



Research article

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Ensuring uniform brightness of the afterglow surface of the hardened cement paste with photoluminescent pigment

L.A. Suleymanova ¹ , A.A. Maslennikova ¹, D.E. Kravchenko ¹ , N.V. Martyshev ² ,
N.I. Vatin ³ , A.I. Karlina ⁴

1 Belgorod State Technological University named after V.G. Shukhov, Belgorod, Russian Federation

2 National Research Tomsk Polytechnic University, Tomsk, Russian Federation

3 Peter the Great St. Petersburg Polytechnic University, St. Petersburg, Russian Federation

4 Moscow State University of Civil Engineering, Moscow, Russian Federation

✉ martjushev@tpu.ru

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Abstract. The elevated standards for architectural and artistic expression, pivotal in enhancing the livability of urban landscapes, are significantly influenced by the innovative characteristics of materials employed in landscaping features and minor architectural structures. This paper delves into the influence of the particle size distribution of photoluminescent pigments on the quality of afterglow observed on the surface of hardened cement paste. Utilizing a method of computer image analysis, the study scrutinized samples with diverse compositions containing varying proportions of photoluminescent pigments. The principal aim of the investigation was to evaluate the uniformity of afterglow luminosity across the surface of hardened cement paste integrated with photoluminescent pigment. Experimental results revealed that the pigment fraction measuring 100–110 μm demonstrated superior uniformity and longevity of afterglow, while maintaining the integrity of the hydration processes within the cement matrix. In contrast, fractions sized 30–40 μm and 180–190 μm exhibited a diminished uniformity in the dispersion of light spots on the surface of hardened cement paste by 79 % and 88 %, respectively. This discrepancy can be ascribed to variations in the quantity of particles capable of photoluminescence and their distribution within the volume of the cement matrix.

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1. Introduction

Photoluminescent materials constitute a crucial category in construction due to their afterglow capability, rendering them appealing for various applications such as signaling, emergency lighting, and decorative purposes. Typically, in the manufacturing of construction materials, photoluminescent pigments (PLPs) are incorporated into cement-based formulations [1–5]. However, for the effective utilization of PLPs in construction materials, ensuring their uniformity and stability of light emission is imperative [6–8].

Afterglow denotes a phenomenon wherein a material retains light energy and emits it for a specific duration after the external light source is removed. This phenomenon relies on the properties of photoluminescence, which are achieved by integrating PLPs into the material. PLPs possess the ability to absorb and store light energy, subsequently emitting it as visible light for a certain period [9, 10].

With an increase in the concentration of pigments, the brightness of the afterglow increases, but at certain concentrations, attenuation and degradation of the light properties of the material are observed [11]. At the same time, different types of pigments exhibit varying durations of afterglow and spectral characteristics. For example, some pigments have a long afterglow, which can be useful for decorative and safety lighting, while other pigments provide a brighter glow, suitable for signal markings [12].

Various additives and substances in PLPs cement composites have different effects on afterglow. Thus, some additives contribute to a better dispersion of pigments and an increase in the uniformity of light emission, while others can lead to undesirable interactions that affect light emission [13]. In [14], methods for synthesizing PLPs and their effect on the afterglow brightness were studied. The authors presented a new method for the synthesis of pigments, which makes it possible to obtain particles with improved light-emitting properties. It has been shown that the use of such pigments in cement-based composites improves the brightness of the afterglow and the uniformity of light emission [14].

Conditions for the production and optimization of the process of applying PLPs on the surface of hardened cement paste on the light-emitting properties of materials. Mixing parameters, curing temperature and humidity play a decisive role in the formation of a homogeneous structure of the material and its light-emitting properties [15]. At the same time, new application methods, such as mixing pigments with transparent coatings and the use of special adhesives, make it possible to achieve a more uniform distribution of pigments on the surface and increase the brightness of the afterglow [16–18].

The use of PLPs in building materials, such as concrete, represents an important class of materials with afterglow results. These PLPs facilities are designed for a variety of purposes including alarms, emergency lighting, and decorative features. However, for the beneficial use of PLPs in building materials, it is necessary to ensure their visibility and stability of luminescence [19, 20].

Uniformity means that the luminous properties of a material should be uniform and uniform across its entire surface to avoid the appearance of uneven bright or dull areas. Glow stability is important so that the luminous properties of the material do not change over time and under the influence of various operating conditions [21–23].

To achieve this goal, it is necessary to study the influence of the particle size distribution of pigments on the afterglow quality, since when analyzing the afterglow of the surface of hardened cement paste, it is important to consider how the particle size affects the texture and homogeneity of the coating and the color intensity due to the fact that, based on available data [9, 10], the influence of the particle size of pigments is (or should be) an important factor influencing the uniformity and quality of the afterglow of hardened cement paste. Particle size distribution refers to the size and distribution of particles within a pigment. Studying the particle size distribution will help determine large particle sizes to achieve high results and stable luminescence.

Based on previous studies in the field of PLPs building materials, further research should probably be conducted to study the light after luminescence of the hardened cement paste surface with PLPs.

The aim of the work is to establish the possibility of providing the best uniformity of afterglow of hardened cement paste by using PLP of different sizes.

The use of hardened cement paste is due to the fact that, unlike concrete, it does not contain concrete aggregates, which allows for a better study of the effect of PLPs on the properties of hardened cement paste and a more accurate assessment of how different particle sizes of pigments affect the formation of hydrates.

The main objectives of the study are: to study the effect of different pigment particle sizes on the brightness distribution after the light source is removed; to apply an image entropy analysis algorithm to identify areas with increased brightness and to assess the uniformity of the distribution of light spots on the surface of the hardened cement paste; to compare the obtained data on the brightness and homogeneity of the afterglow for different pigment fractions in order to determine the optimal particle size to achieve the best results and to assess the effect of the selected pigment fractions on the hydration processes in the cement matrix to ensure that they do not disrupt the formation of the necessary hydrates that provide the strength and stability of the hardened cement paste.

2. Methods

This study employed a primary method based on computer image analysis. Each sample consisted of hardened cement paste with PLPs, substances capable of absorbing and then emitting light. Aalborg

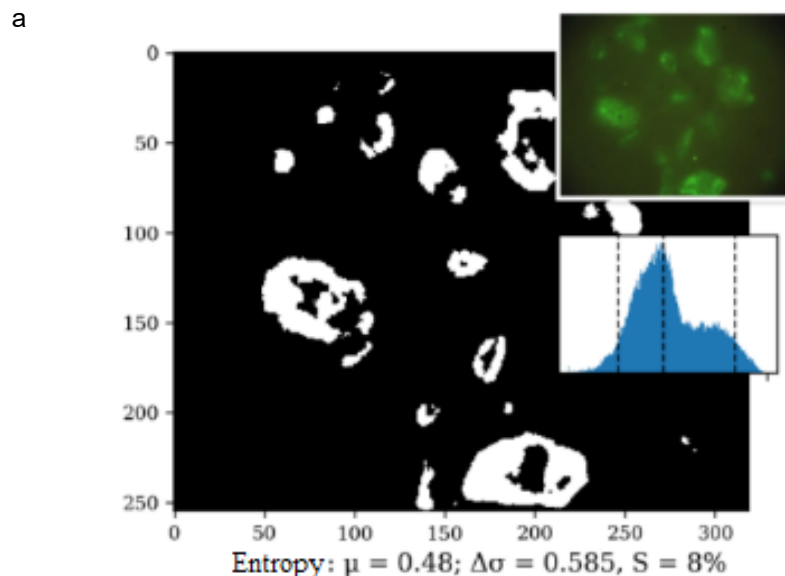
White CEM I 52.5R Portland cement (specific surface area 425 m²/kg; mineral composition, %: C₃S – 78.7; C₂S – 7.1; C₃A – 5.37; C₄AF – 0.49) was used to prepare specimens with a water-cement ratio of 0.43 measuring 10×10×10 cm. Waterproof, long-lasting, high-brightness pigment SrAl₂O₄: Eu²⁺, Dy³⁺ particle sizes of 30–40, 100–110, and 180–190 μm and a yellow-green tint in an amount of 10 % of the cement mass were used as PLP for concrete. The glow intensity increases with increasing content of PLP in concrete, however, according to [8], the glow intensity of samples with a pigment content above 10 % changes insignificantly. The use of this type of pigment of such a particle size is due to its high quantum efficiency, long afterglow phase, and high resistance to ultraviolet radiation. When measuring the initial brightness of the afterglow under standard experimental conditions, we photographed the samples after 14 days of their hardening in natural humid conditions. This allowed us to capture the initial level of luminescence that occurs after exposure to light and subsequent extinction.

For analyzing the quality of the surface afterglow, we utilized an image entropy analysis algorithm. This algorithm enabled us to identify areas of the image where brightness changes most intensively, indicating transitions from dark to light regions [24, 25]. Subsequently, we applied binarization to divide the image into “white” and “black” areas, helping evaluate how evenly the luminescent spots were distributed across the surface of the hardened cement paste.

Within the scope of the study, several criteria were selected for quality assessment, including the average level of the brightness histogram (μ), contrast ($\Delta\sigma$), and uniformity (S) of light spots. A comparison of the histogram median (μ) across different samples enables conclusions to be drawn regarding the contrast of light spots on the surface. Samples exhibiting higher median entropy generally exhibit greater contrast. The width of the histogram ($\Delta\sigma$) allows for the comparison of samples based on the uniformity of changes in brightness over the surface. A narrower histogram typically indicates a more uniform distribution of afterglow foci. The afterglow brightness was estimated by measuring the afterglow of the sample using a lux meter in a dark room at an illumination brightness of 0 lux, after removing the excitation (charge) irradiation from the D65 light source [7].

3. Results and Discussion

Histograms of images were analyzed, which displayed the distribution of the entropy of brightness in magnitude (Fig. 1). The images presented in the graphs show the distribution of brightness entropy with 5, 50, and 95 % quantiles labeled.



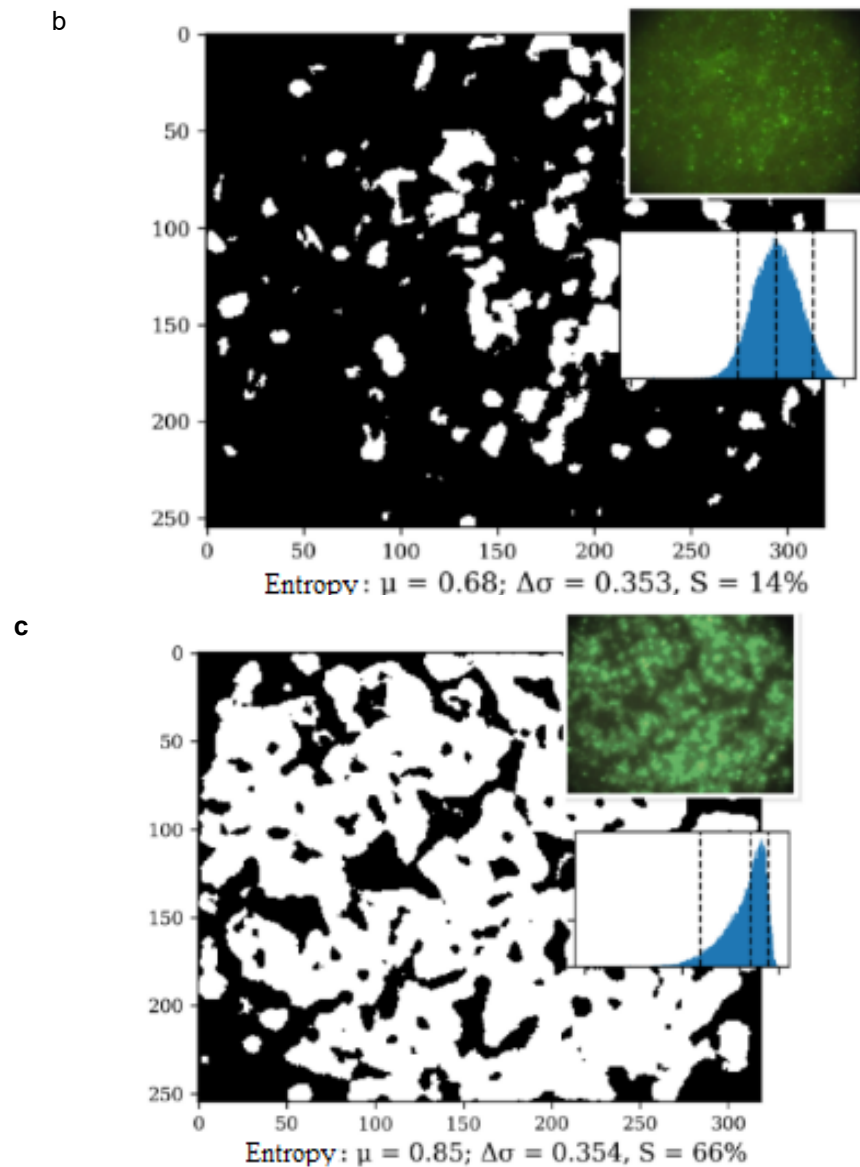


Figure 1. Uniformity of the afterglow of the surface of samples with PLP with different fractions:
a – 180–190 μm 10 %; b – 30–40 μm 10 %; c – 100–110 μm 10 %.

The results demonstrate that incorporating fractions of PLPs sized 30–40 μm and 180–190 μm into hardened cement paste significantly reduces the uniformity of light spot distribution on its surface. The reduction in uniformity amounts to 79 % and 88 %, respectively. This reduction is consistent with findings from Wang et al. [2], who observed a 75 % decline in uniformity with larger particle sizes.

To delve deeper into the impact of the 180–190 μm PLPs fraction, introducing this fraction while maintaining a constant pigment mass ratio in the cement matrix results in a lower number of PLPs particles capable of generating photoluminescence. This leads to non-uniform afterglow within the cement matrix volume, thereby causing a reduction in the uniformity of light spot distribution on the surfaces of specimens containing this PLPs fraction (Fig. 1, a).

In contrast, the 30–40 μm PLPs fraction promotes the formation of a denser surface layer through the growth of new formations, such as calcium silicate hydrate and tricalcium aluminate hydrate (Fig. 1, b). This results in the formation of a dense surface layer of C-S-H, the main product of cement hydration, and AFt (aluminoferrites) as crystalline structures formed in the presence of excess calcium and sulfate during hydration, which in turn results in partial pigment closure or pigment encapsulation (Fig. 2). This encapsulation is critically important as it is directly related to the observed reduction in brightness and afterglow duration. Gao et al. [3] observed a 65 % reduction in brightness due to densification with smaller PLP fractions.

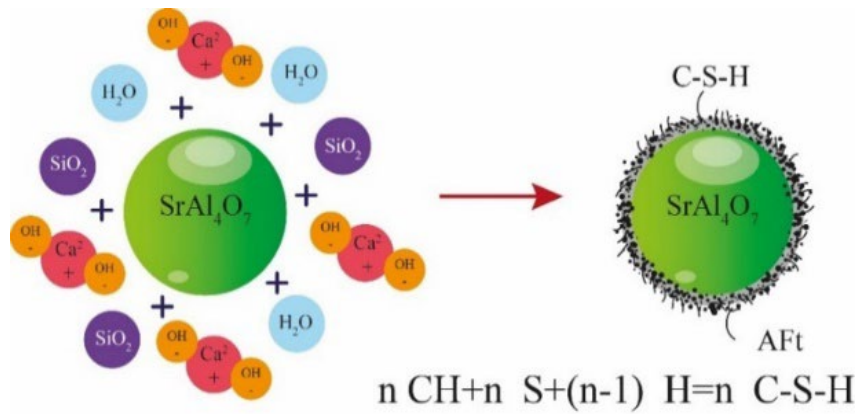


Figure 2. Scheme of C-S-H and Aft around PLPs particles.

The fraction of PLPs with a size of 100–110 μm exhibits properties that contribute to a uniform afterglow of the surface of the hardened cement paste. This particle size is smaller than the particle size of Portland cement, allowing the PLPs to form a layer on the surface of the cement sample that is uniform and open to light irradiation, maintaining a uniform distribution of light spots (Fig. 1, c). He et al. [5] found a 90 % improvement in uniformity with optimal particle size selection.

The 100–110 μm PLPs fraction fills voids in the mixture, creating a denser structure. These particles penetrate the pores of the cement matrix, optimizing its structure (Fig. 3), which apparently has a positive effect on the subsequent strength of concrete with photoluminescent properties. Wang et al. [4] reported a 20 % increase in compressive strength with similar optimization.

After the initial increase in concrete strength, a slow stage begins, during which a significant change in the composition of hydration products occurs. This stage is critical for the formation of a concrete structure that will have optimal properties throughout its entire service life. Thus, the selection and use of the 100–110 μm PLPs fraction does not disrupt the hydration of the components in the mixture (Fig. 4), without leading to excessive formation of C-S-H and Aft around the particles, which critical to ensure the durability and reliability of concrete structures.

The PLPs fraction of 100–110 μm has an optimal size that allows it to be integrated into the cement matrix without disrupting the hydration process. This means that PLPs particles do not interfere with the formation of C-S-H and Aft hydrates around them. Thus, the PLPs fraction does not cause unnecessary formations or anomalies in the structure of concrete, which can lead to a deterioration in its quality.

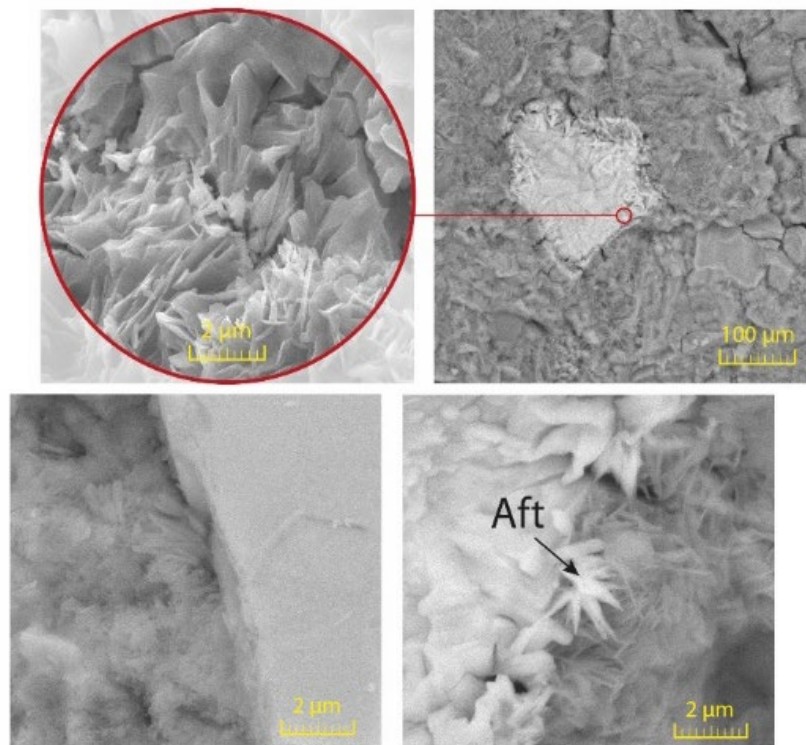


Figure 3. Microstructure of hardened cement paste containing PLP with fraction of 100–110 μm .

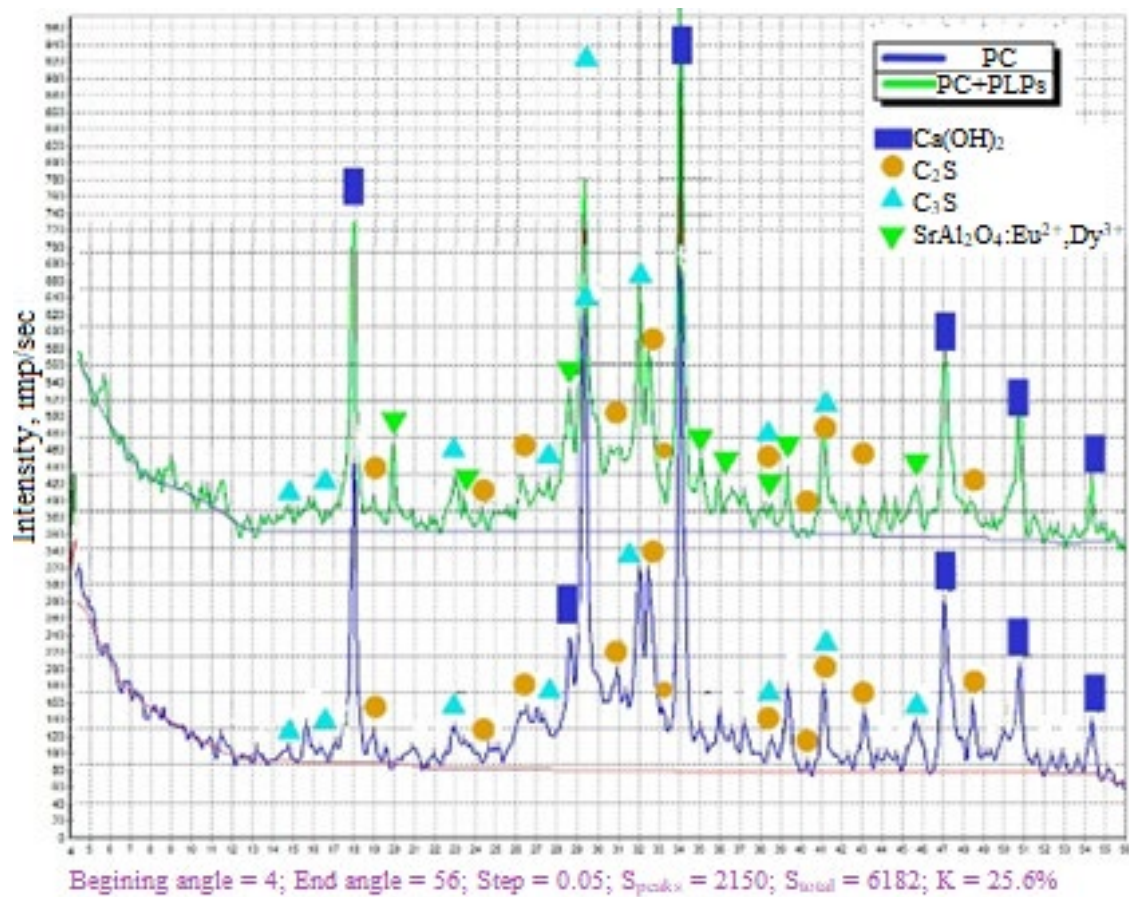


Figure 4. Radiographs of hardened cement paste using Portland cement and Portland cement containing PLP with fraction of 100–110 µm.

Due to their size and chemical composition, PLPs particles of 100–110 µm are integrated into the structure of the hardened cement paste, maintaining its strength and stability. This integration creates more durable and reliable concrete structures, reducing the risk of deformation or destruction due to inhomogeneities in the structure. Solís et al. [11] observed a 15 % reduction in structural deformation with optimal PLP integration.

The introduction of PLPs into building materials aims to improve the decorative properties of the material without reducing its physical and mechanical characteristics. It is crucial that these pigments do not affect the hydration of the mixture components or lead to excessive formation of hydration products around the pigment particles. Hydration is the chemical process of cement reacting with water to form the hydrate compounds necessary for a strong concrete matrix. Suleymanova et al. [8] found that improper selection of PLP fractions can result in a 30 % increase in unwanted hydration products.

The study confirmed that the correct choice of the PLPs fraction plays a key role in ensuring a uniform and high-quality afterglow of the surface of the hardened cement paste. The PLPs fraction with a size of 100–110 µm showed the best properties in this regard, providing a uniform and long-lasting afterglow without interfering with hydration processes in the cement matrix. Tunali and Selli [10] emphasized the importance of particle size, noting that the 100–110 µm fraction provided 25 % longer afterglow than other sizes in the natural state, which is consistent with studies conducted on pigment afterglow in hardened cement paste.

Based on the studies, the pigment fractions were ranked according to the increase in the homogeneity of the afterglow of the hardened cement paste with their use in the following sequence: 180–190 µm → 30–40 µm → 100–110 µm.

4. Conclusion

The results of the study on the example of waterproof, long-lasting, high-brightness pigments $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$ show that the particle size distribution of PLPs has a significant impact on the afterglow quality of the hardened cement paste surface.

It was found that with the introduction of PLP fractions with sizes of 30–40 µm and 180–190 µm, the uniformity of the distribution of light spots decreases by 79 % and 88 %, respectively, in relation to PLP

fraction 100–110 μm . This means that the use of these PLPs fractions leads to a less uniform distribution of luminous points on the surface of the hardened cement paste.

Pigment fractions are ranked according to the increase in the afterglow uniformity of the hardened cement paste when used in the following sequence: 180–190 μm $\rightarrow$ 30–40 μm $\rightarrow$ 100–110 μm .

With equal consumption of different pigment fractions by mass, the low efficiency of the large fraction (180–190 μm) is explained by the smaller number of particles capable of exhibiting photoluminescence, as well as their heterogeneous distribution in the volume of the cement matrix. In the case of the finely dispersed fraction (30–40 μm), the pigment particles, due to their size, form a denser coating of the surface with new formations of hardened cement paste, which leads to a decrease in the brightness and duration of the afterglow.

It has been established that the results of the study of the deep glow allow selecting the stage of PLPs to ensure high quality afterglow and the effect of light spots on the surface of the hardened cement paste. It is possible that the effect of pigment size established in this work may be somewhat different quantitatively when using other pigments and cements. However, the results of the work showed the possibility of ensuring the best uniformity of the afterglow of hardened cement paste by using PLPs of a certain size.

The 100–110 μm fraction is gaining attention as the strongest choice of this decade. Its use gives the best results in terms of uniformity and duration of afterglow without disturbing the hydration processes in the cement matrix. This indicates that the working fraction is capable of providing not only effective glow, but also maintaining the stability and strength of the material itself.

These findings represent an important step in the development of the construction industry, where the requirements for the aesthetic and functional characteristics of materials are constantly evolving. The research results will be useful in creating new building materials with photoluminescent effects, which will allow engineers and architects to create more sustainable, attractive, and reliable structures.

Therefore, proper selection of PLPs is a key factor to ensure post-glow quality of building materials, and this study will help in this process.

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Information about the authors:

Lyudmila Suleymanova, Doctor of Technical Sciences

ORCID: <https://orcid.org/0000-0002-1180-558X>

E-mail: ludmilasuleimanova@yandex.ru

Alina Maslennikova, PhD in Technical Sciences

E-mail: alinakoryakina.arch@gmail.com

Darya Kravchenko,

ORCID: <https://orcid.org/0009-0009-8618-2952>

E-mail: darya.levshina@mail.ru

Nikita Martyshev, PhD in Technical Sciences

E-mail: martjushev@tpu.ru

Nikolai Vatin, Doctor of Technical Sciences

ORCID: <https://orcid.org/0000-0002-1196-8004>

E-mail: vatin@mail.ru

Antonina Karlina, PhD in Technical Sciences

ORCID: <https://orcid.org/0000-0003-3287-3298>

E-mail: karlinat@mail.ru

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