The physical-mechanical influence and chemical effect of calcium formate, vinyl acetate/versatate, and vinyl-acrylate copolymer on the processes of solidification, hardening, and structure formation of the cement matrix were studied during the research reported here. The compositions of mixtures containing additives in the amount of 1, 3, and 5 wt % were investigated. Noteworthy is the water-holding nature of vinyl acetate/versatate and vinyl-acrylate copolymer. With the introduction of appropriate additives, an increase in working time of the mixtures is noticed, which was manifested in prolonging the end time of solidification duration. Calcium formate reduced the end time of solidification duration with an increase in the content of the additive. During 28 days of hardening, a decrease in strength gain was observed with an increase in the content of the additive compared to control samples. At 1 wt % of the additive, the decline in strength on day 28 was 8.7 % for calcium formate, 13 % for versatate, and 15.5 % for vinyl-acrylate copolymer. For versatate and vinyl acrylate with the addition of 3 and 5 wt % in the mixture, the loss of strength is 23–25 % and 27–36.7 %, respectively. 5 wt % calcium formate admixture, compared to 3 wt % on day 7 and day 28 of hardening has a higher strength index. This nature of the effect of additives is explained by the formation of polymer structures throughout the volume of the mixture with the introduction of versatate and copolymer vinyl acrylate, as well as their chemical interaction with the components of the cement binder during hydration in the formed alkaline medium. Calcium formate plays the role of both a filler and a hardening accelerator due to the introduction of an additional amount of calcium ions.

The reported results can be used as a basis for continuing to study the effect of redispersed additives on the durability of cement articles, the development of new formulations for building mixtures, and their potential use in the production of concrete.

Keywords: cement, calcium formate, vinyl acrylate, vinyl acetate/versatate, redispersed polymer, compressive strength

1. Introduction

The need for the use of building mixtures is growing every year in the same way as the need for additives to them. This aspect is associated with the variety of applications of dry mixes in construction. Over the past decade, the use of redispersed powders of various origins has also spread to achieve the necessary physical and mechanical properties of the mobile mixture. In general, such characteristics as water consumption, mobility, solidification duration, water retention, and strength are modified [1].

The variety of additives produced for the construction industry is not limited to one type. Every 1–2 years, new products appear in the additive market with new guaranteed characteristics for modifying the properties of mixtures. This indicates that the production part in the field of building materials is constantly evolving and, thus, new products can have a different effect on the physical, mechanical, and chemical properties of building mixtures. Given this trend in the production of additives, studying the nature of their impact on the properties of dry building mixtures is a relevant task. Also appropriate is the issue of chemical activity of additives in the alkaline medium of the cement matrix which takes place during the hydration of cement minerals.

2. Literature review and problem statement

The use of redispersed powders is directly related to their effect on the strength gain of the silicate composite matrix and rendering them additional characteristics of the finished product. Hydrophobicity, water retention, solidification duration, frost resistance, etc. – this is a list of those characteristics that can be changed in a standard mixture of cement.
binder or other composite binder when administered with redispersed additives. To a greater extent, these additives are both inorganic and organic.

For the most part, in the production of dry building mixtures such as plasters, adhesive mixtures, self-leveling floors, the use of additives is the main factor in creating these types of mixtures. According to research into the market of building additives [2, 3], the use of vinyl copolymer esters and calcium formate is widespread in such products. In studies under [4, 5], comparisons involved various redispersed vinyl copolymer powders. In most contemporary papers [6–14], the authors identified three main additives: vinyl acetate/versatate copolymers (VA/VeoVa) and vinyl acrylic (VA Acrylic), as well as calcium formate powder (FormCa).

Vinyl acetate/versatate (VA/VeoVa) copolymer powders can exhibit a number of separate effects on the cement mixture. Thus, these redesigned powders are generally used as additives to improve adhesion but, in work [4], it was found that this polymer in cement mixtures also affects the water-demand of mixtures and water-holding performance. At the same time, the higher the value of the polymer-cement ratio, the higher the values of water content and water needs, as noted in [7, 8].

The authors of [9, 10] mainly paid attention to the general characteristics of the effects of additives aimed more at affecting the mechanical characteristics, while only superficially describing the chemical effect of additives on the properties of mixtures.

Vinyl acrylic copolymer (VA Acrylic), in turn, exhibits similar characteristics of the effect on cement mixtures as vinyl acetate/versatate copolymers. Vinyl-acrylic copolymer is examined in papers [8, 11], which describe whether the polymer is similar to the copolymer vinyl acetate/versatate in its effect on cement mixtures.

Calcium formate (FormicateCa) is a low-molecular compound of organic origin, namely a salt of formic acid Ca(HCOO)₂. It is an additive that improves the adhesion of cement to surfaces and has the properties of an accelerator of hardening and can also be used as an anti-freeze additive due to its ability to reduce water needs. During hydration processes in the cement matrix, calcium formate affects solidification duration by forming compounds of mono- and tri-hydrate sulphoaluminates. Due to the high dissociation of the compound, the accelerated formation of Ca(OH)₂ occurs while its HCOO⁻ ion is able to penetrate the hydrated layers of alite (C₃A) and belite (C₂A) where it covers their surface [11, 12]. These results are supported by the research reported in [13].

The use of additives in most cases is associated with high efficiency, which is manifested in the effects achieved in the cement matrix when using a small number of additives, this is noted in all works. However, the achievement of these effects, in general, affects the resulting strength of products, as described in studies [9, 10]. According to [10], polymer additives are capable of forming lamellar and needle structures in the thickness of the cement matrix over time. Such formations can be taken as nano-reinforcing components. At the same time, none of the cited works explains the causes of these formations and their potential impact in the long term. The similarity of the nature of the effects of vinyl-acetate/versatate and vinyl-acrylate on the cement matrix is confirmed in [14]. In the case of calcium formate, there is the formation of dense structures, which is noted in works [12, 13], so their effect on strength is greater.

It is worth noting that no work indicates what happens to the additives themselves in the structure of cement stone over a long period. In [14], it is noted that when exposed to an alkaline medium, which is predominant in the processes of structuring cement stone, the dissolution of redispersed additives is impossible, referring to the formed polymer structures. At the same time, in [15], where the reactions of interaction of copolymers of versatate and acrylic were explored directly in an alkaline medium, the course of reactions is described.

In works [8, 13, 14], where there is a question regarding the effects of redispersed additives, the attention was focused on the most common additives in the local market and their actual impact on the mechanical and kinetic properties of cement mixtures. It is worth noting that in different countries the production methods and product quality are regulated differently. This, in general, can affect the end result of the tests.

Thus, the study of this issue is quite expedient not only because of the different approach to the production of additives but also their nature of the impact on the properties of the cement matrix. At the same time, it is important to confirm or refute the chemical interaction of additives with cement components, which can only be obtained through the study of the nature of their action not only on mechanical properties but also on the structure formation of the cement matrix.

3. The aim and objectives of the study

The aim of this work is to identify the nature of the action of the three most common in the production of cement building mixtures of redispersed powders on physical and mechanical properties. The data to be obtained will make it possible to conclude about not only the physical and mechanical nature of the effect on the cement matrix but also confirm or deny the presence of chemical transformations. The would-be results can be applied in practice in the production process to create new formulations of building mixtures, as well as in the production of new concrete compositions.

To accomplish the aim, the following tasks have been set:
- to investigate the effect of the amount of additive on the water-holding capacity of the cement mixture;
- to investigate the effect of the amount of additive on the timing of solidification and the strength indicators of the cement mixture;
- to analyze the chemical interaction of additives with cement particles in a cement matrix in an alkaline medium based on the obtained data and photographs of structure formation.

4. The study materials and methods

4.1. Examined materials and equipment used in experiments

The studies were conducted using standardized cement of the PC I 500N brand. The object of the study was cement mixtures with redispersed additives, namely: copolymers of vinyl acetate/versatate (NeoLith 4400) and vinyl-acrylic (NeoLith 6700). Calcium formate powder (FormCa) was chosen because of its widespread use in dry building mixtures together with redispersed polymers. Due to the method of production of
this low-molecular salt, namely, dispersion of the formed solid mass of calcium formate after filtration and evaporation, some manufacturers label this additive as a redispersing powder.

In determining the compressive strength indicators, the procedure given in DSTU EN 196-1: 2007 (EN 196-1: 2005, IDT) was used. Cement test methods. Part 1. Determining strength. Given that the values first grow rapidly, and then gradually slow down, the test results were processed by logarithmic approximation.

The radio electron microscope (REM 106-I) was used to acquire the results of the interaction of additives on the structure formation of the cement matrix.

4.2. Procedure for determining the indicators of the properties of samples

When conducting these studies, the following main indicators of cement with additives were determined: normal density, solidification timing, compressive strength at the age of 2, 7, and 28 days.

Normal density and solidification timing were determined by the standard procedure on the Vick device in a slurry of normal consistency.

The compressive strength of the samples was determined on samples of small sizes according to the existing procedure given by NIICement. This procedure does not contradict the existing methods for determining compressive strength and is adapted on the basis of the Kind method, where the strength was determined on samples the size of 2×2×2 cm. In the international standard EN 196-1, there is a reference to this procedure, which describes the main aspects of sample preparation for testing.

The concentrations of additives (in wt.%) were selected in accordance with the recommendations of the technical specifications for products provided by the manufacturer. In addition, to increase the correlation of results, a concentration of additives (oversaturation) was chosen to determine the effect of the volume of additives above the norm specified by the manufacturer.

To acquire photographs of the sample structure, after determining the compressive strength, pieces with the least visual damage with viewing surface dimensions of 2×3 mm were selected. The samples were pre-dried at a temperature of 90–110 °C for 4 hours to eliminate moisture, and stored in silica gel desiccators to prevent moisture from entering the samples from the external environment.

5. Results of studying the effect of redispersing additives on the rheological properties of cement

5.1. The effect of the amount of additive on the water-holding capacity of the cement mixture

By the nature of the effect of redispersing additives on the cement mixture, it was found that the additives have a different nature of effect on both normal density and water retention. The results, given in Table 1, indicate a low ability of additives to affect the normal density of cement slurry, as opposed to water retention.

A relatively small change in normal density (0.5–1.5%) indicates a low effect of additives on the mobility of the cement mixture. This indicator suggests that at low concentrations of additives, the water consumption of mixtures as a whole will remain unchanged. This conclusion is confirmed by the values of water retention of cement mixtures when using calcium formate and versatate.

Table 1

| No. | Material         | Cement % | Additive volume % | Normal density % | Water retention % |
|-----|-----------------|----------|-------------------|-----------------|-------------------|
| 1   | Cement          | 100      | 0                 | 27.5            | 93.35             |
| 2   | FormCa          | 99       | 1                 | 28              | 95.61             |
| 3   | Neolith 4400    | 97       | 3                 | 28              | 98.48             |
|     | Neolith 4400    | 95       | 5                 | 98.75           |                    |
| 4   | Neolith 6700    | 99       | 1                 | 28.5            | 96.52             |
|     | Neolith 6700    | 97       | 2                 | 29              | 95.2              |
|     | Neolith 6700    | 95       | 3                 | 30              | 94.6              |

However, vinyl-acrylic copolymer (Neolith 6700), despite the similar effect on the normal density and mobility of the mixture, on the contrary, reduces the rate of water retention. Fig. 1 shows the effect on the water-holding capacity of mixtures.

![Fig. 1. Effect of redispersing powders on the water-holding capacity of cement mixtures](image)

However, despite the relatively low rate of water retention of the mixture without additives (water retention of the cement mixture is 93.35%), it would be incorrect to say that vinyl-acrylic copolymer impairs water retention. Since at a low concentration (1%) the admixture improves this indicator even better than calcium formate and versatate.

The decrease in water retention at higher concentrations of the Neolith 6700 additive can be attributed to its greater chemical activity, as opposed to the more stable versatate and calcium formate. Thus, with an increase in the concentration of copolymer, the proportion of the additive increases, which reacts faster with water, forming polymer structures in the thickness of the cement stone. Thus, covering the particles of cement, slows down the hydration of cement minerals.
5.2. The effect of the amount of additive on the solidification timing of the cement mixture and the acquisition of strength

It has been established that the effect of additives greatly affects the solidification timing of mixtures. With minor changes in normal density, the solidification duration times are radically different from each other depending on the concentration of additives. For versatate and vinyl acrylic, there is no typical, linear increase in the beginning and end of the terms of solidification. This atypicality is confirmed by the results given in Table 2, where one can see a sharp increase in the beginning and end of the terms of solidification at a concentration of 3 wt% additives in the cement mixture.

Table 2

| Material    | Cement volume wt% | FormiateCa        | Neolith 4400 (VA/VeoVa) | Neolith 6700 (VA Acrylic) |
|-------------|-------------------|-------------------|------------------------|--------------------------|
| Normal density  | Solidification duration |
| start | end  | start | end |
| wt% | wt% | % | h-min | h-min |
| Material   | 100  | 0   | 27.5 | 1-10 | 2-15 |
| FormiateCa | 99   | 1   | 28   | 1-03 | 2-30 |
|           | 97   | 3   | 28   | 1-33 | 2-10 |
|           | 95   | 5   | 28.5 | 0-52 | 1-37 |
| Neolith 4400 | 99   | 1   | 27.5 | 1-30 | 2-35 |
|           | 97   | 3   | 28   | 1-03 | 3-33 |
|           | 95   | 5   | 28   | 1-30 | 3-10 |
| Neolith 6700 | 99   | 1   | 28.5 | 1-16 | 2-15 |
|           | 97   | 3   | 29   | 3-29 | 5-14 |
|           | 95   | 5   | 3    | 1-58 | 4-00 |

Special attention should be paid to the indicators of the solidification duration at a concentration of additives of 3 wt%. Increasing the solidification duration for cement mixtures using copolymers is a characteristic mechanism for hardening solutions. Due to the formation of polymer masses in the structure of cement stone, hydration processes slow down accordingly. These formations isolate cement particles, thereby preventing the full course of hydration. Which, in general, increases working time of the mixtures. However, with a further increase in the concentration of additives, one can see a corresponding decrease in working time (the end of the terms of solidification).

This decrease is explained by the fact that with an increase in the concentration of copolymer, due to their relatively high activity before interacting with water, they instantly form coagulation structures. These formations when mixing cement mortar are capable of uneven distribution throughout the entire volume of the mixture. Thereby forming zones with a low concentration of the additive and areas with a high concentration of the additive.

Because of this, such a change in the timing of the beginning and end of solidification is significant for copolymers.

In the case of calcium formate, this change is not radical. Accepting that calcium formate is a derivative of formic acid, when dissociated with water, it partially returns to its original form. Thus, two components are formed in the cement matrix: formic acid and calcium hydroxide. Thus, the redispersed additive acts not only as a water-retaining agent but also as a reinforcing and accelerating hardening agent, which can be traced in Table 2.

By the nature of the effect on the acquisition of grade strength, additives have a similar character as the acquisition of strength by the control mixture (Fig. 2–4).
By the nature of the action, Neolith 4400 and 6700 are similar to typical polymer additives, which, with increasing concentration, increase the solidification duration due to the water-retaining ability of the formed coagulation structures in the cement matrix. This can be seen at concentrations of 1 and 3 wt %. However, with an increase in the amount of the additive (5 wt. %), the start and end time of the period of solidification, in general, decreases compared to the concentration of 3 wt %.

5.3. The effect of additives in the cement matrix and their chemical interaction during hardening

The effect of calcium formate on the terms of solidification in cement mixtures is explained by the fact that during hydration the compound is partially dissociated into formic acid (HCOOH) and calcium hydroxide (Ca(OH)$_2$). Over time, formic acid in the alkaline cement medium forms other formative salts with cement components, namely reacting with CaCO$_3$ residues, reducing to calcium formate or in the presence of potassium oxides according to the following mechanism:

$$2\text{HCOOH} + \text{CaCO}_3 = (\text{HCOO})_2\text{Ca} + \text{H}_2\text{O} + \text{CO}_2, \quad (1)$$

$$(\text{HCOO})_2\text{Ca} + 2\text{H}_2\text{O} = 2\text{HCOOH} + \text{Ca(OH)}_2, \quad (2)$$

$$\text{K}_2\text{O} + \text{H}_2\text{O} = 2\text{KOH}, \quad (3)$$

$$\text{HCOOH} + \text{KOH} = \text{HCOOK} + \text{H}_2\text{O}. \quad (4)$$

Over time, the resulting formic acid under the action of the formed compounds and in the presence of water-negative agents decomposes into water (H$_2$O) and carbon monoxide (CO). In turn, carbon monoxide with atomic water is converted into carbon dioxide (CO$_2$), which already negatively affects the integrity of cement stone by activating carbon-oxygen corrosion, while the resulting new formats form crystalline structures, or partially destroyed.

The active formation of calcium hydroxide in the initial stages of hardening leads to an acceleration of the timing of the onset of solidification, which affects the working time of building mixtures. Fig. 5 shows the structure of cement stone when applying calcium formate admixture.

Fig. 5 clearly shows the amount of Ca(OH)$_2$ formed and the size of their crystals in the early (7 days) periods (red circles), and the proportion of non-reacted calcium formate (blue circles).

The decrease in strength and increase in the working time for copolymers is explained by the fact that the polymer structure formed in the cement matrix acquires stability and hydrophobicity due to the versatate group for samples with the addition of Neolith 4400. Since the versatate molecule itself is hydrophobic and larger in size than acetate (Fig. 6), it is the main factor in the manifestation of hydrophobicity.

Copolymer additives are activated in the presence of moisture by creating polymer chains, which, as already indicated, cover particles of cement, thereby reducing its activity and intensive hydration of calcium oxide (Fig. 7).

However, the vinyl-acrylate copolymer Neolith 6700 (VA Acrylic), which does not have such a hydrophobic group as versatate in its structure, is two separate copolymers – Vinyl Acetate and n-Butyl Acrylate. These two copolymers, which are bound by a soluble colloid, when interacting with water in an alkaline medium of the cement matrix, form particles of polyvinyl alcohol (Fig. 8).
Alkaline medium of cement mixtures. Confirmed by chemical transformations of additives in an ensuing hydration period. Cement hydration is associated with water retention values for redispersed powders are caused. At the same time, the rehydration and changes in the physical and mechanical characteristics are due to the presence of coagulation formations of the copolymers Neolith 6700 in the cement medium.

This reaction occurs due to the fact that, unlike acrylate, acetate is more active during hydrolysis, provided that it is not protected by a hydrophobic radical as is the case with versatate (Neolith 4400). At the same time, the formed calcium acetate and polyvinyl alcohol, in turn, are characterizing reaction products that confirm the decline in the strength of the mixtures.

Calcium acetate has characteristics similar to formate. Therefore, it, like calcium formate, eventually dissociates due to the presence of excess water.

Polyvinyl alcohol also acts as a coagulant which, in the same way as the versatate group, gives hydrophobicity to the mixture due to the presence of coagulation formations (Fig. 9) on the surface of the cement while increasing its working time (terms of solidification).

That is why the decline in strength when using additives and changes in the physical and mechanical characteristics of cement mixtures are caused. At the same time, the resulting water retention values for redispersed powders are confirmed by chemical transformations of additives in an alkaline medium of cement mixtures.

Our results of the nature of the effect of redispersed additives on the cement matrix indicate their water-holding nature, which corresponds to the obtained indicators (Table 1, Fig. 1). At the same time, calcium formate and vinyl acetate/versatate have a more pronounced characteristic, with an increase in their content in mixtures than vinyl acrylate. This effect is explained by the fact that the resulting polyvinyl alcohol (Fig. 8) is more hydrophobic than versatate (Fig. 7), which is why, with an increase in the concentration of the Neolith 6700 additive, its water-holding capacity decreases.

At the same time, the nature of the progress of chemical reactions (1) to (4), as well as the nature of the hydrophobicity of materials (Fig. 6, 8), explain the changes in the solidification timing of the cement matrix (Table 2) during hydration. For calcium formate, this is the course of chemical reactions (1) to (4). These changes, in turn, are supported by indicators of changes in the acquisition of strength (Fig. 2–4), which are already explained by the nature of the structure-building of the cement matrix using chemical additives (Fig. 5, 7, 9).

The acquisition of strength by the samples occurs gradually and its character can be fitted to the logarithmic equation (Fig. 2–4). At the same time, the indicator of reliability of data approximation is close to 1 at certain concentrations.

In fact, the introduction of additives of organic origin, to a certain extent, reduces the grade strength of cement on day 28 of hardening. For concentrations of redispersed additives of 1 wt. %, such a decrease is 9–15 %, while the strength value for mixtures remains within the permissible limits for cement mixtures of grade 500 (42.5–50 MPa). Instead, the concentrations of 3–5 wt. % reduce the strength of mixtures to values of the grade 400.

Characteristic curves indicate that with further hardening of samples, the strength of the material will increase. This, in turn, is confirmed by the water-holding effect of additives.

Our verification of the results indicates that for the addition of vinyl acetate/versatate (Neolith 4400) at a concentration of 3 wt. %, the value of reliability is less than 0.8 (Fig. 3). This decrease is due to the water-holding nature of the additive and its greater resistance to conversion compared to vinyl-acrylic copolymer (Neolith 6700). It is this characteristic that affects the acquisition of strength by cement in the early hardening periods, which is why the reliability factor is of such importance. At the same time, it is worth noting that the use of calcium formate as a water-retaining additive compared to Neolith 4400 and Neolith 6700 is also advisable at low concentrations.

Comparing the methodology for studying the effects of all three additives to papers [8, 10, 13], there is a complete absence in the latter of determining the chemical effect on changes in the characteristics of the cement matrix. This is explained by the fact that the cited works [8, 10, 13] were more focused on the narrow production sphere, which, in turn, is advisable only for one range of production. Moreover, the comparison of the
obtained results indicates a significant difference in the methods of production of additives in different countries. 

To prove this statement, regarding the orientation only to the physical and mechanical indicators of the cement matrix, it is enough to compare the research methodology to those more similar in works [4, 14]. The authors of [4, 14] also associated changes in the solidification duration and strength gain due to the formation of polymeric structures on the surface of cement minerals but did not indicate which transformations took place. This means that most of the research on this issue is focused only on the practical component compared to the scientific one, thereby not giving a more detailed answer to the question of the durability of these products. This question is answered by the study of chemical transformations that take place during the hydration of cement components in the matrix.

Such conclusions can be considered appropriate both from a practical point of view and a scientific point of view. Since the results allow for a more reasonable approach to the issue of determining the impact on the characteristics of the cement matrix in the long-term aspect of using a particular additive. From a practical point of view, their main direction of application and their effectiveness in obtaining the necessary properties for mixtures are confirmed (Tables 1, 2 and Fig. 1–4). Instead, from a theoretical point of view, our results (1) to (4) and Fig. 5–9) can be used to design mixtures and solutions for special purposes, as well as to predict the durability of articles.

The limitation of this study is that the design and study of the compositions of mixtures with these additives was carried out regarding their direct effect to the cement matrix in a controlled environment. This is the basis for the need for additional research related to the issue of durability of articles for the use of redispersed powders in their practical application. At the same time, these studies will make it possible to expand both the practical and theoretical bases for studying the issue of the influence of additives on the strength acquisition by the cement matrix. Therefore, further research should be aimed at studying the issue of their impact on the durability of the cement matrix. The nature of the interaction with aggressive media of mixtures with redispersed additives is also quite important.

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7. Conclusions

1. Given the different origin of the additives, all three powders exhibit a water-retaining character. Calcium formate, not being a polymer, at low concentrations has an approximate water retention value, as well as redispersible polymer powders. This approximation ranges from 0.3 to 2 % in accordance with the increase in the concentration of additives in mixtures. Thus, it can be noted that by the nature of calcium formate, it is similar to polymeric powders of water-retaining action.

2. The use of additives of organic origin leads to an uneven effect on the solidification duration and the acquisition of strength depending on the concentration of the additive in the mixture. For Neolith 4400 and Neolith 6700, the optimal concentration as hardening retarders is 3 wt %. This statement is confirmed by a decrease in the working time of the mixtures (the end of the terms of solidification) at concentrations above 3 wt %. Calcium formate, on the other hand, demonstrates the character of a hardening accelerator. In general, their use reduces the overall strength of the samples on day 28. At a concentration of 1 wt %, a decrease is 9–15 %. Instead, the concentrations of 3–5 wt % reduce the strength of samples by 15–25 %.

3. By the nature of the chemical action, the calcium formate manifests itself as an accelerator of hardening due to dissociation and the active formation of calcium hydroxide. Neolith 4400 and Neolith 6700 perform more as hydrophobic agents due to the versatate group for “4400” and polyvinyl alcohol for “6700”, which is formed in an alkaline cement stone medium. The existing formations of calcium acetate according to the reaction equations may indicate that over time, the strength of the cement matrix is likely to increase.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.
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