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# Symbiotic Impact of Carbon Quantum Dots on Microstructural Development and Mechanical Behaviour of Cement Mortars

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#### Abstract:

Carbon Quantum Dots (CQDs) are a new environmentally pleasant material of the carbon family that has gained great interest in construction engineering due to their unique and valuable properties, such as high stability, minimizing toxic activity, and good water solubility. Therefore, this study aims to synthesize the CQDs using glucose as a precursor and apply it as nano-reinforcements in cement mortar. X-ray diffraction (XRD), Fourier Transform Infrared Spectroscopy (FTIR), and transmission electron microscopy (TEM) technologies were used to characterize the generated CQDs. The mechanical properties of the created CQDs-cement mortar were investigated by assessing the compressive and flexural strengths as well as the workability of the mortar samples. Experimentally, four different percentages of the developed CQDs— ranging from 0.005% to 0.030%—were added to the cement mortar. The acquired results showed that, at the age of 56 days utilizing 0.025% CQDs, the compressive and flexural strengths of the CQDs-cement mortar had grown by 72% and 59%, respectively. The workability study's findings revealed no evidence of a significant impact of CQDs on the mortar's flow characteristics. In conclusion, the findings of this study may suggest that CQDs can be used to enhance the mechanical properties of materials based on cement.

# **Keywords:**

Carbon quantum dots; Nano-reinforcement; mechanical properties

#### 1. Introduction

Ordinary Portland Cement (OPC), which is frequently used in construction, has inadequate tensile strength, heat of hydration, and strain capacity. This results in limited structural capabilities [1, 2]. These vulnerabilities result from pre-existing defects, including pores, shrinkage-induced fissures, and flimsy phase boundaries. As a result, reinforcements like steel and fibre are frequently added to OPC-based materials to increase their tensile strength and resilience to stress [3-5]. The addition of specific amounts of various nanomaterials, such as nano-iron oxide, nano-alumina, and nano-titanium oxide, to cement and concrete has also opened up new options due to recent advances in nanoscience and nanotechnology [6-12]. The literature supported the benefits of nanoparticles on the durability and mechanical characteristics of cement-based products such as cement mortars. However, it was also noticed that adding nanoparticles to cement-based materials increases the water demand and negatively affects the flow ability of fresh mixes, leading to diminished workability. Therefore, several researchers were devoted to developing nanomaterials to minimize the dehydration of cement-based materials. In this context, nanosilica, which has a round form, a diameter of less than 25 nm, and a specific surface area of 300 m/g, has been found to encourage cement hydration [13, 14]. The low aspect ratio, however, restricts the capacity to prevent microcracks and enhance post-peak toughness. Carbon nanomaterials like carbon black, carbon nanotubes, graphene oxide, and graphene nano-sheets have also gained attention for improving mechanical properties and controlling cracks at the nanoscale level [15-18]. Carbon nanotubes (CNTs) are extremely strong and have a large surface area, particularly, and have demonstrated superior mechanical performance in cement [19]. However, their efficiency in reinforcement is limited by poor dispersion in water and lack of interfacial area between cement and CNTs, leading to decreased efficiency of reinforcement and non-uniform distribution of CNTs over cement composite [20-22]. The addition of dispersing agents has been shown to improve CNTs-cement bonding and increase the mechanical strength of the complex.

Carbon atoms are organised in a single plane to form a sheet in the structure of graphene, and it also has sp2-joined carbon atoms with strong mechanical and inherent strength [23]. Graphene sheets form a significant contact area from their top and bottom surfaces, which results in extremely good contact with the host materials, such as concrete. A single graphene sheet has a theoretical surface area of about 2600m<sup>2</sup>/g, which is higher than the surface area of CNTs [24, 25]. Graphene was considered a viable option to enhance the characteristics of cement-based materials due to its high surface area and unique physical and chemical properties; nevertheless, its extensive use in many industries is constrained by its expensive manufacture and challenging dispersibility. In recent years, graphene oxide (GO), which contains nano-layer sp<sup>2</sup>-hybridized carbon atoms and oxygen functionalized groups [10, 25-27], the presence of oxygen functionalized groups in GO can alter the van der Waals interaction enhance their dispersion in water [27]. Also, the carbon-cement-based nanocomposites can also be able to perform new functions, such as sensing and temperature monitoring etc. it, depending on the type of nanomaterials used.

Since their discovery in 2004, carbon quantum dots (CQDs), which consist of an aromatic sp2-hybridised domain and an sp3-hybridised matrix, have advanced as an emerging class of nanomaterials. With a diameter of 1–10 nm and oxygen functionalized groups, carbon quantum dots are a relatively new class of carbon nanomaterials. They have gained attention recently for their excellent solubility, low toxicity, tunable fluorescence, superior electronic, and catalytic properties, among other things [28]. CQDs have evolved as a new rising star of the carbon family. They are currently considered a potential candidate for different applications, such as sensing [29], catalysts [30], solar cells and cement-based materials [31]. Incorporating CQDs in a cement matrix is still a new application, and the literature does show enough evidence of their effects on cement-based bodies [1, 4]. In the field of civil engineering, carbon quantum dots (CQDs) are proving to be a capable material with abundant uses. Despite being less well-known than other materials in the area, they have special qualities that make them useful in a variety of applications [32-34]. The following are some of the main ways that CQDs are crucial in civil engineering: Environmental Remediation: CQDs can be used to clean up polluted water and soil. Because of their large surface area and strong adsorption properties, they are effective at removing heavy metals and organic pollutants from water and soil, preserving the environment [35]. Building Materials: CQDs can be used to improve the quality of building materials. They can enhance mechanical strength, durability, and even self-healing qualities when added to concrete or asphalt. Infrastructure may become more durable and resilient as a result [36]. Applications in Nanotechnology: CQDs may be used in civil engineering nanotechnology. They can be applied to create nanocomposites for use in building and infrastructure projects that have better qualities, like advanced strength and lighter weight [37]. Design for Sustainability: Including CQDs in civil engineering projects is in line with the increasing focus on sustainability and green construction practices. Their use can encourage the creation of environmentally approachable infrastructure and lessen negative belongings on the environment.

Therefore, a series of laboratory experiments are used in the current work to examine the impacts of CQDs on the microstructure and mechanical characteristics of the cement mortar.

#### 2. Materials and Methods

#### 2.1 Materials

#### **2.1.1 Cement**

A compound that binds different materials together by chemical processes, frequently aided by water, is referred to as "cement" in popular usage [30]. Ordinary Portland Cement (OPC) of 53 Grade was used in this study for the preparation of mortars.

#### 2.1.2 Fine Aggregate

Fine aggregates are the major part of the mortar in weight, and they play a vital role in contributing to the strength parameters of the mortar. The manufactured sand utilized in the study as fine aggregates and it has passed all the tests according to Indian Codal provisions. Due to the construction industry's explosive growth, manufactured sand, also known as M-sand, has developed a viable alternative to natural river sand. The scarcity of suitable river sand worldwide has prompted the extensive adoption of M-sand. Its utilization is driven by factors such as accessibility and costeffectiveness, as it can be easily obtained by crushing hard granite rocks found nearby, eliminating the need for expensive transportation from distant riverbeds.

Consequently, incorporating M-sand into construction projects can significantly reduce expenses. Additionally, M-sand offers the advantage of being dust-free and allows for easy control of particle sizes, ensuring compliance with specific grading requirements [38]. The high-quality M-sand used in this scenario was procured from a local crushing plant testing to zone II (IS 383) 1987 [39], and other properties are investigated as per the recommendation of IS: 2386 (Part III) – 1963 [28], obtained results are presented in Table.1

Sl.No	Property	No of trail	Value	Average	Deviation	
1	Specific gravity	T-1	2.625	2.596	+0.029	
		T-2	2.603		+0.007	
		T-3	2.561		-0.035	
2	Water Absorption	T-1	1.38	1.25	+0.13	
		T-2	1.10	Ċ	-0.15	
		T-3	1.28		+0.03	
3	Fineness Modulus	T-1	2.90	2.86	+0.04	
		T-2	2.86		0.0	
		T-3	2.82		-0.04	

Table 1: Preliminary test results

# 2.1.3 Carbon Quantum Dots (CQDs)

Due to their distinctive features, CQDs are a type of nanomaterial that have grown significantly during the past several years [40]. CQDs have received considerable interest across multiple sectors, including the construction domain, due to their nanoscale nature and carbon-based composition [41]. With sizes usually below 10 nanometers, these dots comprise carbon atoms in crystalline or amorphous formations, and they exhibit distinct properties in terms of chemistry, rendering them highly advantageous for diverse applications [27].

In light of the mentioned merits of the CQDs, they have been used in this study to produce cement mortar and examine its effects on the properties of the produced mortars.

# **2.1.4 Components of Cements**

Due to its adhesive and binding qualities, cement is a central component in construction. Typically, limestone, clay, shale, and other materials are the basic elements of cement. These elements go through a number of processes to create the finished product.

				-					
Elements	В	CaO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	MgO	SO <sub>3</sub>	K <sub>2</sub> O	Na <sub>2</sub> O
wt%	0.023	65.2	20.8	6.17	3.97	1.10	1.12	0.15	0.17

Table 2: Components of Cements in Table 2

#### 3. Preparation and characterization of samples

#### 3.1 Synthesis of CQDs sample

CQDs can be produced by relatively straightforward and affordable processes, including hydrothermal synthesis, solvothermal synthesis, or microwave-assisted synthesis [42]. They are accessible to researchers thanks to their simplicity in synthesis, which also makes mass production possible. Figure 1(a) shows a schematic illustration of carbon quantum dots made using hydrothermal processes. 100ml of DI water and 1.5 grams of the precursor were dissolved and agitated for 30 minutes. The solution is transferred to hydrothermal autoclaves and heated at 1800°C for 12 hours in a hot air oven. The heated solution is then retrieved and centrifuged for 10 minutes at 15000 rpm to eliminate the unreacted carbon product. In the end, the supernatant was collected, a one kDa membrane dialysis experiment was carried out for three days, and transparent yellow-coloured liquid CQD was obtained, as shown in Figure 1(b). These liquid CQDs were then stored for use in the preparation of cement mortar, and the chemical equation for the synthesized CQDs is given in eq. (1), along with a diagram of its chemical structure in figure 1(c).

 $C_6H_{12}O_6 + H_2O \rightarrow CQDs + 6CO_2 \tag{1}$ 









method. (b) Synthesized CQD sample. (c) Chemical Structure of CQDs.

#### 3.2. Preparation of CQDs-Cement mortar

The CQDs-cement mortar samples were made of cement M-sand with the ration of 1:3 added with water and CQDs. The water-to-cement ratio was 0.5, which indicates that there are 0.5 units of water weight in the concrete mix for every unit of cement utilized. A crucial factor in the mix is the water-to-cement ratio, which affects the workability and strength of the composite and CQDs were added in amounts ranging from 0.005 to 0.03 wt% of cement. Three samples for mortar cubes and prisms are cast for each weight percentage of CQDs using identical methods. The cement was mixed with the water and CQDs in a plastic bowl and agitated with a hand mixer for 5 minutes. Then, the mixture was placed in cubic (70.6 x 70.6 x 70.6 mm) and prism molds (40 mm x 40 mm x 160 mm), and each mold was vibrated for 15 to 30 seconds vibration table, see Figure 2. All the mortar specimens were immersed in water for 24 hours before demolding them. Mechanical properties of the mortar samples were studied at ages of 7, 28 and 56 days, and average results are discussed in Figures 7 (a) & (b) & 8 (a) & (b).



Figure. 2 Mini-Conical Slump Flow Test

#### 3.3. Characterization of CQDs and CQDs-Cement mortar

Using a Bruker AXS D8 Advance X-ray diffractometer and CuK radiation with a 1.54 Å wavelength, the crystal structure of the produced CQDs was identified. Using a JASCO FTIR-6100 spectrometer, Fourier transform infrared spectroscopy (FTIR) was used to examine the chemical bonding nature of the samples. The Raman spectrum was measured using a Renishaw Invia laser Raman microscope with a 532 nm LASER excitation wavelength. The transmission electron microscope was carried out Using a Philips CM20 analytical transmission electron microscope and a thermionic lanthanum hexaboride (LaB6) electron source operating at a 200 kV accelerating voltage; pictures of the CQDs structure were captured using transmission electron microscopy (TEM). A drop of CQDs was drop cast onto a Holey carbon film on 400 mesh Cu grids after being diluted 1000 times, sonicated for 30 minutes, and then used to prepare the sample (CQDs, 5.0 mg/mL). The grid was used for TEM investigation after being dried out at room temperature for 24 hours. To evaluate the compressive strengths and flexural strength, the mixes were moulded into cuboids of 70.5mm x 70.5mm and 40 mm  $\times$  40 mm  $\times$  160 mm, respectively (Figure 3). A Compression testing machine with a load capacity of 1000 kN (ASTM D642 standard) and a Universal testing machine with a capacity of 1000 kN was used to assess compressive strength and flexural strength, respectively.



**Figure. 3** (a) CQDs-cement Mortar cubes for the compression test, (b) CQDs-cement mortar prisms for the flexural test.

#### **3.4 Characteristics of the CQDs**

Various analytical techniques were used in this study to understand the size, morphology, crystallinity, and superficial characteristics of CQDs. The size, morphology and crystal phase of the as-synthesized CQDs were examined using the Transmission electron microscope (TEM) and displayed in Figure 4. The obtained images of the CQDs samples exhibited a uniform spherical shape with particle sizes ranging between 5-10 nm confirmed by histogram images of CQDs(Figure 4.(c)) [29, 43].



Figure. 4 (a) TEM (b) HR-TEM and (C) Histogram images of CQDs

X-ray diffractometer images of the synthesized CQDs are shown in Figure 5(a). The diffraction peak obtained at 23.39° corresponds to the amorphous phase of carbon. Further, the inter-planer distance value of CQDs was found to be 021nm.

FTIR spectroscopy was adopted to analyze the surface functional collections and chemical bonds of as-made CQDs. Figure 5(b) shows the CQDs FTIR spectrum. The broad characteristic peak observed around  $3100 \text{ cm}^{-1} - 3400 \text{ cm}^{-1}$  is attributed to -OH stretching vibrations of amino and hydroxyl groups. The peaks obtained at 2857 cm<sup>-1</sup>, 1566 cm<sup>-1</sup> and 1408 cm<sup>-1</sup> correspond to C-H, C=C, -COOH vibrational stretching, respectively. These functional groups play a crucial role in increasing the cement hydration properties. Similarly, the peaks around 1630 cm<sup>-1</sup> correspond to C=O, stretching mode [44].

The Raman spectrum of CQDs is depicted in Figure 5(C). The latter exhibits the characteristic peak at 1586 cm<sup>-1</sup> that is attributed to the G band, which arises due to the first-order scattering of the E2g mode in carbon materials [45]. The other peak is observed at 1368 cm<sup>-1</sup> attributed to the D band, which corresponds to the presence of defects in CQDs; these defects are helpful in increasing the mechanical strength of the cement. A fraction of the carbon particles in the CQDs are sp2-hybridized, and topological flaws may be found in the CQDs, according to recent literature [17, 18]. These flaws would unavoidably result in significant flatness deformation [19]. The wrinkly exterior might offer mechanical interaction between the CQDs and cement matrix, improving the interfacial adhesion improving load transmission and binding strength between the CQDs and the cement medium [46].



Figure. 5 (a) XRD pattern, (b) FTIR spectra (c) Raman spectra of carbon quantum

#### 4. Results and Discussions

# 4.1 Mini-Conical Slump Flow Test

Cement-based materials must have a high flow ability, moderate viscosity, and acceptable cohesive stability to achieve the homogenous distribution of engineering qualities in cement composite [29]. The impact of CQDs on the performance of mortar was evaluated by a mini-conical slump flow test at different mass ratios of CQDs ranging from 0.005% to 0.030%. The results showed that the CQDs-cement mortar samples exhibited strong preliminary slump and moderate slump loss at different mass ratios of CQDs, which could be attributed to the abundance of hydrophilic groups, such as carboxyl and amide groups. Since CQDs are hydrophilic and the acidity is increased by the breakdown of hydrogen particles into protons, the - OH- link's polarity increases, and Ca<sup>2+</sup> produces ligands with carbon of amide structure [22, 23]. To minimize the unfavorable impacts of a high surface area and high water absorption on cement slump, the carboxyl and amide clusters of CQDs will influence the concentration of Ca<sup>2+</sup> in the cement pore solution.

The results showed that the slump diameter decreased when 0.025% CQDs were added to the mixture; standard deviation values are also mentioned in the graphical representation (Figure 6). This finding may be explained by the CQDs' surface area, which decreases the amount of water in the fresh mixtures. However, it was noticed that the slump diameter increased by 48.9% when 0.020% CQDs were added at w/c = 0.5. According to the literature, poly-carboxylate admixtures can considerably improve the workability of CQDs-cement mortar [21]. Poor workability is commonly coupled with poor compaction ability due to entrapped significant air gaps in the material [22].





# 4.2 Mechanical Properties of CQDs-Cement Mortar

This study used three samples for each test to assess the compressive and flexural strengths of CQDs-cement mortar samples at various ages. The results show that the compressive and flexural strengths of the CQDs-cement mortar samples have significantly increased when compared to the reference samples. In general, it was found that adding CQDs enhanced the compressive and flexural strengths after 56 days in comparison to the reference samples by 73 and 59%, respectively. Below is a list with further information regarding the findings attained.

#### 4.3 Compressive strength

The ability of mortar to withstand compressive forces or loads is referred to as its compressive strength. It is a crucial characteristic that affects the overall toughness and longevity of structures. Three cubic specimens (70.6 mm x 70.6 mm x 70.6 mm) were tested in each stage of this study to assess the compressive strength of the CQDs-cement mortar and reference samples. The compressive strength test was carried out with a loading rate of 0.05 MPa/s. Figure 7(A) shows the results obtained from the tests along with standard deviation values, and it can be clearly seen that the compressive strength increased when the CQDs/cement ratio increased from 0.005% to 0.025%, but it suddenly decreased when the CQDs/cement ratio increased more than 0.025%. This increase in strength is due to the influence of CQDs in the cement mortar. Therefore, it can be concluded that the optimum dosage of CQDs for cement mortar is 0.025%. The fact that the CQDs have a strong seeding impact on the cement hydration kinetics and provide more sites for the nucleation formation of new areas of hydration products may be the cause of the increase in compressive strength with an increase in the CQDs ratio. Also, the presence of oxygen-functionalized groups in the CQDs accelerates the hydration process.

The influence of age on compressive strength is depicted in Figure 7(B), and it is clear that as mortar ages, its strength gradually increases. Additionally, it should be noted that the compressive strength rises as the CQDs ratio rises until it reaches 0.025%. (it reached 73% of the compressive strength of the reference samples after 56 days), then it decreases when the CQDs ratio becomes 0.03%.





**Figure. 7** (a) Effect of CQDs ratio on the compressive strength (b) Effect of age on the compressive strength.

#### 4.4 Flexural strength

Another crucial mechanical characteristic of cement mortar is called flexural strength, which assesses the material's capacity to sustain bending or flexing forces without cracking. The standardized modulus of rupture test, which is commonly used in the literature, was used in this study to assess the flexural strength of CQDscement mortar samples, according to ASTM C307. The prism specimens (40mm X 40mm X 160mm) were tested for flexural strength using a Universal Testing Machine (UTM). The obtained results with the standard deviation measurements are shown in Figure 8a, which shows that increasing the ratio of CQDs from 0 to 0.025% improved the flexural strength of the mortar by 59% (after 56 days) compared to the reference samples, Quantum dots make the material more homogeneous, which lowers flaws and improves overall structural integrity. When compared to materials without CQDs additions, this greater uniformity can produce materials with higher flexural strength. It's also critical to remember that the success of CQDs materials in improving flexural strength depends on a number of variables, including the kind, size, and concentration of the components, their compatibility with the matrix material, and the manufacturing technique used to make the composite. However, increasing the CQDs ratio to 0.03% had negative effects on the flexural strength.





flexural strength.

# 4.5 Mechanism of effects of CQDs on the Mechanical Properties of Cement Mortar

The CQDs' preliminary process for increasing their mechanical strength Based on the aforementioned findings, cement mortar was explained. The mechanism was comprehended by using SEM pictures for 7 days, 14 days, and 28 days to prepare CQDs cement mortar. Portlandite (defined as CH) and ettringite are two of the three products of C-S-H and C-OH that are primarily generated during the hydration process [47]. We presume that C-S-H entirely encircled the cement particles' surface or border due to the surface hydrophilic functional groups, fillers, nucleation function, quantum confinement effect, and CQDs [48]. Additionally, CQDs that are dissolved in water and are difficult to agglomerate can act as plentiful and uniform nucleation sites, which is advantageous for the development of additional C-S-H. The structure of cement-based materials becomes more compact with the creation of C-S-H phases because they concurrently cover the cement particles' surfaces and those of the CQDs [49]. Additionally, CQDs can fill the gaps or voids in cement particles by acting as Nanospheres. CQDs also include a lot of oxygen-containing hydrophilic functional groups, which helps them adhere to cement particles and create more C-S-H species as a result [50]. Reduced porosity and higher mechanical strength of cement-based materials are the results of the aforementioned joint effects, which create a denser and less permeable microstructure[51-53].

The pore configuration and porosity of the cement mortar determine how well hydrated cementitious composites operate, which in turn affects the service life of Cement mortars through their permeability and ion diffusive properties. The addition of CQDs can improve cement cement mortars' pore configuration by reducing the macrospore volume and critical pore size because the CQDs fill the bigger pores in cement mortars, which enhances pore configuration. Due to their nanoscale size, CQDs provide more opportunities for the hydration of the products and, thereby, improve the mechanical properties by filling fractures, creating crystalline formations during cement particle hydration.

The organization, size, form, and distribution of the CQDs inside the mortar matrix are all taken into consideration when describing the microstructure of CQDs-cement mortar. Thus, SEM images were obtained for the developed samples in this study (Figure 9), and it was noticed that small particles are present over cement sheets, which confirms the reinforcement of cement with CQDs. Most of the phases formed during the hydration process are composed of three C-S-H products: portlandite (CH) and ettringite (AFt Ettringite, often known as AFt (short for "aluminium ferric trisulfate"), is one of the early hydration products in cementitious systems.). The surface hydrophilic functional groups of CQDs, fillers, and nucleation function are nano in size, which boosts the mechanical strength of the CQDs-cement mortar. Water-soluble, hard-to-agglomerate CQDs nanoparticles can serve as numerous homogenous nucleation sites, which is beneficial for producing more C-S-H. Although there is a chance that CQDs and the C-S-H phase could interact, the bonding mechanism would most likely involve a mix of chemical and physical interactions at the interface between the two phases, such as electrostatic interactions and van der Waals forces. Further study is required to explore and optimize the interactions between CQDs and cementitious materials to improve the performance of these composites in various applications.

The development and understanding of CQDs-cement mortar are still relatively new fields of research, and the specific details of the C-S-H bonding mechanisms between cement and CQDs can vary depending on the experimental conditions and the specific properties of the CQDs and cementitious materials used. Portlandite (CH) and ettringite (AFt) are two of the three C-S-H products that make up the common phases that occur during hydration. For cement particles that weren't moist, it is assumed that C-S-H covered the area or boundary. The external hydro-philic functional groups of CQDs and the fillers and nucleation function they provide as an outcome of the nano-size effect boost the mechanical strength of the specimens when CQDs are introduced to cement. More C-S-H can be produced using water-soluble, difficult-to-aggregate CQDs nanoparticles as multiple, homogenous nucleation sites. The simple cement paste displays a porous microstructure; how-ever, adding 0.025% CQDs (wt% of cement) caused a significant change in the microstructure, with CQDs particles producing a very thick and dense texture of hy-drated products by both filter and pozzolonic effects.



Figure. 9 SEM image of plain cement mortar and CQDs-cement Mortar of differ-

ent ages.

# Conclusions

This research examination concentrated majorly on the effects of CQDs on the mechanical properties and flow ability of cement mortar through a detailed experimental investigation. The microstructure of the composites made of cement was also examined in the study.

The results obtained indicated that adding a reasonable ratio of CQDs to the mixtures of the cement mortar improves the mechanical properties of the cement mortars that are produced, but high ratios of CQDs negatively impact the quality of the cement mortars. Generally, it was found that the best compressive and flexural strengths can be obtained using a CQDs/cement ratio of 0.025% after 56 days of curing as per test results, where it was noticed that this ratio of CQDs improves the compressive and flexural strengths by 79% and 59%, respectively, compared to the reference samples.

The results of this study could be preliminary evidence of the possibility of using the CQDs as reinforcements in cement-based combined materials.

However, there is still a need for more studies to better understand the mechanism of the effects of the CQDs on the properties of the cement-based combined materials.

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# **Conflicts of Interest:**

The authors declare no conflict of interest.

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# **Declaration of interests**

 $\boxtimes$  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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