load.

Nanoindentation is usually conducted using a so-called Berkovich type indenter, which is a triangular pyramid whose ratio of cross section to indentation depth

* Corresponding author: libor.topolar@vutbr.cz

is the same as in the case of a Vickers indenter [13]. Nanoindentation can provide basic information about the depth profile of hardness and modulus by thickness, consistency of the layer, its cracking or creep.

Another available surface layer test is the scratch test (Fig. 1). Scratch tests are used to examine the cohesive and adhesive properties of thin films and coatings. The principle of the test is based on the movement of a tip under load across the examined surface with a simultaneous and continuous monitoring of the applied force and the respective position of the tip, or the frictional force. The resulting formation on the surface is called a scratch track [14]. The Rockwell conical indenter is the most commonly used indenter for this type of testing, more accurate measurements utilize a so-called Cube-corner indenter, and the load is either constant or linearly increasing. Several gradual types of layer-substrate system defects can be observed during the test, from the formation of microcracks through cohesion defects to complete delamination. The purpose of the scratch test is to identify the limit loads corresponding to the formation of these defects. In general, defects in a layer-substrate system during a scratch test occur as a result of a combination of elastic-plastic indentation stress, friction and internal stress [15,16].

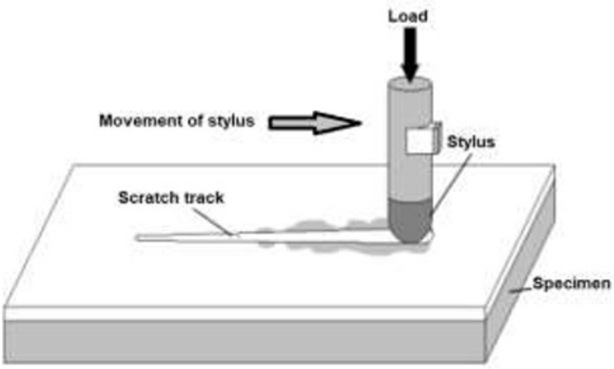

Fig. 1. Schematic representation of a scratch test [14] the stylus in the described experiment was replaced by a needle and a blade.

It is known that shock acoustic waves (usually in the range of 150-300 kHz) can be generated in the areas where the material undergoes abrupt changes in stress or pressure. This phenomenon is known as acoustic emission (AE) and is typically detected by an audio sensor attached to the surface of a given material. The formation and growth of microcracks, interface bond breaking, and delamination of thin films are typical examples of phenomena which produce acoustic emissions. Employment of acoustic wave sensors makes it possible to acquire interesting data regarding the various processes that occur during indentation or scratch testing [17-19].

Another possibility to determine the properties of a surface layer is a modified SASW (Spectral Analysis of Surface Waves) method [20]. The SASW method is a seismic testing method used to determine the velocity of shear waves, shear modulus profiles of soils and Young's modulus of asphalt pavements. The key point of SASW testing is the generation and measurement of surface waves (Rayleigh waves). The source is a vertical impact (or in the described case of the Hsu-Nielsen test – breaking of a pencil lead) on the surface of a sample (Fig. 2). The impact generates a group of Rayleigh waves (R-waves) of various frequencies, which are recorded by two acoustic emission sensors placed on the surface of the sample (Fig. 3).

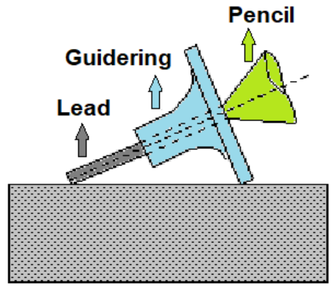

Fig. 2. Hsu-Nielsen Source (pencil lead break). Lead hardness-2H, lead diameter - 0.5 mm, length - (3.0 ± 0.5) mm.

2 Pilot experiment setup

A cement paste (CEM I 42.5R; Českomoravský cement, a.s. – plant Mokrá) was chosen as the test material. The water coefficient (the water to binder weight ratio) for the preparation of the paste was 0.29. Test specimens were produced according to ČSN EN 196-1 and were in the shape of beams with dimensions of 40×40×160 mm. Demoulding of the specimens was conducted 24 hours after mixing and was followed by a standard ageing process lasting 28 days.

The reference sample (REF) was represented by a simple, untreated paste whose surface was ground with a grinding wheel with a grain size of 200 and 80 in order to remove the carbonation products. The treated sample (LIT) was prepared by applying lithium water glass (Lithium water glass; silicate module 3.1; solid content 20.4%; SChem a.s.) onto a sample that had also been ground as in the case of the reference sample. The application of lithium water glass was performed by immersion in a bath for 5 minutes. Excess water glass was removed by wiping with a brush. The specimens were ready for testing after subsequent ageing for 24 hours.

Acoustic non-destructive tests were divided into three separate parts. The first part involved a modified SASW method. The sensors were placed according to Fig. 3 and a pencil lead was broken five times for each sensor including each of the five specimens. Consequently, 50 mechanical wave transit times were recorded. The results have been processed using box plots (Fig. 5).

 https://doi.org/10.1051/matecconf/202236403009

Fig. 3. Location of piezoelectric sensors during testing.

The second part employed a simple scratch test using a needle (Fig. 4 - up) and recording of acoustic emissions using two acoustic emission sensors. Four scratches were made on each of the five specimens. Amplitude was selected as the monitored parameter of the acoustic emission hits. A higher amplitude value indicates higher damage to the monitored specimen or its structure. The results have been processed using box plots (Fig. 6).

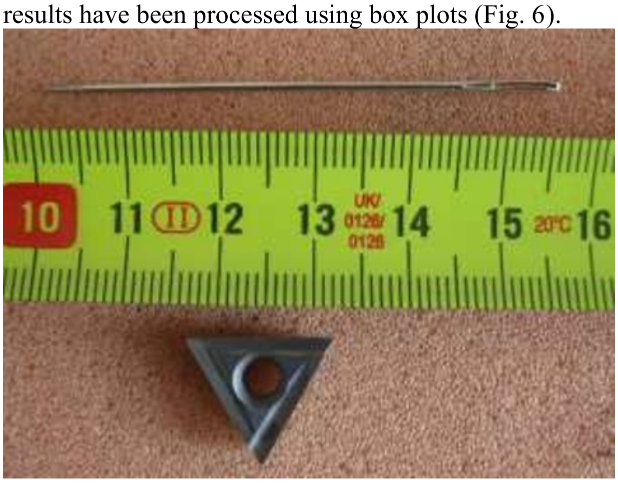

Fig. 4. Scratch test tools used during pilot experiments.

The final third test was very similar to the second one with only the needle being replaced by a metalworking blade (Fig. 4 - down). The results have been processed using box plots (Fig. 7).

Recording of mechanical waves was conducted using a Dakel ZEDO device. The parameters set for recording of mechanical waves were as follows: the total gain was 60 dB (34 dB pre-amplification + 26 dB gain on the recording card), the detection threshold was automatically at 112% above the noise level.

3 Results

The results are presented using box plots, which constitute one of the methods for graphical visualization of numerical data using their quartiles. The middle part of the diagram is bordered on the top by the 3rd quartile, on the bottom by the 1st quartile, and there is a line designating the median between them. The graphs (Fig. 5 - 7) also include a cross designating the arithmetic mean. The box plots also contain lines perpendicular to the middle of the diagram, which express the minimum and maximum values. Outliers are then plotted as individual points.

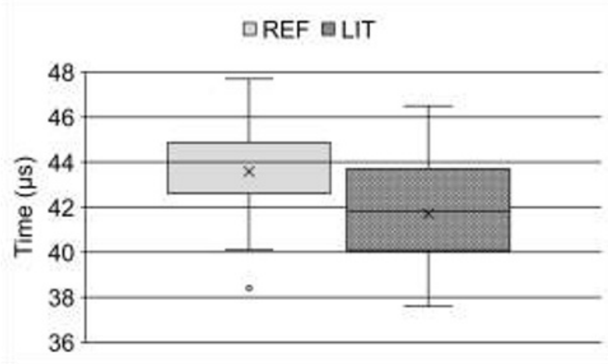

Fig. 5. Surface wave transit time from the first test.

In the case of the results of the first test (a modified SASW method), the graphical representation of the transit times is shown in Fig. 5. The results demonstrate a difference between the untreated reference cement paste and the one treated with the lithium sealer. The transit time is slightly lower in the case of the treated specimens, which indicates higher surface wave velocity (at the same base length). The higher surface wave velocity is a consequence of a more compact structure of the surface treatment. The pores that had formed during the ageing process of the cement paste had probably been sealed too. The results also show a slightly larger variance of values in the case of the treated specimens, which may be due to a not yet fully mastered technique of surface treatment using the lithium sealer.

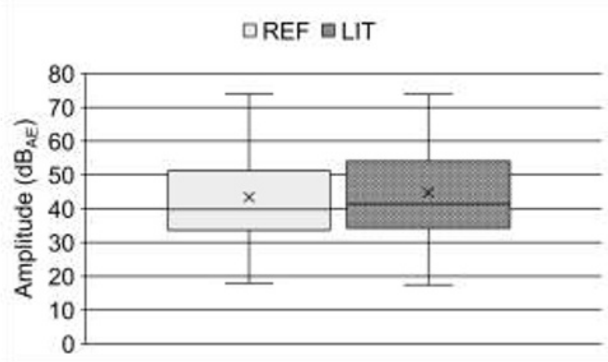

Fig. 6. Size of recorded amplitudes - scratch test needle.

In the case of the results of the second test (scratch test using a needle), the graphical representation of the recorded amplitudes is shown in Fig. 6. In this test, the results are very similar; however, the paste treated with the lithium sealer did have slightly higher amplitude values. The slightly higher amplitude will probably not be associated with higher degree of damage, but rather with increased hardness of the treated surface. (*In the following research, the authors will also focus on a microscopic analysis of the scratch tracks*).

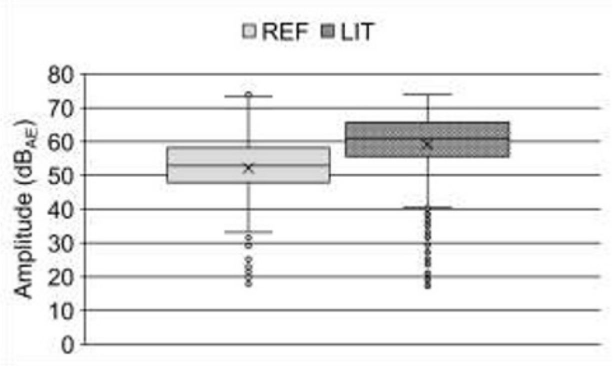

Fig. 7. Size of recorded amplitudes - scratch test blade.

In the case of the results of the third test (scratch test with a blade), the graphical representation of the recorded amplitudes is presented in Fig. 7. In this case, a slightly more significant difference between the individual sets of specimens was observed. The specimens that were surface-treated with the lithium sealer again reach higher amplitude values. Again, this will probably be associated with the harder material in the form of the lithium sealer rather than with greater damage. In this test, the evaluation also found more outlying values of lower amplitudes, which are most likely related to the greater depth of the scratch.

4 Conclusions

Three tests of surface-treated cement pastes were conducted within the pilot measurements of the starting project. All three tests were conducted on two sets of five specimens. One set was for reference (i.e. untreated) and the other set of specimens was surface treated with a lithium sealer. The following experiments were conducted: a modified SASW method and two scratch tests (a needle and a blade). The following preliminary conclusions can be drawn from these experiments:

- surface wave velocity is higher in the case of treated specimens, which is probably caused by better sealing of surface imperfections by the lithium sealer, which then allows mechanical waves to pass more easily across the surface of the material;

- the amplitude of the recorded signals is higher in the case of the surface-treated specimens, which is probably a result of stronger surface layer structure in the form of the lithium sealer.

During the pilot tests, we focused on the mechanical macro damage of the treated samples. The aim was to select a suitable tool for the indentation test and to determine the difference between untreated and treated surfaces by analysing the mechanical waves recorded by piezo-sensors.

The results obtained so far will need to be confirmed by further experiments. The following work within the project will focus on a deeper analysis of the results of the SASW method and a more detailed analysis of the scratch test signals. In the case of the scratch test, attention will also focus on the image and depth analysis of the scratch tracks after testing.

This research was funded by the Czech Science Foundation, GA22-04828S, “Advanced lithium silicate sealers: on the way to sustainable building materials”.

References

1. X. Pan, et al., A review on concrete surface treatment Part I: Types and mechanisms, *Constr. Build. Mater.*, **132**, pp.578-590, (2017)
2. Z. Song, et al., Experimental exploration of the waterproofing mechanism of inorganic sodium silicate-based concrete sealers, *Constr. Build. Mater.*, **104**, pp. 276-283, (2016).
3. J. Kim and R. Kitagaki, Behavior of hydrates in cement paste reacted with silicate-based impregnant, *Cem. Concr. Com.*, **114**, (2020).
4. E. Franzoni, et al., Ethyl silicate for surface protection of concrete: Performance in comparison with other inorganic surface treatments, *Cem. Concr. Com.*, **44**, pp. 69-76, (2013).
5. S.-S. Park, et al., Evaluation of Concrete Durability Performance with Sodium Silicate Impregnants, *Adv. Mater. Sci. Eng.*, **2014**, pp. 1-11, (2014).
6. L. Jiang, et al., The investigation of factors affecting the water impermeability of inorganic sodium silicate-based concrete sealers, *Constr. Build. Mater.*, **93**, pp. 729-736, (2015).
7. M.H.F. Medeiros, et al., Reducing Water and Chloride Penetration Through Silicate Treatments for Concrete as a Mean to Control Corrosion Kinetics, *Int. J. Electrochem. Sci.*, **7**, pp. 9682-9696, (2012).
8. H.Y. Moon, et al., Evaluation of the durability of mortar and concrete applied with inorganic coating material and surface treatment system, *Constr. Build. Mater.*, **21(2)**, pp. 362-369, (2007).
9. M. Ibrahim, et al., Use of Surface Treatment Materials to Improve Concrete Durability, *J. Mater. Civil. Eng.*, **11(1)**, pp. 36-40, (1999).
10. L. Baltazar, et al., Surface skin protection of concrete with silicate-based impregnations: Influence of the substrate roughness and moisture, *Constr. Build. Mater.*, **70**, pp. 191-200, (2014).
11. J.L. Thompson, et al., Characterization of silicate sealers on concrete, *Cem. Concr. Res.*, **27(10)**, pp. 1561-1567, (1997).
12. X. Pan, wt al., A review on surface treatment for concrete – Part 2: Performance, *Constr. Build. Mater.*, **133**, pp. 81-90, (2017).
13. A.C. Fischer-Cripps, A review of analysis methods for sub-micron indentation testing, *Vacuum*, **58(4)**, pp. 569-585, (2000).
14. N.M. Jennett, and S. Owen-Jones, The scratch test: calibration, verification and the use of a certified

reference material, (National Physical Laboratory, 2002).

15. N.X. Randall, et al., The effect of intrinsic parameters on the critical load as measured with the scratch test method, *Surface and coatings technology*, **137**, pp. 146-151, (2001).
16. J. Tomastik, et al., On the importance of combined scratch/acoustic emission test evaluation: SiC and SiCN thin films case study, *Coatings*, **8.5**, pp. 196, (2018).
17. C.U. Grosse, et al., Acoustic Emission Testing: Basics for Research-Applications in Engineering. (Springer Nature, 2021).
18. A.V. Danyuk, et al., Anisotropy of the acoustic emission signal on scratch testing of a single crystal of aluminum, *Letters on Materials*, **9.1**, pp. 130-135, (2019).
19. M. Yahiaoui, et al., Acoustic Emission Characterization of Transgranular Cracks in WC–Co Cemented Carbides During a One-way Scratch, *Tribology Letters*, **69.4**, pp. 1-8, (2021).
20. S.I. Moon, et al., Non-destructive Testing Method Using Surface Acoustic Waves Generated by Inter-digital Transducer in Wall Thinned Pipeline, *15th Asia Pacific Conference for Non-Destructive Testing (APCNDT2017)*, (2017).