Rice Husk and Old Corrugated Container Cement Boards: Performance of Nano-SiO_2 on Strength and Dimensional Stability

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ABSTRACT: *In this study the effect of nano sized silica particles (nano-SiO_2) on the physical and mechanical properties of rice husk and Old Corrugated Container (OCC)-cement boards was investigated. Modulus Of Rupture (MOR), Modulus Of Elasticity (MOE), Internal Bonding Strength (IB), density, water absorption and thickness swelling after 24 hours immersion in water and hardness were measured. Results showed that rice husk-cement boards with 2% nano-SiO_2 demonstrated the best physical and mechanical properties. It is related to better distribution and compaction of particles during compression without particles agglomerating. A higher property of 2%-nano-SiO_2 content rice-husk-cement boards was confirmed by their higher hydration temperature. Addition of 3% nano-SiO_2 to the mixture reduced the density because of both the substitution of denser cement particles by lighter nano-SiO_2 particles and air-entrapment in the boards. Furthermore, Scanning Electron Microscopy (SEM) analyses showed that the optimum amount of nano-SiO_2 (2%) can fill micro pores and make a uniform structure with a rough surface which improves properties of composite boards.*

KEY WORDS: *Cement boards; Nano-SiO_2; Rice husk; Old Corrugated Container (OCC); SEM.*

INTRODUCTION

The rapid growth of the world's population and consequent different demands has resulted in increasing consumption rate of organic and inorganic, as well as renewable and non-renewable sources of raw materials. This has the potential of diminishing raw material availability and leading to environmental damages. Therefore, in recent years the development and application of lightweight, sturdy and, durable construction materials has been of great interest. As a result, wood-cement bonded boards are widely used in building constructions throughout the world as suitable candidate for construction materials. Wood-cement boards,

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as compared to other construction materials, have many advantages including; increased strength, light weight, high heat and sound isolating capability, and fire resistance. Furthermore, it is a cost-effective and eco-friendly alternative for construction materials. However, decreasing forest resources made it necessary to find and replace a substitute for wood in the manufacturing of fiber cement boards. Therefore, utilization of wood processing residues, agricultural residues, wastepaper, as well as pulp mill wastes have increased dramatically in recent years.

Agricultural wastes such as rice husk, coconut coir, peanut shells, and bagasse were used to manufacture the natural fiber-cement boards. There was no adverse effect of these materials on Portland cement hardening and strength of resultant board [1]. *Asasutjarit et al.* reported that the coconut coir-based lightweight cement boards can be used as a building insulation material to reach energy conservation in buildings [2]. The use of arhar stalk, as a suitable residue, in cement-bonded composite building products was also investigated by *Aggarwal et al.* [3]. In an investigation of the potential reuse of waste rice husk as fiber composites in concrete, it was described that boundary of fiber-matrix transition zone has excellent adhesion and that rice husk in concrete is not subject to damage over the years [4]. Among agricultural residues, rice husk contains rather high silica content and has a very good potential to be used as additive in cement boards. Over half a million tons of rice husk is produced in Iran per year, but it is currently burned by farmers in farms [5]. The practice does not only lead to air pollution and soil nutrition loss, but also reduces crop yield. Moreover, the quantities of wastepaper generated worldwide have recently increased due to the high consumption of paper and paper products. Reusing and recycling of waste paper in panel product industry have been the most favorable solution to providing a suitable substitute for virgin fibers as well as asbestos fibers, especially in developing countries. Wastepaper fibers, sludge, and other recycled sources of pulp materials are already used in cement bonded products [6]. Wastepaper is suitable for reinforcing cement composites, even though it contains undesirable amounts of fiber fragments due to excessive processing [7]. Moreover, the technical properties of three layer Cement-Bonded Boards (CBBs) produced from wastepaper and sawdust was introduced by *Fuwape et al.* [8]. Therefore, consumption of these kinds of waste is at the center of attention of many researchers and industries to obtain resistant, renewable, and cost-effective materials with added-value. Generally, the field of CBBs manufacturing may introduce a potential utilization of wood wastes and low quality forest thinning in the US, as well as other countries [9].

Different studies conducted evaluate the effectiveness of different additives in enhancing the hardening reactions of cement in the presence of lignocellulosic materials. In addition to usual chemicals, utilization of nano particles in the manufacturing process of composite materials with new functional properties has been studied by many researchers [10-15]. Nano-SiO_2 generates thicker cement paste, accelerates the cement hydration process, and increases the paste strength [16, 17]. Nano-SiO_2 decreases the setting time of mortar when compared with silica fume and reduced bleeding water and segregation by improving its cohesiveness. Additionally, it can control the degradation of calcium silicate hydrate reaction that is caused by calcium leaching in water, blocking water penetration and leads to improvements in durability [18]. *Wen et al.* reported that nano-SiO_2 improves the mechanical properties of gypsum particle-boards [19]. Three percent nano-SiO_2 was the optimum amount added to these boards to enhance both Modulus Of Rupture (MOR) and Modulus Of Elasticity (MOE) at 30°C and 40°C, respectively. Nano-SiO_2 behaves not only as a filler to improve mortar cement microstructure, but it also enhances pozzolanic reactions [20]. It was reported that adding nano-SiO_2 increases the strength, flexibility, and aging resistance of polymers [21]. SiO_2 is known to improve the fire-resistant properties of wood-inorganic composites [22]. *Li et al.* described that the compressive and flexural strength of cement mortars with nano-SiO_2 were higher than cement mortars without the addition of additives at similar wood/binder ratio [23]. Accordingly, waste lignocellulosic materials combined with mineral filler (nano-SiO_2) can be successfully used to manufacture filler reinforced composites (FRCs) [24]. Use of rice husk and municipal wastepaper in wood cement board production will address environmental challenges associated with these and yet provide an advantage in the production of lightweight, low-cost, and renewable construction material.

This study seeks to investigate the potential use of rice husk particles and OCC (Old Corrugated Container)

Table 1: Physical and chemical properties of nano-SiO_2.

Properties	Average value
Specific surface area(BET), m^2/g	200±25
Average primary particle size, nm	7-14
Tapped density, g/L	50
Moisture, 2 hours at 105°C, wt.%	<1.5
Ignition loss, 2 hours at 1000°C based on material dried 2 hours at 105°C, %	<1
pH in 4% dispersion	3.7-4.7
SiO_2 content based on ignited material, wt.%	>99.8

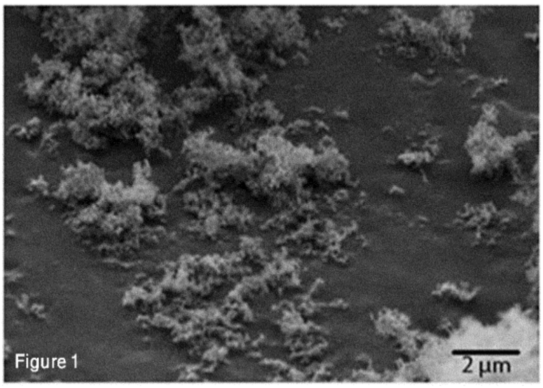

Fig. 1: SEM micrograph of Nano-SiO_2 .

fibers for manufacturing of cost-effective natural fibers-cement boards with improved properties. In order to determine the optimum content of nano-SiO_2 as reinforcement in natural fiber-cement board composition, the effect of different weight percentages of nano-SiO_2 on the properties of the manufactured boards was also investigated. Moreover, the physical and mechanical properties of the fiber-cement boards manufactured by rice husk and OCC were compared.

EXPERIMENTAL SECTION

Materials and preparation

Two lignocellulosic materials were used; namely, OCC and rice husk. The OCC was randomly collected from different stores in Karaj, and rice husk was collected from a rice-grinding mill. Both materials were ground by a rotary mill with a feed size 1.5 mm mesh into smaller particles. Dimensions of lignocellulosic materials were measured by optical microscopy (Olympus, Japan). The average length and diameter of OCC fibres were 1.2 mm and 39.2 µm and the average dimensions of rice husk particles were 0.77 mm and 0.28 mm. Type II Portland cement, provided from Abyek factory, was used in this study as specific surface area, specific gravity and SiO_2 content of cement were 2600 cm^2/g, 3-3.25 ton/m3 and 20.5 % (w/w), respectively. The nano-SiO_2 powder with purity of 99.8% SiO_2 was applied as a replacement of the cement at four weight percentages (0, 1, 2 and 3% by dry weight of the cement). Nano-SiO_2 was purchased from Plasma Chem. GmbH (Berlin, Germany). Its physical and chemical properties and SEM micrograph are given in Table 1 and Fig. 1, respectively.

Methods

A total of 8 combinations (two lignocellulosic materials and four fractions of nano-SiO_2) were applied and for each composition three replicate boards were produced. To manufacture the boards, at first Portland cement and lignocellulosic material (rice husk and OCC) were initially hand-mixed. Dispersed nano-SiO_2 in water was then added to the mixture and the mixture was stirred for 15 minutes using an electric stirrer. The target density and thickness of boards were 1.3 g/cm^3 and 12 mm, respectively. Next, the mixture was uniformly distributed in a mold with dimensions of, 35 cm × 27 cm × 4 cm, which was then placed on a metal plate and covered with cellophane (this prevented the board from sticking to the plate). Another plate was placed on the top of the mat. The mat was cold-pressed using a hydraulic press, which applied 3 MPa pressure for 10 minutes. For primary curing to occur, the boards were held for 24 hours. The boards were then removed from the mold and conditioned at 95% relative humidity and room temperature for 28 days for final curing of the boards. The cured boards

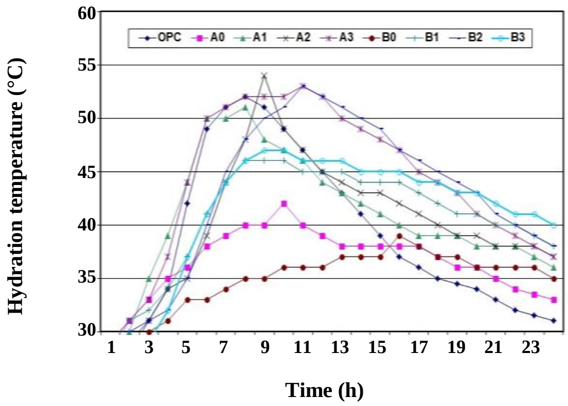

Fig. 2: Hydration temperature of treatments during 24 hours (OPC: Ordinary Portland Cement, A: rice husk, B: OCC, indices are nano-SiO_2 contents from 0-3%).

were dried at 65±2% relative humidity and 20±1°C for one month, until equilibrium was reached.

To perform the hydration test, type II commercial Portland cement was used in 200 g batches. Distilled water (90.5 mL) was added to the mixture of cement (200 g) and rice husk and OCC (15 g oven dry basis) in a polyethylene bag for 3 min [25-28]. The mixture was placed in an insulated flask that is equipped with a thermocouple wire. The hydration temperature of the mixture was measured and plotted against time for a period of 24 hours.

Sample preparation and measurement of physical and mechanical properties were carried out according to DIN 634-1 and 2 [29, 30].MOR and MOE, Internal Bonding strength (IB), density, water absorption and thickness swelling after being immersed in water for 24 hours, and hardness were also measured. Samples for all tests were cut from the bending specimens. Likewise, Scanning Electron Microscopy (SEM) was used for microstructural observation. Analysis of variance (ANOVA) was used for statistical analysis of the data. Under circumstances where significant differences were observed, Duncan multiple range test was used to group the averages. The experiment was arranged as a 2 by 4 factorial in a completely randomized design.

RESULTS AND DISCUSSION

Hydration temperature

Fig. 2 illustrates the hydration temperature of all samples plotted against time. Rice husk-cement boards with 2% and 3% nano-SiO_2 content, due to high activity and large specific surface of nano-SiO_2, exhibit higher maximum hydration temperatures than Ordinary Portland Cement (OPC). It seems that these materials do not have any retarding effect on cement hydration and, enhance the rapid formation of hydrated products. Moreover, nano-SiO_2 improved the performance of lignocellulosic material. Impregnation of calcium content on the fiber walls showed better strength enhancement [4]. Fig. 2 confirms that nano-SiO_2 is responsible for a higher hydration degree of cementitious compounds [31].

Nano-SiO_2 has high surface energy and atoms in the surface have a high activity, which leads the atoms to react on outer ones easily. When nano-SiO_2 is added to cement, it acts as the micro-filler of the cement particles, which can reduce the amount of water.

The presence of many unsaturated nano-SiO_2 (bonds ≡Si-O- and ≡Si-) on the surface, the reaction process between SiO_2 and Ca $(OH)_2$ may be as follows:

≡Si-O-Si≡ +H-OH → ≡Si-OH (react quickly)

≡Si- +OH→ ≡Si-OH (react quickly)

≡Si-OH +Ca $(OH)_2$ →Ca-Si-H

It is therefore possible that the nano-SiO_2 is accelerating the cement setting and the hydration processes [16, 32].

Physical and mechanical properties of natural fiber-cement boards

The average values of physical and mechanical properties (MOR, MOE, IB, Density, Hardness, and Water absorption and Thickness swelling after 24 hours immersion in water) of boards containing 0-3% nano-SiO_2 at 28 days are summarized in Table 2.

Rice husk-cement boards manufactured by 2% nano-SiO_2 showed the highest bending strength. This is due to the boards having higher hydration temperature and density of these composites. Increasing the nano-SiO_2 content (in optimum percent) contributes to a denser microstructure and leads to an increase in Ca-Si-H chain dimension and also Ca-Si-H stiffness [33]. The quantity of nano-SiO_2 present in the mix is close to the amount required to combine with the liberated lime during the process of hydration [34, 35]. Reaction between nano-SiO_2 (with high specific surface) and calcium hydroxide, generating Ca-Si-H gel, improves this property. The correlation between hydration temperature, bending strength, and internal bonding strength has been reported [36].

Table 2: The average of physical and mechanical properties of boards at 28 days

Treatment	MOR (MPa)	MOE (GPa)	IB (MPa)	D_o (g/cm^3)	Hardness (MPa)	WA_{24} (%)	TS_{24} (%)
25% Rice husk + 0% nano-SiO_2	5.8 (0.5)*	4170 (176.5)	1.49 (0.4)	1.38 (0.09)	15.7 (3.3)	18.6 (10.1)	0.75 (0.3)
25% Rice husk + 1% nano-SiO_2	7.5 (1.02)	5489 (446.5)	2.14 (0.4)	1.39 (0.06)	22.1 (5.3)	17.5 (3.7)	1.1 (1.5)
25% Rice husk + 2% nano-SiO_2	10.6 (1.3)	6484 (323.2)	2.33 (0.2)	1.33 (0.03)	30.4 (2.7)	21.6 (6.1)	0.7 (0.4)
25% Rice husk + 3% nano-SiO_2	7.1 (0.5)	5777 (266.3)	2.34 (0.2)	1.28 (0.60)	19.8 (3.05)	22.8 (1.1)	0.8 (0.5)
12% OCC+ 0% nano-SiO_2	6 (1.28)	3043 (516.8)	1.01 (0.2)	1.18 (0.07)	22.5 (9.1)	32.2 (14.4)	3.7 (2.03)
12% OCC+ 1% nano-SiO_2	4.9 (0.9)	2232 (848.7)	1.06 (0.2)	0.99 (0.05)	30.2 (6.89)	32.5 (20.2)	3 (1.3)
12% OCC+ 2% nano-SiO_2	8.1 (1.02)	3167 (603.2)	1.04 (0.3)	1.05 (0.06)	23.7 (12.7)	32.7 (15..2)	2.3 (1.2)
12% OCC+ 3% nano-SiO_2	4.2 (1.1)	1912 (750.7)	0.93 (0.2)	1.00 (0.06)	26 (7.04)	37.1 (11.7)	1.9 (1.08)

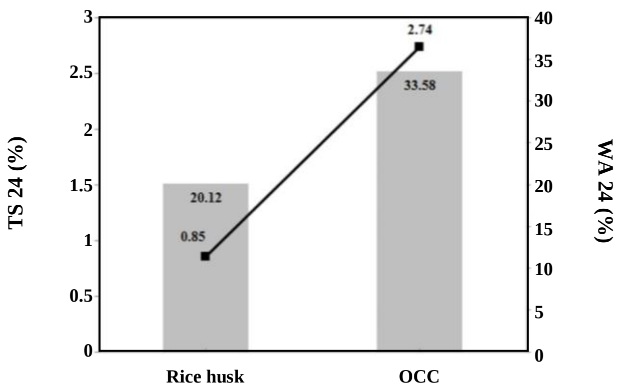

Fig. 3: Effect of lignocellulosic materials on WA and TS after 24 hours immersion in water.

Higher content of nano-SiO_2 by ≥ 3% leads to a lower level of MOR and MOE, due to agglomeration caused by hardly dispersed particles during the mixing stage [37]. Addition of 1% nano-SiO_2 to OCC-cement boards decreased MOR and MOE; in other word, formation of OCC-composite containing 1% nano-SiO_2 was frustrated. Difference between treatments applying 0, 1 and 3% nano-SiO_2 was not statistically significant.

Rice husk-cement boards manufactured without nano-SiO_2 showed lowest IB and the difference among boards incorporating nano-SiO_2 was not statistically significant. Improving IB by adding nano-SiO_2 can be a consequence of its high activity and large specific surface (200±25 m^2/g). Adding nano-SiO_2 causes a decreasing of $Ca(OH)_2$ crystals and consequently improves the bonding strength between the lignocellulosic material and cement [31].

Rice husk-cement boards demonstrated the IB measure two times more than OCC-cement boards. Water absorption and swelling of OCC fibers was higher than rice husk particles which consequently decreased internal bonding strength. A small increase in the thickness of OCC boards compared to those of rice husk confirms this statement.

Natural cement boards incorporating 3% nano-SiO_2 had the lowest density. Nano-SiO_2 addition to cement pastes and mortars decreases the apparent density and increases the air content in these mortars [38]. A reduction in density occurs due to both the substitution of denser cement particles by lighter nano-SiO_2 particles and air-entrapment in the boards. Pores and non-uniform distribution of 3% nano-SiO_2 particles can be seen in Fig. 4. Nanoparticles were uniform distributed in boards incorporated 2% of nano-SiO_2. Cement composites with suitable amount of nano-SiO_2 produce more stable bonding framework [36]. The density of rice husk-cement boards was 18% higher than OCC-cement boards. The geometry of OCC fibers, their high water absorption and swelling, and perhaps, insufficient removal of moisture during pressing could be the reason for the spring-back and for that matter, the thickness of these boards being more than rice husk boards which results in lower densities.

According to the results obtained by statistical analysis, the effect of nano-SiO_2 on hardness of boards

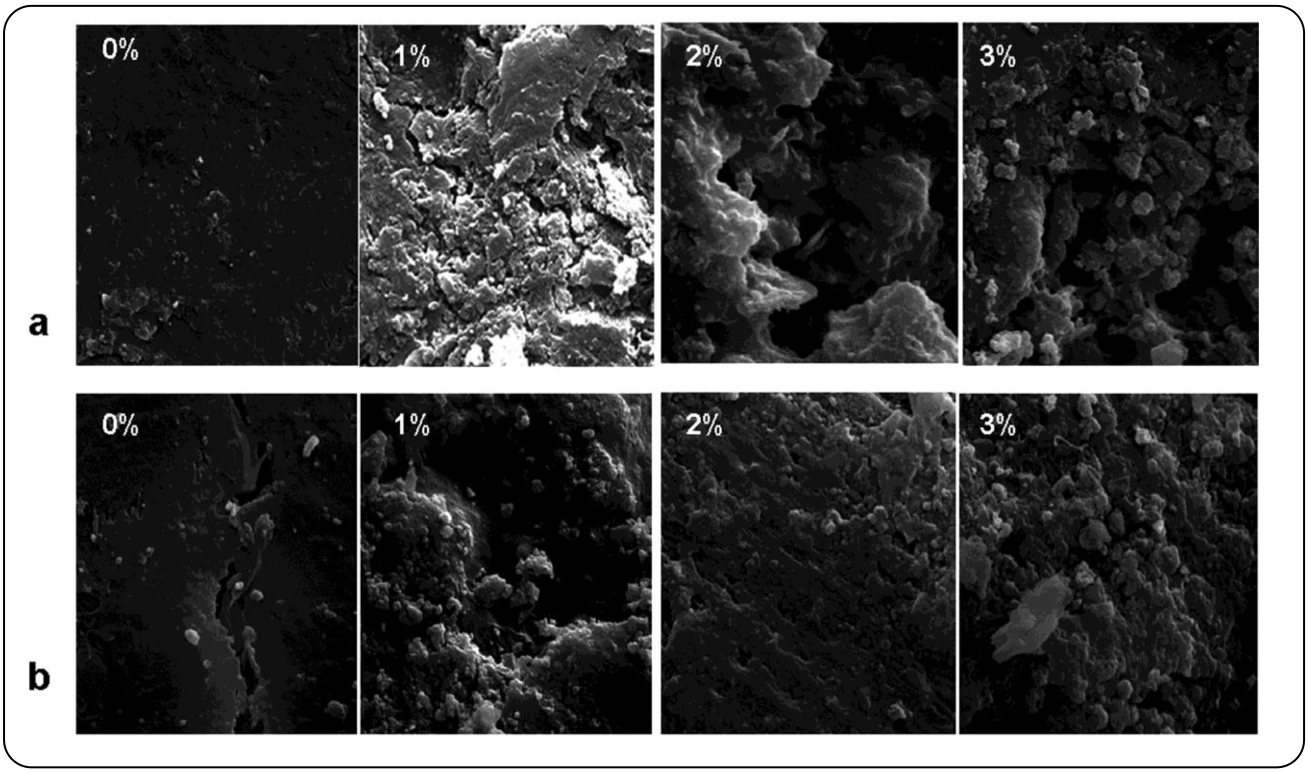

Fig. 4: ***SEM micrographs (×20,000) of rice husk (a) and OCC (b)-cement boards incorporating 0, 1, 2, 3% Nano-SiO_2.***

was significant at 95% confidence interval. Natural cement boards incorporating 1% and 2% nano-SiO_2 had the highest hardness. OCC boards with 1% nano-SiO_2 showed a high hardness (30.22 MPa) performance as high as the best nano-SiO_2 rice husk boards (30.36 MPa). Nano-SiO_2 showed a special structure of a three-dimensional network and is well-known for its high hardness and stability [39]. The difference between boards manufactured by 0 and 3% nano-SiO_2 was not statistically significant.

The effect of nano-SiO_2 on the WA and TS after 24 hours immersion in water was not statistically significant. In comparison with OCC-cement boards, rice husk-cement boards had 40% and 69% lower WA and TS, respectively, after 24 hours immersion in water (Fig. 3). This can be related to high ash content in rice husk. Also, reducing the density of OCC-cement boards increases the WA and TS of these boards.

SEM observation

In order to assess the effect of existence and also different contents of nano-SiO_2 on the microstructure of natural cement boards, SEM micrographs were taken from the central part of the samples. Fig. 4 shows the SEM micrographs of boards manufactured by both rice husk and OCC containing different amounts of nano-SiO_2.

In OCC-cement boards manufactured with no nano-SiO_2, cracks are visible. Low hydration of cement and spring-back after pressing induce microscopic cracks that decreased the strength. Addition of nano-SiO_2 up to 2% leads to reduction of $Ca(OH)_2$ crystals and increases the calcium silicate hydrate (Ca-Si-H) gel. This gel can fill the existing micro pores in the mixture leading to dense and uniform structure and improving the properties of the boards [18]. Nano-SiO_2, due to its high specific surface, can both improve the cement hydration through increasing the hydration temperature and affect the performance of lignocellulosic material.

Decreasing the strength of boards with the nano-SiO_2 content of 3% is due to agglomeration of nano particles in the mixture. When 3% nano-SiO_2 is added to the mixture, it generates higher tendency toward accumulation and agglomeration in contact with water. Porous and non-uniform structure in boards containing 3% nano-SiO_2 confirms their lower physical and mechanical properties. Therefore, optimum amount of nano-SiO_2 is suggested as small dosage.

CONCLUSIONS

Based on the results of the study, the following conclusions were reached:

● Rice husk-cement boards manufactured by the 2% nano-SiO_2 showed the highest bending strength, this is due to better compaction and distribution of particles and higher hydration temperature of boards.

● Addition of 3% nano-SiO_2 to the mixture leads to reducing density which can be related to both the substitution of denser cement particles by lighter nano-SiO_2 particles air-entrapment in the boards, and formation of clogs.

● SEM analysis showed that 2% nano-SiO_2 resulted in a decrease in the $Ca(OH)_2$ crystals with an increase in calcium silicate hydrate (Ca-Si-H) gel. The formation of this gel, helping hydration reactions to be developed, is likely to fill the micro pores in the mixture and therefore, leads to uniform structure and improved properties.

● Rice husk-cement boards showed superior physical and mechanical properties than OCC-cement boards. It can be due to compression stress releases when in contact with water thereby causing the spring-back of OCC-cement boards which decrease the strength.

● Optimum treatment was related to rice husk-cement boards manufactured with 2% nano-SiO_2.

According to this research, OCC fibers and rice husk can be used for the manufacture of cement based composite boards, as value-added materials, and their properties can be enhanced by selectively adding nano-SiO_2 particles which could enable the introduction of these materials in residential and industrial applications.

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