Kaunas University of Technology

Faculty of Civil Engineering and Architecture

The Influence of Zeolite on Building Composites Reinforced with Wood Fibres

Master's Final Degree Project

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Structural and Building Products Engineering (6211EX008)

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Raena Sehreen Athher Ahmed

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Declaration of Academic Integrity

I confirm that the final project of mine, Raena Sehreen Athher Ahmed, on the topic „The Influence of Zeolite on Building Composites Reinforced with Wood Fibres“ is written completely by myself; all the provided data and research results are correct and have been obtained honestly. None of the parts of this thesis have been plagiarised from any printed, Internet-based or otherwise recorded sources. All direct and indirect quotations from external resources are indicated in the list of references. No monetary funds (unless required by Law) have been paid to anyone for any contribution to this project.

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Kaunas University of Technology

Faculty of Civil Engineering and Architecture

Task of the Master's final degree project

Study programme: STRUCTURAL AND BUILDING PRODUCTS ENGINEERING

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(English):

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CEOLITŲ ĮTAKA STATYBINIAMS KOMPOZITAMS ARMUOTIEMS MEDŽIO PLAUŠU

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Analytical research	☐
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Other information (if needed):

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(contribution 100 %) *position, name, surname* *signature*

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Summary

The purpose of this work is to study the effects of incorporating zeolitic waste and synthetic zeolite on the physical and mechanical properties of cement composites reinforced with wood fibres. Wood fibres cement composites have the advantages of low-density, high insect resistance, high fungal resistance, strong acoustic insulation and good thermal insulation properties. These natural (wood) fibres are lignocellulosic materials with significant amounts of lignin, hemicellulose, sugars and extractives that inhibit the setting time of cement hydration. To reduce the retardation caused by the wood fibres, zeolitic waste and synthetic zeolites are introduced into the wood fibres-cement system. Zeolites are chosen in this study as they exhibit pozzolanic properties which can be useful to increase the workability and mechanical properties of the cement composite system. Three types of wood fibres cement composites are prepared for investigation, the first type of composites contains 5% untreated wood fibres and different amounts of zeolitic waste (0%, 1%, 2%, 5%, 10%) based on the weight of cement; the second type of composites contains 5% ultrasonically treated wood fibres and zeolitic waste (0%, 1%, 2%, 5%, 10%); the third type of composites contains 5% ultrasonically treated wood fibres and synthetic zeolite (hydrosodalite) obtained from silica-gel waste (0%, 1%, 2%, 5%, 10%). From the obtained results, Type-1 composites with untreated wood fibres and zeolitic waste indicated agglomeration of wood fibres in the structure of the samples. Wood fibres cement composite with ultrasonically treated wood fibres and 10% zeolitic waste (Type-2) showed the most optimal results in terms of mechanical strength and physical properties. In this research, the investigated density of samples with treated wood fibres revealed that it was possible to produce lightweight composites with the densities ranging from 1000-1200 kg/m^3.

Athher Ahmed, Raena Sehreen. Ceolitų įtaka statybiniams kompozitams armuotiems medžio plaušu. Magistro baigiamasis projektas / vadovė prof. dr. Danutė Vaičiukynienė; Kauno technologijos universitetas, Statybos ir architektūros fakultetas.

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Kaunas, 2021. 59 p.

Santrauka

Šio darbo tikslas yra ištirti ceolitinių atliekų ir sintetinio ceolito poveikį fizinėms ir mechaninėms cemento kompozitų, sustiprintų medžio pluoštu, savybėms. Medienos pluošto cemento kompozitai turi palyginus mažą tankį, didelį atsparumą medžio kenkėjams ir medį ardantiems grybas, stiprias garso izoliacijos ir geras šilumos izoliacijos savybes. Šie natūralūs (medžio) pluoštai yra lignoceliuliozinės medžiagos, kuriose yra nemažas kiekis lignino, hemiceliuliozės, cukrų ir ekstraktų, kurie stabdo cemento hidrataciją ir kietėjimą. Siekiant sumažinti medienos pluoštų sukeltą hidratacijos ir kietėjimo trukmių pailgėjimą, ceolito atliekos ir sintetiniai ceolitai dedami į medienos pluošto-cemento sistemą. Ceolitai yra pasirinkti šiame tyrime, nes jie pasižymi pucolaninėmis savybėmis, kurios gali būti naudingos sutrumpinant kompozicinės cemento sistemos kietėjimo laiką ir pagerinant mechanines savybes. Darbe tirti trijų rūšių medienos plaušų cemento kompozitai, pirmojo tipo kompozituose naudotas neapdorotas medienos pluoštas ir skirtingi ceolitinių atliekų kiekiai (0%, 1%, 2%, 5%, 10%), atsižvelgiant į cemento svorį; antrojo tipo kompozituose buvo 5% ultragarsu apdorotų medienos pluoštų ir ceolito atliekų (0%, 1%, 2%, 5%, 10%); trečiojo tipo kompozituose naudoti 5% ultragarsu apdorotų medienos pluoštų ir sintetinio ceolito (hidrosodalito), gauto iš silikagelio atliekų (0%, 1%, 2%, 5%, 10%). Iš gautų rezultatų nustatyta, kad 1 tipo kompozitų struktūroje su neapdorotais medienos pluoštais ir ceolito atlieka bandiniuose susidarė medienos pluoštų aglomeracija. Medienos pluošto cemento kompozitas su ultragarsu apdorotais medienos pluoštais ir 10% ceolitinėmis atliekomis (2 tipas) turėjo optimalius mechaninio stiprumo ir fizinių savybių rezultatus. Šiame tyrime buvo gauti lengvasvoriai kompozitai, kurių tankis svyravo nuo 1000 iki 1200 kg/m^3, naudojant apdorotą medienos pluoštą.

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Abbreviations:

FCC – fluidized catalytic cracking;

XRD – X-ray diffraction;

XRF – X-ray fluorescence;

SEM – scanning electron microscope;

ZW – zeolitic waste;

WF – wood fibres;

SZ – synthetic zeolite;

CFA – coal fly ash.

Introduction

According to EU sustainable goals of 'Responsible consumption and production' [1], it is essential to reduce waste materials production or re-use those materials. Zeolites and zeolitic wastes are used in this research to add to this goal. Utilization of waste materials like silica-gel by-product in the synthesis of zeolites; zeolitic waste obtained from fluidized catalytic cracking (FCC) process helps in the reduction of these residues and benefits the circular economy.

The main aim of the final degree project is to experimentally combine zeolites in wood fibres cement composites investigating its influence on the mechanical properties. Zeolites are introduced into the composite system to enhance wood cement compatibility. To study the results, an assessment will be carried out by comparing three types of wood-cement composites. In this research, wood fibres cement composites are selected as they are lightweight composites that have possible applications in the sound and heat insulation of buildings.

To achieve the goal of this work, the following tasks have been set:

- Review literature regarding zeolites, zeolitic-waste from fluidized catalytic cracking (FCC) process, building cement composites from wood-based materials and incorporation of zeolites in cement systems. From the existing research, study the experimental methodology including advantages, disadvantages of investigative methods and select the required quantity of materials to be added for optimal results in the experiment.

- Experimentally incorporate the selected amount of zeolitic waste (FCC), Portland cement and untreated wood-fibres producing the first type of building composites. Investigate its physical and mechanical properties to analyse the results.

- Experimentally incorporate the selected amount of zeolitic waste (FCC), Portland cement and ultrasonically treated wood-fibres producing the second type of building composites. Investigate its physical and mechanical properties to analyse the results.

- Hydrothermal synthesis will be carried out to transform waste silica gel (source of Si and Al) to synthetic zeolites. The properties of the obtained synthetic zeolites and zeolitic waste will be investigated by X-ray fluorescence (XRF) analysis, X-ray diffraction (XRD) analysis, Scanning electron microscopy (SEM) and finesse analysis.

- Experimentally incorporate the selected amount of synthetic zeolites (obtained from waste silica gel), Portland cement and ultrasonically treated wood-fibres producing the third type of building composites. Investigate its physical and mechanical properties to analyse the results.

This project work involves experimental research, to formulate conclusions on the utilization of zeolitic waste or synthetical zeolite (based on silica-gel waste) in building composites reinforced with wood fibres.

1. Literature review

The purpose of the literature review was to focus on the existing research in the field of zeolites, synthesis methods and their incorporation into cement systems including, the changes in the morphology of these cement pastes. Scientific research analysis was summarized on the main properties, characteristics and limitations of wood-based cement composites and zeolitic waste cement composites. All these topics were elaborately discussed and reviewed in this chapter.

1.1. Synthesis of zeolites from waste materials composed of silicon and aluminium compounds

Zeolites are synthesized from waste materials like fly ash, bottom ash or silica gel residues which are amorphous structures composed of mainly aluminium compounds, silicon compounds and other impurities (heavy metals) [2,3,17]. During the process of synthesis, these amorphous aluminosilicates present in the residual ash dissipate in solution (usually, alkaline), then they bundle, multi-nucleate and crystallize to form zeolites [11].

In this section, different processes of zeolite synthesis from raw materials like fly ash or silica gel by-product are analysed. It is noticed that each method can lead to different types of zeolite with certain limitations. Belviso [38] listed some of the main experimental methods used for zeolite synthesis namely: the hydrothermal conventional method; the microwave heating process; the alkaline-fusion assisted hydrothermal method; the sonication method along with a hydrothermal process; and the multi-step treatment process. Fig. 1 shows the schematization of different methods employed for the synthesis of zeolite.

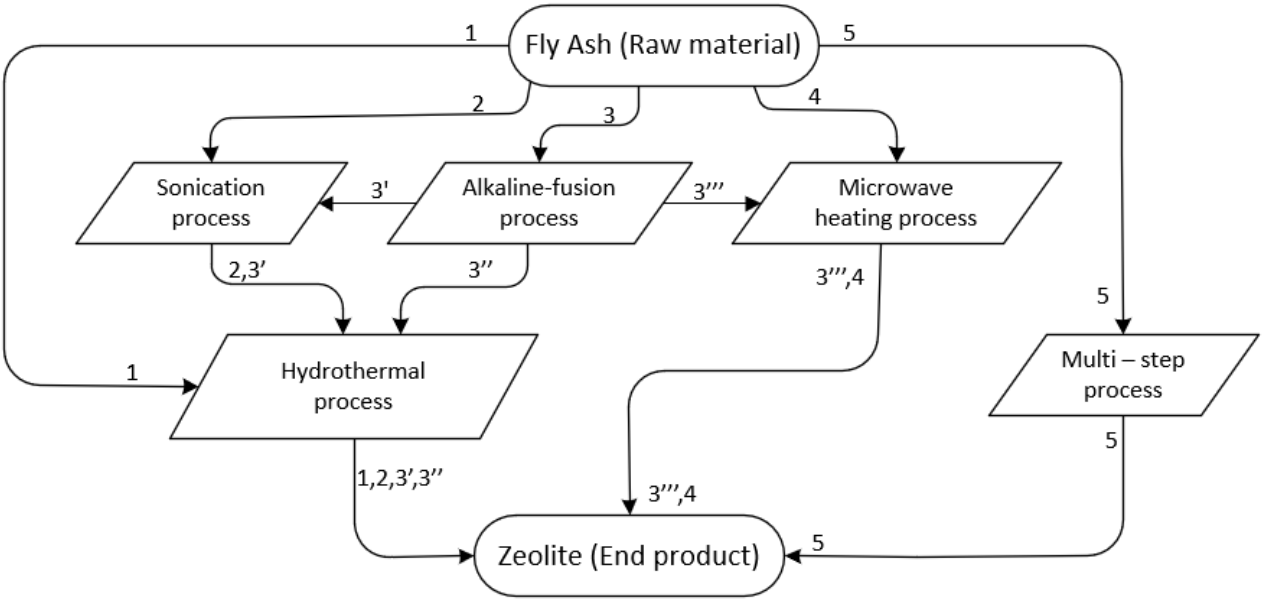


Fig. 1. Scheme of different methods of zeolite synthesis

In the synthesis of zeolite, many factors and conditions can be altered to achieve the crystallization of different types of zeolite. The stages of preparation of zeolites can be classified into the characterization of initial materials, the reaction of components and the crystallization of the required zeolitic phases [7].

Czuma et al. [2] investigated the possibility of obtaining zeolites through the use of by-product solutions from the previous synthesis. These solutions contain a significant concentration of alumina ions, silica ions and other heavy metal compounds. The study resulted in the crystallization of zeolite P1 and sodalite by using this type of cyclic two-step synthesis process; moreover, further tests proved that the produced zeolites can be used as sulphur dioxide sorbents. This paper provided new insights on the usage of post-production wastes of preceding synthesis, contributing to the ecological aspect of recycling materials.

Almost pure zeolite A with a degree of crystallinity of 92.52 %, cation exchange capacity of 5.14 meq/g, and specific surface area of 37.12 m^2/g was synthesized using alkaline fusion assisted hydrothermal process by Wulandari et al. [3]. The sequence of the synthesis process included the pre-treatment of coal fly ash, followed by the fusion with sodium hydroxide, which was then subjected to the ageing process and the final step was the hydrothermal treatment. The authors noted that the optimal time for hydrothermal treatment was 1 h with the Al/Si molar ratio of 0.75. It was additionally observed that when the hydrothermal time increased there was a reduction in the degree of crystallinity due to the rapid formation of zeolite A.

A comparative study was performed on the use of the alkaline fusion method and the alkaline fusion with ultrasonic dispersion method for the synthesis of zeolite X from coal fly ash. The process is illustrated in Fig. 2. The research implied that the crystallization temperature was one of the crucial factors in the synthesis, as it can influence the type of zeolite formed and the time taken for the aging step. It was noted that the crystallization time reduced from 24 h to 2 h with the use of ultrasonication approach [4].

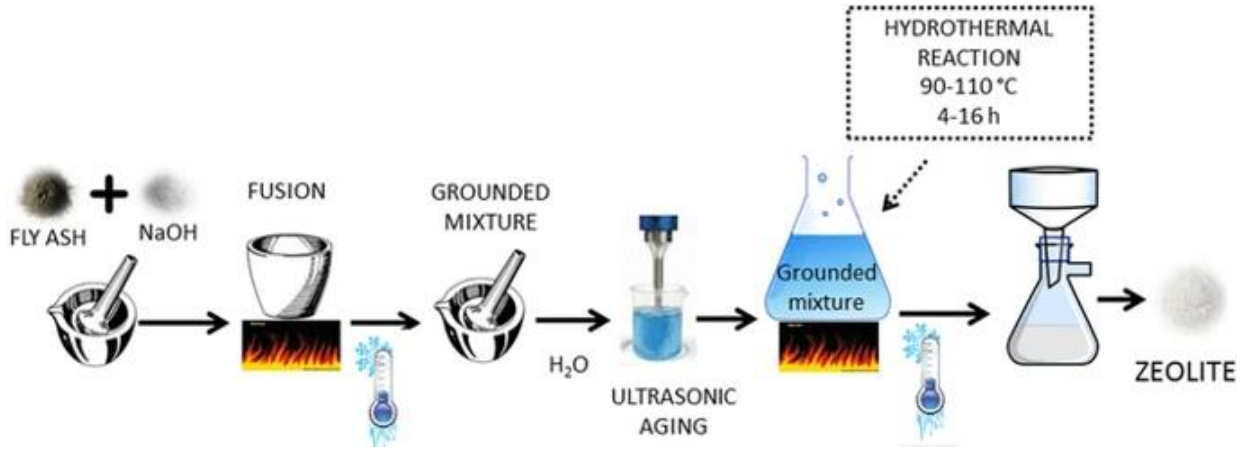


Fig. 2. Alkaline fusion followed by ultrasonic aging method [4]

As indicated by Belviso [38], during the conventional hydrothermal process, the fly ash is treated with alkaline solution (NaOH or KOH) of various concentration subjected to different temperatures (approx. 80-100°C) for varying time which turns the amorphous material in fly ash into the crystalline zeolite. Fukasawa et al. (2017) [5] performed hydrothermal synthesis by preparing an aqueous solution of incinerated biomass ash (10-15 g) in deionized water (50 ml) with constant stirring for 10 min then the solution was filtered; 0.14-1.12 g of potassium hydroxide was added to the extracted solution bringing the concentration to 0.05-0.4 mol/L. Then, 10 g of coal fly ash was added to the prepared solution which was heated at 160°C for 24 h with constant stirring. Then the end product was filtered, washed and dried at 100°C for 24 h. This experiment resulted in the successful synthesis

of K-zeolites which showed as a good adsorption material for caesium ions, pointing its possible future applications in the field of radioactive waste treatment. While Feng et al [6] performed zeolite synthesis using two-step hydrothermal method for which sodium hydroxide and coal fly ash were incorporated together with weight ratio of 0.87-2 and heated at 600°C for 2 h. The mixture was cooled and added to purified water that was subjected to rigorous stirring of 20 min; then the solution was heated at a temperature of 80°C for 24 h, after which it was washed, dried and filtered. This investigation lead to the production of highly crystalline zeolite A.

Belviso et al. [8] researched the influence of the ultrasonication step on the formation of zeolites during synthesis. For the experiment, 3 different types of fly ash were used, two sets of syntheses were performed, one was a traditional hydrothermal method and the other was a ultrasonication treatment followed by a hydrothermal method. In the traditional hydrothermal synthesis: coal fly ash was mixed with sodium hydroxide (NaOH) and heated for 1 h at 550°C (pre-fusion treatment), then products were powdered, added to deionized water and incubated for 4 days, the solid compounds were separated by a centrifuge and dried for 12 h at 80°C. In the sonication followed by hydrothermal method, the aqueous solution containing fly ash, sodium hydroxide in distilled water prepared in the same manner as the traditional method was subjected to ultrasonic dispersion for 1 h before the incubation period. In both the methods hydrothermal temperature for crystallization was ranging between 25°C to 60°C. The results of the experiments showed that the synthesis of zeolite X occurred at considerably low temperatures when it was subjected to 1 h ultrasonic dispersion before incubation than the products which were synthesized only with hydrothermal treatment. Since ultrasonic dispersion facilitates the supersaturation of Si/Al ions which increases the rate of nucleation of zeolitic phases. The authors concluded that the ultrasonic treatment can be used to lower temperatures for crystallization and reduce the issues of variable Si/Al ratios in fly ash.

The three main factors affecting the commercialization of zeolite synthesis are the time taken for the synthesis process, consumption of energy required for the process and the liability of harmful components release during the process. A study was performed to synthesize zeolites using thermo-sonical treatment to make the conditions of synthesis economical. For this experiment, an alkaline 1 M solution was prepared by adding 0.8 g of sodium hydroxide to 20 ml of distilled water to which 2 g of coal fly ash was added. The reaction was conducted in a controlled temperature environment using an oil bath with a constant temperature of 100°C, the prepared mixture was exposed to varying periods of hydrothermal process, ultrasound radiation and their combinations. It was noted that Na-P zeolite was synthesized in this investigation with the percentage of crystallinity of 87% at the optimal conditions of 1 h hydrothermal treatment assisted by 3 h of ultrasound radiation, due to the sonication the rate of nucleation increases leading to reduced time of crystallization [9].

Vaičiukynienė et al. [44] researched the synthesis of zeolites from by-product silica. The by-product silica was obtained from the industrial production waste of AlF_3. This study resulted in the crystallization of Zeolite Na-A with the highest degree of crystallinity of 25.81% in the presence of ultrasound waves of 20 kHz. The authors pointed out that the use of the sono-chemical method produced higher crystallinity of the obtained zeolites. A microwave-assisted heating synthesis of CFA was carried out by Tanaka et al. [10] which involved two steps: the treatment of fly ash by refluxing under microwave heating followed by the hydrothermal treatment. In these investigations, the authors in the first step, subjected the fly ash to microwave heating for 1 h after which the fly ash was added to NaOH-$NaALO_2$ solution with Si/Al molar ratio of 0.4-10. The dissolved solution was incubated for 24 h at 85°C. Na-A zeolite was obtained at the end of the process which showed good results in

eliminating heavy metals such as Cd, Pb, Zn. The crystallization rate of zeolite was found to be higher due to the pre-treatment of CFA in microwave which induced faster dissolution of ions.

1.2. The effects of zeolite on hardened cement systems

Zeolites are usually used in the cement systems for their effect as a pozzolanic substance, due to the presence of large amounts of aluminosilicates which react with water and $Ca(OH)_2$. A study was conducted to investigate the properties of hardened cement paste with the addition of hydrosodalite. In the experiment, three samples of blended cement were prepared by adding hydrosodalite powdered (0%, 5% and 15% of cement weight) and superplasticizer (2% of cement mass) to the Portland cement CEM I 42.5R. The water/cement ratio was maintained as 0.38 for all samples. The results of the experiment showed that as the amount of hydrosodalite increased there was a steady decrease in the density and consequently the compressive strength of the hardened cement samples with an increase in the time taken for cement hydration. The authors illustrated that the decrease in strength of hardened cement samples was due to the varying micro-structures of the different samples [12].

Nagrockiene, Girskas and Skripkiūnas [13] studied the effects of synthetic zeolites in cement systems subjected to cycles of freeze-thaw tests. The authors used synthetic zeolite which was obtained from a combination of aluminium fluoride (AlF_3) manufacturing by-product, sodium hydroxide compound (NaOH), aluminium hydroxide compound $Al(OH)_3$ altered with a solution of calcium chloride ($CaCl_2$) synthesized at 105°C. Hardened cement samples were made by mixing CEM I 42.5 R (w/c of 0.27) with synthesized zeolite (0%, 5% and 10% of cement weight) and superplasticizer (0.5% of cement weight) , the dimensions of samples were 40 x 40 x 160 (length x width x height) mm. The above investigations concluded that when 10% of synthetic zeolite was added to the samples: (i) during 28 freezing-thawing cycles showed maximum reduction in mass loss. (ii) ultrasonic pulse-velocity test achieved 446 m/s after freeze-thaw cycles due to the increase in concrete density. Additionally, the authors noticed that there was a change in the hydration products formation due to the added zeolite, which decreased the amounts of calcium hydroxide and ettringite present, while increased the C-S-H gels. This made the cement composite effectively improve strength, frost resistant, and density. It was also found in X-ray analysis that a layer of $CaCO_3$ formed at the top of the samples which prevented the attack of salts providing durability to the cement composites.

Further Girskas and Skripkiūnas [14] continued their research on synthetic zeolite and their freeze-thaw resistance and microstructure, this time the same synthetic zeolite was obtained (from AlF_3 by-product, NaOH and $Al(OH)_3$) same as previous paper but the temperature of synthesis was reduced to 95°C. In this experiment, the scientists also studied the effect of adding air-entraining agent (a substance used to create tiny voids of air in concrete/cement) to the cement system with synthetic zeolite. The outcome of this experiment revealed that: the water adsorption value of samples with the absence of air entraining mix decreases with an increase in zeolite percentage added but for the samples with air entraining mix the absorption value increased because of the new air pockets created which allow the permeability of water in them; for samples with 10% zeolites with air entraining agent - the amount of pores increased 1.9 times, the distance between pores reduced from 190 μm to 130 μm, the surface loss during free-thaw cycles reduced 3.5 times and without air entraining agent - the amount of pores decreased 4 times, pores distance decreased from 360 μm to 190 μm, while surface loss during free-thaw cycles reduced 1.6 times. Therefore, the reduction in the mass loss indicated that the durability of the cement system resistance towards the free-thaw cycle increased.

A study was performed to use zeolite A as an additive material to cement mortars, which incorporated zeolite A in 5%, 20% and 30% by cement weight. The water/cement ratio was maintained at 0.4 for all sample mortars. Portland cement (OPC-I) was mixed with zeolite A and poured into moulds of size 50 x 50 x 50 mm which were cured in atmospheric pressure and room temperature. After testing of the cement blended mortars for the compressive strength, it was found that with an increase in amount of zeolite A in the samples there was a decrease in the compressive strength of samples. The author explained that zeolite A addition to cement lead to a reduction in the calcium hydroxide or portlandite $Ca(OH)_2$ which accelerated transformation of semi-amorphous calcium hydrosilicates into monosulfate and monocarbonate phases which could be the reason for the decrease in compressive strength of the hardened blended cement mortar [15].

An experimental investigation was conducted to analyse the significance of hydrosodalite in cement composites. The experiment involved using modified hydrosodalite in cement composite system, the hydrosodalite was synthesized in laboratory with temperature at 105 °C and modified through ion exchange process with calcium chloride where Na^+ gets replaced by Ca^{2+} ions. The cement samples were made with W/C of 0.55 including the above altered hydrosodalite in 0%, 5%, 10%, 15% and 20% by cement mass. The composite mix was added to a mould of 20 x 20 x 20 mm. Pozzolanic activity was observed in the hydration of the cement and modified hydrosodalite which achieved low number of voids and greater compressive strength with an increase in the percentage of hydrosodalite. In the analysed results, it was pointed out that the maximum compressive strength of the cement composite was found for samples with the addition of 15% of zeolite while the lowest compressive strength was observed for samples with the addition of 5% of modified hydrosodalite [16].

Vaiciukyniene et al. [17] studied the reaction of synthetic zeolites specifically zeolite A with Portland cement systems. Two types of zeolites were synthesized from the residue of aluminium fluoride manufacturing process, for the production of one type of zeolite the residue was mixed with water and treated to heated suspension at 110°C for 240 min. The second type of zeolite was produced in which the residue was grinded in mechanical mill for 15 min and dried at 80°C for 24 hr. With the Chapelle test the authors evaluated the two synthesized zeolite A and the test showed significant pozzolanic reaction between the zeolites and cement (CEM I 52.5 R) in the composite. Zeolites were added to the Portland cement in percentages of 0;5;10 different for each type of the zeolite. The graph drawn from the compressive strength recorded in tests of all the samples revealed that the composites with the addition of 5% of zeolite type 2 had the maximum compressive strength than other samples. It was also noted that the compressive strength of such sample was 9% more than the control sample. This experiment points further application of zeolites as cement replacement material which has environmental and economic benefits.

A paper was published on the influence of zeolite synthesized in laboratories on the cement viscosity in which three main components of the composites was taken as cement, synthetic zeolite and superplasticizer. The synthetic zeolite was obtained in the same manner as in the previous paper [13]. Zeolite was added with 0 %, 5% and 10% of cement mass, amount of superplasticizer remained the same in all samples with water/superplasticizer ratio 0.27, the water/cement ratio varied in all 3 samples taken as 0.27, 0.28 and 0.30 respectively. The value of slump which is taken as the mean value of spread in two perpendicular directions reveals the consistency of the fresh cement composite paste. The results of the experiment informed that the samples with 0% of zeolite had consistence of 149 mm and after 30 min viscosity was noted to be 81.1 mPa s, 5% of zeolite had slump/consistence of 85 mm and after 30 min viscosity was noted to be 746.5 mPa s and 10% of zeolite had slump of

70 mm and after 30 min viscosity was noted to be 827.1 mPa s. The results illustrate the linear influence of zeolites in terms of cement viscosity i.e. there is a gradual increase in viscosity with the increase in zeolites. This study has future applications for researchers who aim to produce specific type of consistence of cement composites with the addition of zeolites [18].

Jakevicius et al. [19] studied the influence of using ultrasonication on the temperature of hydration of hardened Portland cement paste which contained synthesized amorphous zeolite. The samples contained cement with zeolites 0%, 5%, 10% and 15% amount by cement weight. The hardened cement pastes were subjected to ultrasonic irradiation 28 kHz for a duration of 15 min using a transducer of 60 Watts-power. Highest temperature in the hydration process was recorded for the samples with 5% zeolite which was subjected to ultrasonication. This proved the initial hypothesis that cements with zeolites subjected to ultrasonic radiation will lead to an acceleration of the exothermic reaction in the hydration of cement pastes.

1.3. Cement composites containing wood-based materials

In chemistry, wood is a polymer-based material made up of many components like cellulose, lignin, hemicellulose, extractives and other inorganic substances. Cellulose consists of several glucose units which are insoluble in solvents like water, alkaline and other organic solutions. Lignins are aromatic amorphous polymers which like cellulose do not dissolve in various solvents but can be combined with hemicellulose forming complex carbohydrates. Hemicellulose is a polymer which contains different types of sugars, unlike cellulose, they dissolve in alkali solvents and rarely in water, they release additional extractives. Extractives present in the wood are natural non-polymeric substances which can be easily dissolved in water and other solvents. One of the main inhibitors in lignocellulosic cement systems during cement hydration was researched to be soluble sugars, hemicellulose, and extractives (starches and tannins). Scientists have established that increase in the number of extractives in concrete or other composites which include alkali solutions hinder the hydration of products reducing the setting time [22, 23].

Ashori et al. [20] performed a study to make wood wool cement boards with two types of wood, eucalypt and poplar. Before making the composites, eucalypt and poplar were treated with water at temperature of 50°C for 2 days, then dried, cut and shaved to form wood wool particles with dimensions of 100-250 mm length, 1-5mm width and 0.2-0.5 mm thickness. These treated wood-wools were mixed with cement by weight ratio of 40:60 and 60:40, a series of compositions were made with varying amounts of eucalypt and poplar wood-wool 100:0; 80:20; 50:50; 20:80 and 0:100 respectively, and calcium chloride 5% and 8% was additionally added to the samples. The samples underwent procedures to be pressed and assembled into cement wood-wool composite boards. The mechanical properties of the formed boards were investigated which revealed that the addition of wood-based material inhibits the process of hydration by reducing the maximum temperature of hydration and increasing the time of hydration. In general, cement wood-wool composite boards made of poplar wood had better mechanical properties than eucalypt which could be due to fewer extractives in poplar. It was also stated that 5% of $CaCl_2$ enhanced the mechanical properties of the composites.

In another study, cement wood-wool boards were prepared by mixing ordinary Portland cement with wood-wool in the ratio of 2:1 and water/cement ratio was taken as 1:1. The aim of this study was primarily to develop low-density cement wood-wool boards which will provide efficient sound and

heat insulation, secondly to investigate the influence of wood-wool size (1.5 mm, 2.5mm, and 3.5mm) on the cement composite boards. Before making the composite boards, the wood-wools were treated to remove soluble sugars and redundant extractives from wood, this was done by soaking the wood-wool in room temperature for 1 day, then dried up to moisture content of approximately 12%. The results of this research laid out that the mechanical strength of the wood-wool cement boards was found to be directly proportional to the density and the density was indirectly proportional to the wood-wool size i.e. as the size of the wood-wool increased, the density and the strength decreased significantly. The compressive strength of the wood-wool cement composite boards ranged from 0.02-0.11 MPa and the highest flexural strength and elasticity was noticed for boards with 1.5 mm size of wood-wool [21].

Vaickelionis et al. [23] researched the harmful effects of wood extractives. In this investigation a pozzolanic substance - carbonated opoca was added to two different Portland cements to determine the influence of extractives in the cement composites. After the completion of the experiments the authors summarized that the extractives in wood saw dust dissolved in alkali environment producing more of these extractives; the soluble sugars in cement inhibit hydration process. The addition of pozzolanic substance (carbonated opoca) decreased this effect because the pozzolan adsorbed the soluble extracts, this property depends on their surface area. It was pointed out that in this experiment the setting time of cement hydration depended not on the type of wood species used but on the concentration of wood extracts present.

To avoid the conflicts with wood-cement compatibility research is being done to treat the wood prior to forming composites. The conditions to which the wood is subjected plays an important role in the mechanical properties of the wood-cement composites. A study was performed on sap wood fibres (from Poplar species of wood) to be added in Portland cement system, to be treated in three different methods water extraction, alkaline hydrolysis degradation and retention of redundant extractives. In the water extraction treatment was done by adding wood fibres to a water bath at 25°C, 62.5°C and 100°C with duration of 5 min, 25 min and 45 min respectively, water/wood ratio was kept at 20,30 and 40. In the second treatment, wood fibres were added to $Ca(OH)_2$ solution of 5.6 g/l, 11.1 g/l and 22 g/l for a duration of 4 h, 24 h and 72 h, here water/wood ratio was kept same as water extraction. In the third method, primarily, a coating agent (styrene) was used to form a thin layer on the surface of wood fibres then it was immersed in water for 5 min, water/wood ratio was kept 26.67 for all. The wood fibres were dried before using in the synthesis of composites. For the synthesis of the wood-cement composites water/cement ratio was kept as 0.93, here 840 cm^3 of water was added per 900 g of Portland cement, to this paste 450 g of wood fibres was immersed and mixed thoroughly before adding to the moulds of dimensions 300 x 300 x 70 mm. The wood cement paste was not vibrated due to the need of a homogeneous paste because the vibration might lead to the cement settling at the bottom. After performing tests on the formed wood fibres cement composites, it was established that the compressive strength of the wood-cement composites where, wood was treated by the water extraction and the coating agent was higher than the composites with the alkaline treatment. However, the highest compressive strength of 0.15 MPa was achieved through the hydrolysis treatment. The authors hypothesized that this decrease in the strength maybe due to the alkaline medium which helps in the disintegration of soluble sugars, reducing the bond between cement and wood surfaces. It was indicated that compressive strength for composites with wood treated by water extraction was almost same for all, approximately 0.10 MPa. This relationship between the treatment of wood and mechanical strength can be interpreted as the dependence of fibres interaction on the immersed

solution being favourable or non-favourable. For wood-cement composites, the elasticity properties was directly proportional to the water/wood ratio and decreased with duration of immersion in coating agent medium. Therefore, all the treated wood fibres composites had higher modulus of elasticity than those of untreated wood fibres. It is recommended to use different wood treatment approaches based on the need, for stiffening structures hydrolysis method could be used, while for coating method could be used for acoustic insulation of structures [24].

1.4. Zeolitic waste incorporated in cement systems

A great deal of scientific research is carried out in the utilization of residues from fluidized catalytic cracking (FCC) process which is employed in the oil refinery industry to transform crude oil into gasoline and other fuel products. Zeolites are used as catalysts in the process of fluidized catalytic cracking (FCC) which helps in the breakdown of hydrocarbons to produce fuel products, after many cycles this catalyst becomes spent in the end. The spent fluidized cracking catalyst can be also known as zeolitic waste which is produced after many cycles of refinery procedures containing high amounts of silicon dioxide (SiO_2), aluminium oxide (Al_2O_3), heavy metals and other organic impurities. This waste generated is highly pozzolanic in nature [25,26,27,28].

Hassan et al. [26] investigated the impact of adding these synthetic zeolites catalyst in the production of green cement (eco-friendly cement) to reduce the CO_2 emission during the process. The synthetic zeolites used in the experiments were obtained as the residual waste of fluidized catalytic cracking (FCC) process with compositions of 1, 3, 5, 7, 10% by cement mass. Results obtained from the experiments showed that the substitution of cement clinker with zeolite waste from (FCC) up to 10% acts as a pozzolanic material without any adverse effects to the properties and characteristics of the cement. This research is useful in broadening the residual FCC application fields, showing it can be useful to form green cements.

A paper is written on the effects of using treated waste fluid catalytic cracking catalyst (FCCC) in Portland cement system. This waste product (FCCC) contains many organic pollutants which are known to inhibit the hydration process to eliminate such impurities it was treated with a clear solution of 17% hydrogen peroxide (H_2O_2) for 1 day after that subjected to centrifugation and dehydrated in an oven at 100 °C. Two types of samples of hardened Portland cement pastes were formed of dimensions 20 x 20 x 20 mm, one with treated waste FCC catalyst and other with waste FCC catalyst only. The water/cement ratio was 0.4 consistent for all samples, and the proportions of added purified waste FCCC and waste FCCC was 0%, 10%, 20% and 30% of cement mass. After 28 days, the samples were tested for compressive strength it was observed that all samples with treated waste fluid catalytic cracking catalyst had developed higher compressive strength than the samples with non-treated waste FCC catalyst. Regarding the amount of replacement it was pointed out that all treated and non-treated waste FCC catalyst with 10% and 20% zeolites had higher strength which decreased with 30% zeolites and ultimately, the highest compressive strength of 65.25 MPa was noted for 10% purified waste FCC catalyst whereas the control sample with 0% zeolite has only reached the strength of 46.80 MPa. The authors mentioned that the portlandite released during hydration process undergoes a pozzolanic reaction with the added treated and non-treated waste FCC catalyst producing cement equivalent products responsible for the increased strength in these blended cements. This study indicates that the usage of wastes from FCC catalyst in Portland cement system leads to an increase in the strength of these samples. Purifying such waste FCCC with the use of oxidizers like hydrogen peroxide improves the results of the compressive strength significantly [25].

Pinto et al. [27] studied the residual fluidized cracking catalyst (FCC) pozzolanic activity during the initial levels of the hydration process of cements. For the experiment, Portland cement was mixed with calcium carbonate and spent residual catalyst with water/materials ratio of 0.5 and replacement of zeolites was 0, 5, 10, 15, 20, and 30% of cement mass. In conclusion, three systems of thermal analysis was conducted on the cement-zeolitic waste pastes: (i) By use of non-conventional differential thermal analysis (NCDTA) which works on the same principle as DTA except there is no supply of heating/cooling to the system, it was observed that the increase in replacement of cement with the residual waste catalyst up to 30% yielded increase in the formation of ettringite and portlandite in the cement composites pastes. (ii) Thermal analysis (TA) technique showed that the increase in the amount of waste FCC catalyst depleted higher amounts of calcium hydroxide indicating high pozzolanic activity after one day of hydration. (iii) Differential scanning calorimetry (DSC) also showed an increase in the pozzolanic activity by the increase in zeolitic residual waste replacement in cement. Through this study it is evident that the residual catalyst behaves as a pozzolan in reaction with cement systems accelerating the setting time significantly.

According to a study performed to find the durability properties of concrete with added residual spent fluidized cracking catalyst. The composition of the concrete included cement (280 kg/m^3), residual FCC catalyst of 15% by cement mass, corrosion retarder dimethylaminoethanol (12 kg/m^3) and cement/water ratio of 0.7. It was observed that 15% of spent residual catalyst enhanced the chloride resistance of the concrete composites whereas, reduced the carbonation resistance. The added corrosion inhibitor showed positive impact on the chloride attack and the carbonization of concrete. However, there was no synergic effect of the corrosion retarder and spent FCC zeolites. The author summarized that the spent fluidized catalyst can be used as a supplementary cement material in the concrete with regards to durability enhancement of concrete, there is a lack of information on the reasons of this correlation [28].

Allahverdi et al [29] researched the physical and the mechanical properties of Portland cement pastes with added spent catalyst residue (FCC) as well as the pozzolanic behaviours of the spent residue FCC. To study the pozzolanic activity, micro silica and metakaolin were used as control pozzolans. From the lime-combinability test, spent catalyst residue exhibited the strongest pozzolanic behaviour after micro silica better than metakaolin therefore, the spent catalyst residue can be applied as pozzolanic cement supplementary material. The experimental compressive strength tests showed that the replacement of cement with zeolites residual FCC by 15% and 20% of cement mass increased the 360th day compressive strength up to 108 MPa which is 45% more than the control samples which had the compressive strength of 75 MPa. On the 28th day cured samples, the addition of residual FCC increased the water adsorption, exposed pores volume, and compressive strength of the hardened Portland cement paste, usage of this material effects the paste workability and setting times which decreased with an increase in percentage of cement replacement by zeolites.

In another research, Izquierdo et al. [30] prepared hardened cement pastes with and without adding waste FCC, metakaolin and silica fumes and studied the hydration mechanism of such pastes. The compositions of the hardened Portland cement paste were made by adding waste FCC 0%, 10% and 20% of cement weight, and for reference samples metakaolin and silica fumes of 10% were added. After subjecting the samples to XRD, TG, SEM and NMR characterization techniques, the major hydration products for Portland cement blended with waste FCC were calcium silicate hydrate (C-S-H), hydrated calcium aluminium sulphate hydroxide (ettringite), calcium aluminate hydrates (C-A-H) and hydrated calcium aluminosilicates (C-A-S-H). The pozzolanic reactivity of waste FCC 10%

blended cements was indicated by the consumed lime of 61% at 360th day curing exceeded the values of metakaolin and silica fumes blended cement pastes of the same percentage. The author noted that the addition of waste FCC 10% up-to 30% increased the strength of the hardened blended-cement pastes indicating its future application as cement replacement material. Castellanos et al. [31] performed investigations on the mechanical properties of cement pastes with added residual FCC of percentages 10, 20 and 30, and for control samples cement with and without addition of 20% metakaolin was used. From the results it is evident that samples with residual FCC 10% had compressive strength and modulus of elasticity higher than the control sample with and without metakaolin due to the early pozzolanic reactivity. The best mechanical characteristics was noted for sample with 10% FCC waste as cement supplementary material than others and these characteristics decreased with an increase in the waste FCC percentages.

1.5. Conclusion of literature review

Through the scientific research analysis, it is evident that there exists a research gap where zeolites/zeolitic wastes which exhibit pozzolanic activity can be added to wood fibres-cement composites. To observe the influence of zeolites on the compatibility of wood-cement composites, will be crucial to determine positive or negative changes in the properties of these composites. This research will provide a basis for future studies on this type of wood-fibres cement composite system.

2. Experimental investigation methods and materials

The main task of the experiment is to investigate the influence of zeolites on building composites reinforced with wood fibres. The experimental samples are analysed using several investigation techniques to characterize and compare the results of these tests. It is essential to understand and employ the investigative methods which are relevant to this research. The examination of the obtained results will determine the usage value of such composite systems.

2.1. Investigation methods

To achieve the objectives of this project, the following investigation methods were employed.

2.1.1. X-ray fluorescence (XRF) analysis

X-ray fluorescence analysis was carried out to determine the chemical composition of initial materials like zeolitic waste, Portland cement and silica gel by-product. The spectrometer used for the investigation was Bruker S8 Tiger series 2, which consists of rhodium (Rh) tube with anode voltage up to 60 kV and electric current up to 130 mA. The pressed sieved samples were measured in helium atmosphere using SPECTRA Plus QUANT EXPRESS method [34].

2.1.2. X-ray diffraction (XRD) analysis

X-ray diffraction analysis of the crushed sample materials was carried out using the D8 Advance diffractometer (Bruker AXS) operating at the tube voltage of 40 kV and tube current of 40 mA. The X-ray beam was filtered with ferromagnetic metal (Ni) 0.02 mm filter to select the CuKα wavelength. The specimens were scanned over the range $2\theta = 3–70°$ at a scanning speed of 6° min^{-1} using a coupled two theta/theta scan type [35].

2.1.3. Determination of particle size distribution

A laser particle size analyser CILAS 1090 LD was used to determine the particle size distribution and the specific surface area of the materials in intervals from 0.04 μm to 500 μm. The distribution of solid particles in air stream was approximately 12-15 wt.%, where compressed air (2500 mbar) was used as a dispersing phase. The measuring time for the experiment was 15 s [36].

2.1.4. Scanning electron microscope (SEM) analysis

To study the morphology and compositional microstructure of the initial materials as well as the hardened composite samples, a high-resolution scanning electron microscope (SEM) ZEISS EVO MA10 was used in this work [37].

2.1.5. Determination of pH

The pH measurements of the hardened samples - water suspensions were conducted by using pH-meter Adwa AD8000 (200V) with an accuracy at 20°C of ±0.01 pH [32]. The water suspensions of the samples were prepared with the ratio of water (W) and solid material (S) W/S taken as 10.

2.1.6. Determination of water absorption

Water absorption analysis was performed on two types of hardened wood fibres cement composites (Type-2 and Type-3). Firstly, all the hardened samples were dried in the oven at 50°C for 24 h, then

they were weighed (initial mass). After that, the dried samples were added in water for about 24 h, then they were removed from the water and weighed (final mass). The percentage of water absorption capacity of the hardened composite samples was determined by calculating the difference in the measured mass of the samples.

2.1.7. Determination of semi-adiabatic calorimetry

The hydration temperature curves of two types of composite cement pastes was measured with the 8-channel USB TC-08 Thermocouple Data Logger. The temperature accuracy for this device was found to be ±0.2% of recorded readings and ±0.5°C of recorded temperatures [39]. The experimental setup for the calorimetry analysis is illustrated in Fig. 3.

Fig. 3. Semi-adiabatic calorimetry experimental setup

2.1.8. Determination of compressive strength, flexural strength and density

The compressive strength and flexural strength of the hardened cement samples was tested after 28 days of curing, according to LST EN 196-1:2016. Toni technik 2020 (Fig. 4) is a fully automated press machine with computerized control which was used to compress the specimen cubes. It has the load capacity from 0 till 600 kN [40]. Three-point bending strength test was performed for the samples using the same machine with a different setup. The density of the samples was determined according to the standard LST EN 12390-7:2019.

Fig. 4. Toni Technik 2020 hydraulic press machine

The compressive strength of the samples is calculated according to Formula (1):

$$\text{Compressive strength} = \frac{F}{A}, \quad (1)$$

Here: F – applied force at which specimen fails, N;

A – area of the specimen, mm^2.

The flexural strength of the samples is calculated according to Formula (2):

$$\text{Flexural strength} = \frac{3 \cdot Fl}{2 \cdot bh^2}, \quad (2)$$

Here: F – applied force at which specimen fails, N;

l – length between the bottom supports, mm;

b – breadth of the specimen, mm;

h – height of the specimen, mm.

The density of the samples is calculated according to Formula (3):

$$\rho = \frac{m}{V}, \quad (3)$$

Here: ρ – density of the specimen, kg/m^3;

m – mass of the specimen, kg;

V – volume of the specimen, m^3.

2.1.9. Determination of ultrasonication treatment

In this study, the wood fibres of Type-2 and Type-3 composites were treated by using sonication treatment. The ultrasonic homogenizer equipment SONOPLUS electronic UW3400 manufactured by the company Bandelin was used as shown in Fig. 5. The procedure for ultrasonic treatment lasted for 1 min using 20 kHz and 250 W power ultrasonic waves.

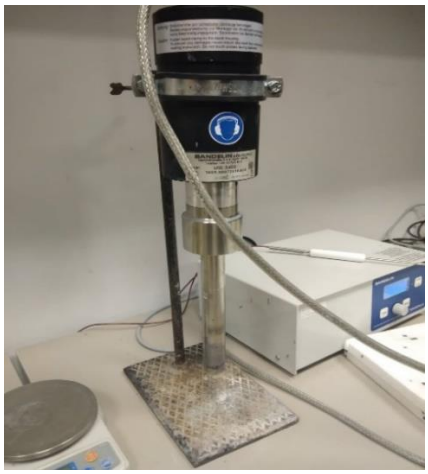


Fig. 5. Equipment used for ultrasonic treatment

2.2. Initial materials

In this section all the raw materials required for the experimental research are explained in detail along with their characteristics.

2.2.1. Zeolitic waste

In simple terms, zeolitic waste can be described as a by-product of oil refining industry, it is obtained when zeolites are used as catalysts in the fluid catalytic cracking (FCC) process to transform crude oil into gasoline and other fuel products [24,26]. The quantity of the residual zeolitic waste of spent FCC catalyst is gradually increasing due to the current expansion of the petroleum (crude) refining industry. Therefore, it is crucial to reduce the quantity of this waste material to improve its environmental impact. Many scientists are now employing this residual FCC catalyst in building materials. To enhance properties of the composite mortars or concrete, thereby producing eco-friendly or green concrete. The chemical composition of the spent FCC catalyst (zeolitic waste) depends on the type of the petroleum refining process and the manufacturer.

For this study, the residual spent fluid catalytic cracking (FCC) catalyst was used as zeolitic waste. This zeolitic waste contains high amounts of silicon dioxide (SiO_2), aluminium oxide (Al_2O_3), and smaller amounts of carbon, titanium, magnesium, lanthanum and other impurities. The chemical composition of the zeolitic waste is presented in Table 1 which was measured according to X-ray fluorescence (XRF) spectrometer.

Table 1. Chemical composition of zeolitic waste

Element	Amount, wt.%
Oxygen	54.51
Carbon	3.3
Aluminium	23
Silicon	16.18
Magnesium	0.44
Sulphur	0.15
Sodium	0.07
Iron	0.81
Titanium	0.39
Lanthanum	1.15

The compositions of the prepared hardened cement paste samples varied in the contents of zeolitic waste as supplementary material: 0%, 1%, 2 %, 5%, and 10%. The characteristics of the particle size distributions of zeolitic waste mixture is presented in Table 2. For this material the mean diameter was found to be approximately 97.95 μm with the lowest particle size of about 19.80 μm.

Table 2. Characteristics of the powder mixtures obtained from particle size distributions

Material	d90, μm	d50, μm	d10, μm	Mean diameter, μm
Zeolitic waste	177.81	90.91	19.80	97.95

The particle size distribution histogram is illustrated in Fig. 6, for this distribution the particle diameter of zeolitic waste ranges from 0.1 μm to 450 μm. Through the investigation of scanning electron microscope, it can be noted that the microstructure of zeolitic waste is predominantly round and oval shaped as shown in Fig. 7.

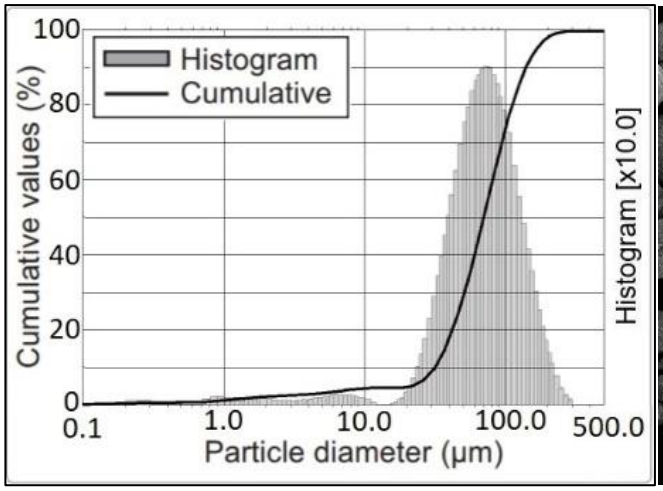


Fig. 6. Histogram of particle size distribution of zeolitic waste

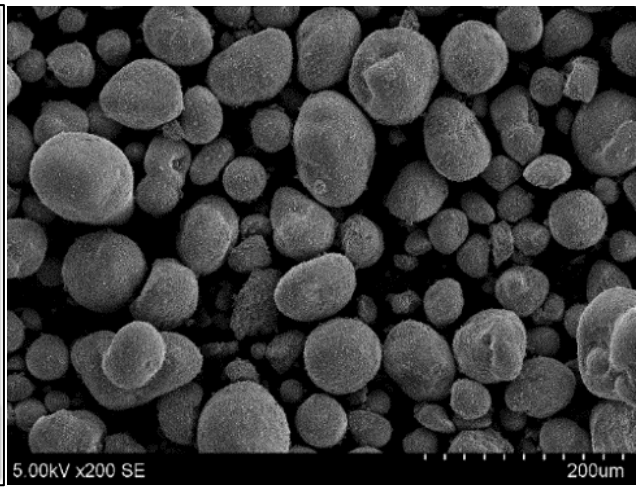


Fig. 7. Scanning electron micrograph of zeolitic waste

The X-ray diffraction peaks of the zeolitic waste show, zeolite-Y dominating in the waste FCC catalyst as presented in Fig. 8. Zeolite Y is noted as a porous aluminosilicate having a high surface area with a crystalline structure and a geometry consisting of tunnels and cages.

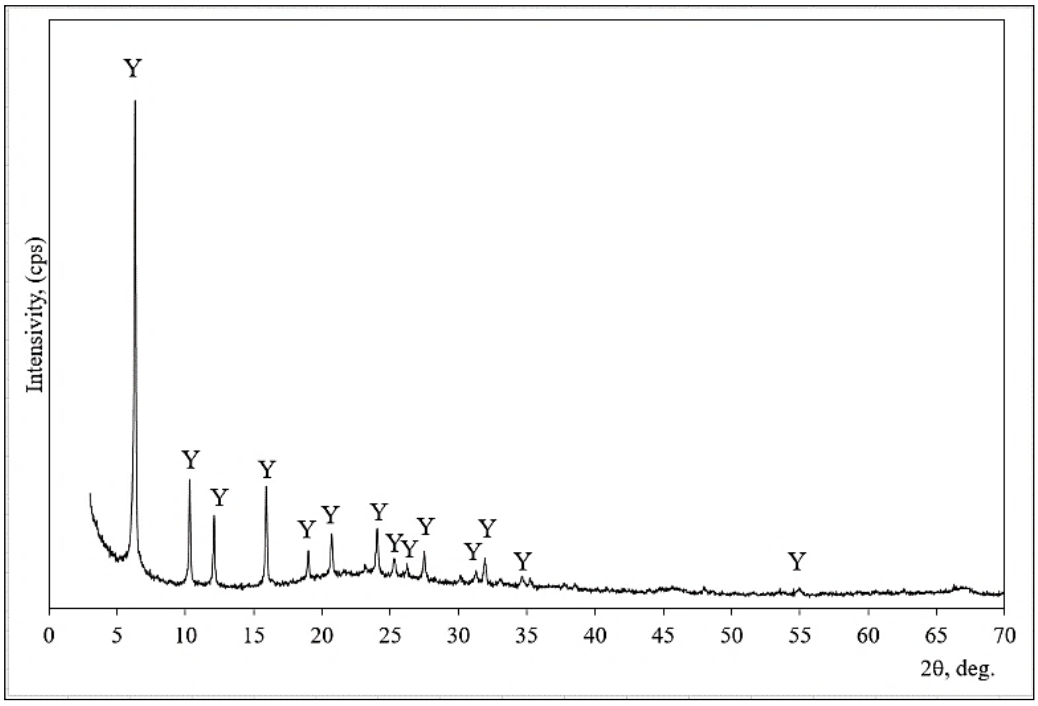


Fig. 8. X-Ray diffraction pattern of zeolitic waste. Notes: Y is zeolite $Al_{60.352}{\cdot}Si_{139}{\cdot}O_{371.52}{\cdot}H_{5.984}$ (73-2313)

2.2.2. Ordinary Portland cement

In this study, commercial ordinary Portland cement type CEM I 52.5R, which conformed to the European standard EN 197-1:2011, was used as a binding material for the preparation of the samples. The notations in the cement type 'R' and '52.5' correspond to a high reactivity at the early stages of hydration and a minimum compressive strength of 52.5 MPa observed on 28th day respectively. The particle size distribution of the Portland cement is plotted on Fig. 9.

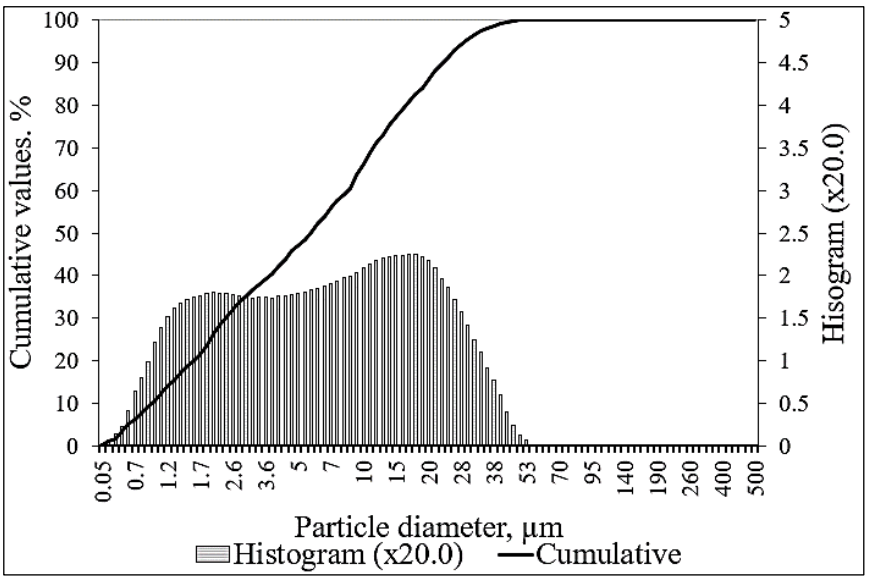


Fig. 9. Particle size distribution of Portland cement CEM I 52.5R

Portland cement CEM I 52.5R can be categorized as a hydraulic cement with the mineral composition C_3S = 50.7 %; C_2S = 18.5 %; C_4AF = 14.2 %; C_3A = 9.7 %. The chemical composition of the Portland cement is given in Table 3 which shows that the highest amount of oxides present in this cement are calcium oxide (CaO) and silicon dioxide (SiO_2). The amount of these oxides present in the cement impacts different properties of the cement.

Table 3. Chemical composition of Portland cement CEM I 52.5R, wt.%

Compound	Amount, wt.%
SiO_2	21
Al_2O_3	3.91
Fe_2O_3	2.91
MgO	2.71
CaO	66.01
SO_3	3.4
Cl	0.06

2.2.3. Water

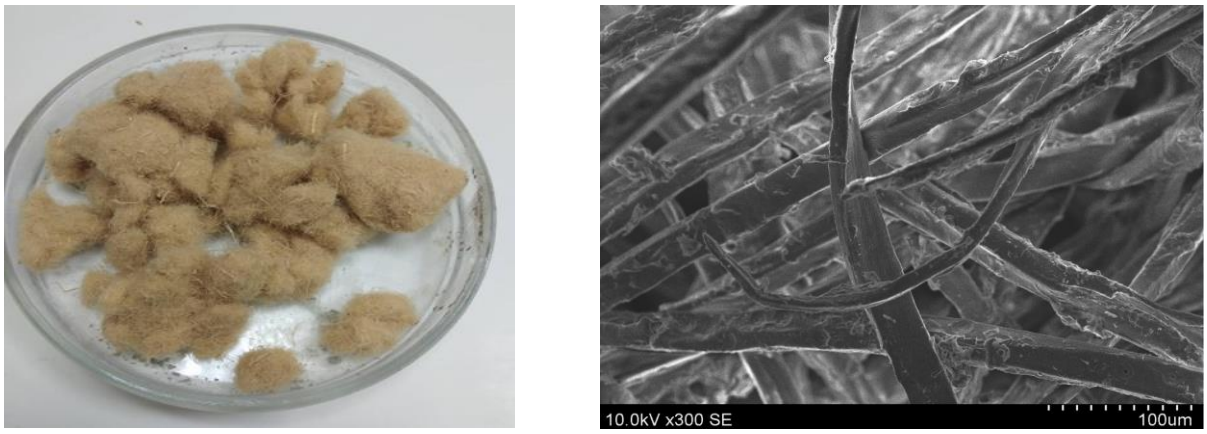
Water (H_2O) or deionised water with no impurities was used in the experiments. It is essential to make sure there are no heavy metals included in the water as they can be harmful to the composite

cement systems. The water/cement ratio was maintained as 0.66 for the first and the second type of wood-cement composites.

2.2.4. Wood fibres

The wood fibres (Fig. 10) used as reinforcement material in this study are commercially produced "Steico-zell" (wood fibres) obtained from the manufacturing company STEICO, Germany [41]. These wood fibres are used as building insulation material and have possible applications in:

- Prefab walls
- Timber roofing cassettes
- Insulating material for timber elements
- Insulation for renovation works (walls, roofs, floors)
- Joint free insulation
- Interlocking system of wood fibres provides good slump resistant insulation



a b

Fig. 10. Structure of wood fibres. Here a - macrostructure of fibres; b - microstructure of fibres

According to the manufacturer, the wood fibres as a bulk material have a moisture content of approximately 5-6% [42]. As seen in Fig. 10, the microscopic structure of the wood fibres demonstrates a complex system of interlocked fibres with sharp needle-shaped features. The characteristic values of different parameters (properties) of the manufactured wood fibres are presented in Table 4.

Table 4. Characteristics of Steico-zell wood fibres [41]

Parameter	Value
Declared thermal conductivity - λ_D	0.038 W/ (m*K)
Specific heat capacity - c	2100 J / (kg*K)
Fire classification according to EN 13501-1	B-s2, d0
Fire class according to EN 13501-1	E
Water vapour diffusion resistance	1-2 μ

2.2.5. Synthetic zeolite

For the synthesis of zeolite, the following raw materials were used:

1. By-product silica gel
2. NaOH - sodium hydroxide
3. $Al(OH)_3$ reagent
4. H_2O

By-product silica gel rich in Si and Al, is an amorphous form of SiO_2 contaminated with fluoride compounds obtained from aluminium fluoride (ALF_3) production waste [44]. From (Fig. 11) X-ray diffraction analysis it can be noted that the waste silica gel contained amorphous $SiO_2 \cdot nH_2O$ and crystalline $AlF_3 \cdot 3.5H_2O$ because SiO_2 is in amorphous phase it is not detected in the XRD pattern.

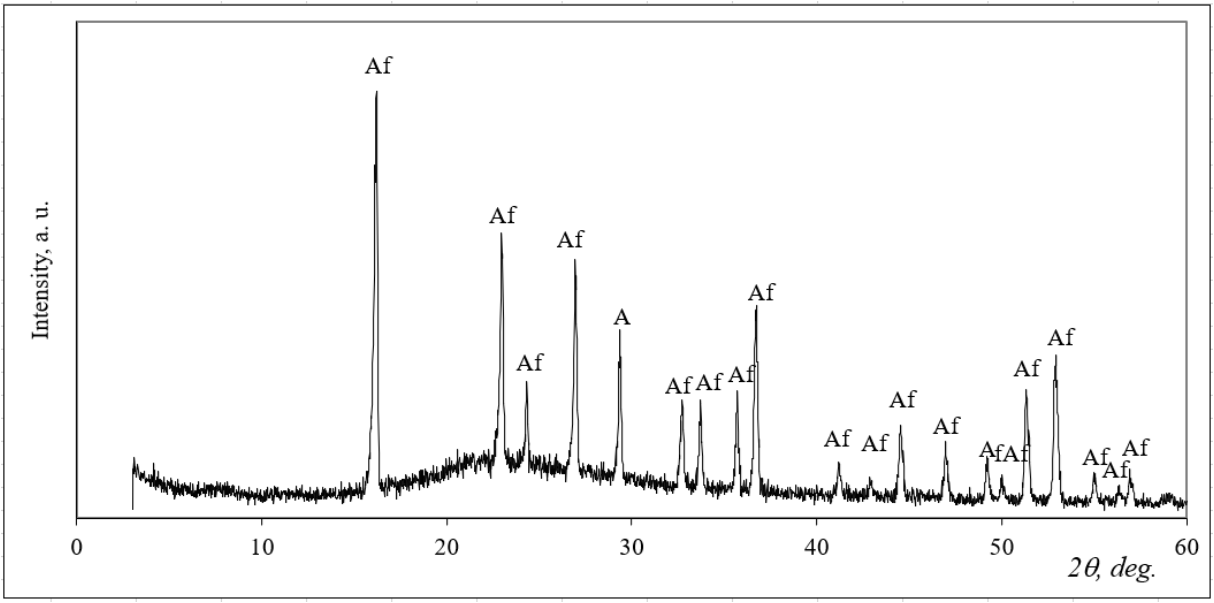


Fig. 11. XRD pattern of waste silica gel. Note: Af - (35-827) hydrated Aluminium fluoride ($AlF_3 \cdot 3.5H_2O$)

The chemical composition of the by-product silica gel was analysed using the XRF technique and the findings are presented (Table 5). The microstructure of the silica gel waste obtained from scanning electron microscope is illustrated in Fig. 13.

Table 5. Chemical composition of by-product silica gel by XRF measurement

Compound	Amount, wt.%
CaO	0.42
SiO_2	72.2
Na_2O	0.02
Al_2O_3	5.68
MnO	0.01
F	21
SO_3	0.01
Fe_2O_3	0.66

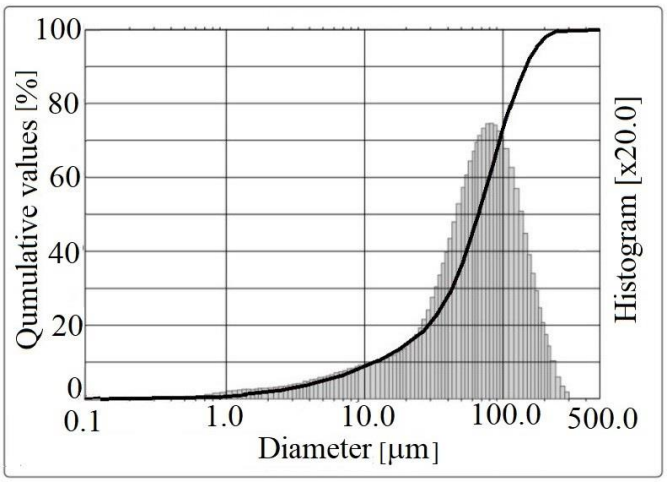


Fig. 12. Silica gel by-product particle size distribution

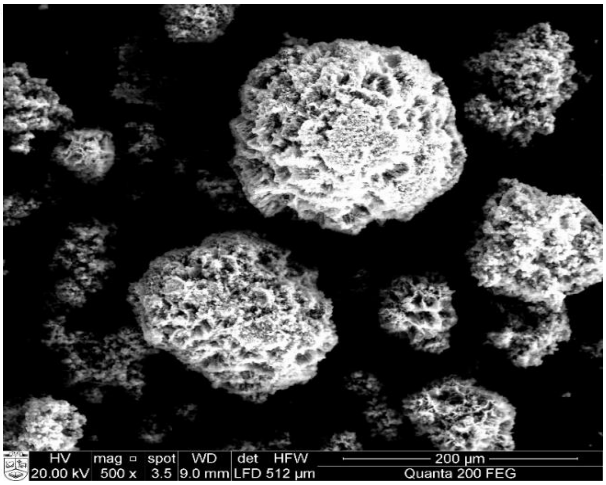


Fig. 13. Scanning electron micrograph of silica gel by-product

From the particle size histogram taken in this study (Fig. 12), the specific surface area of silica gel by-product was equal to 716 m^2/kg recorded using laser particle size analyser "CILAS 1090 LD", it was also found that the particles of this material have diameter ranging from approximately 1.0 µm to 450 µm with a mean diameter of 74.7 µm. The peak of the histogram was noted to be at 95 µm with 3.65% of all particles.

Preparation of synthetic zeolite:

In this study, the synthetic zeolites were prepared from the molar ratio of reagents which were taken as Na_2O: Al_2O_3: SiO_2: H_2O = 2: 1: 2: 10.

- Initially, silica gel by-product and $Al(OH)_3$ were mixed thoroughly and added to NaOH solution to form a homogeneous slurry.
- Low temperature (105°C) synthesis of zeolite was carried out in an enclosed plastic bottle containing the formed slurry with an isothermal holding time of 2 h.
- After that the samples were removed from the oven, once it cooled down to room temperature, the cooled product was washed with deionised water, and vacuum filtered through a Buchner funnel (Fig. 14) to remove impurities such as sodium fluoride (NaF).
- The obtained synthesized product (zeolites) was dried at 100°C for 24 h.

Fig. 14. Vacuum filtration by Bruchner funnel

The synthesized zeolites acquired using waste silica gel as source of silica was analysed by using X-ray diffraction analysis. The XRD pattern resulted in a good crystallinity of hydrosodalite (H) as well as trace amounts of Zeolite A and Zeolite C also synthesized as shown in (Fig. 15).

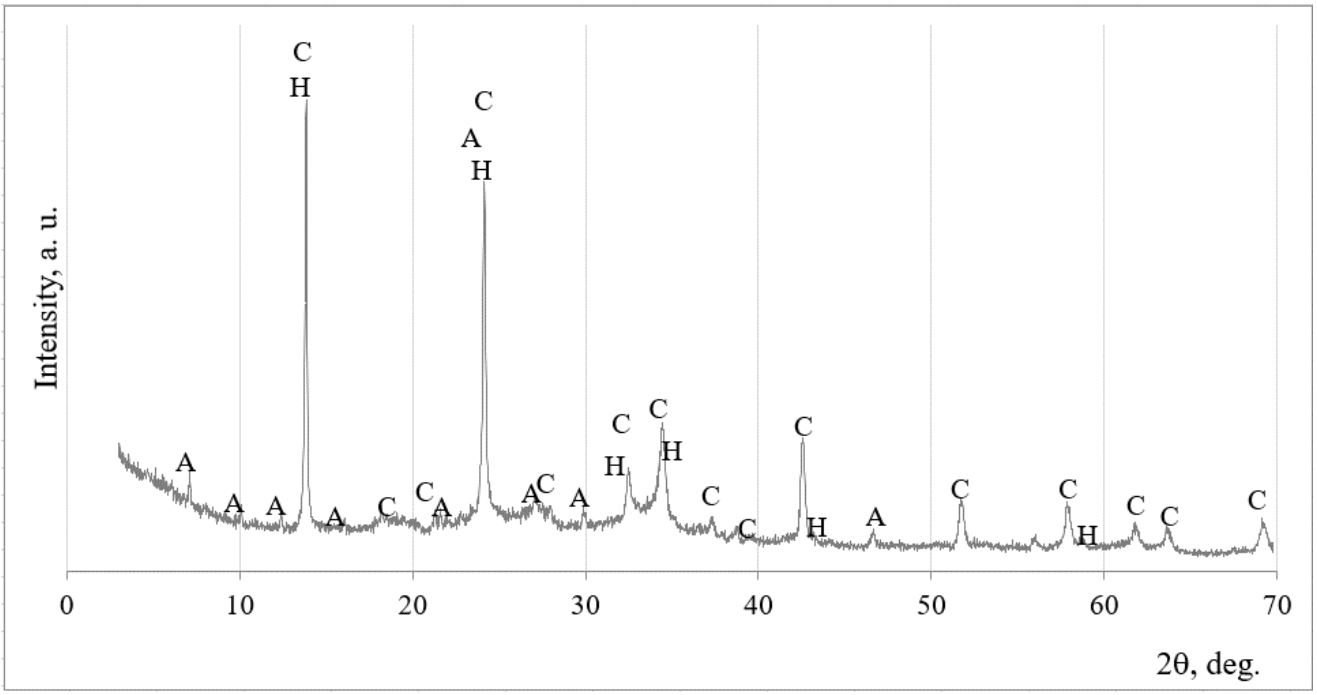


Fig. 15. X-ray diffraction analysis of synthetic zeolite. Notes H - hydrosodalite Na_4.. (76-717); A - Sodium aluminium silicate; C - hydroxycancrinite

3. Methodology of applied research

For this research, there are three types of composite samples made, out of which two types contain zeolitic waste along with untreated and ultrasonically treated wood fibres, respectively. The third sample type contains zeolite synthesized in laboratory and wood fibres ultrasonically treated. This chapter will further explain the methods used for preparing the samples, their compositions and other supplementary procedures involved.

3.1. Preparation of Type-1 wood fibres-cement composites

The first type of composite samples consists of Portland cement, zeolitic waste, untreated wood fibres and water. Here, zeolitic waste was added by the percentage of cement weight: 0%, 1%, 2%, 5% and 10% increments. The water/cement ratio for all samples was maintained as 0.66. The reference sample (Table 6, No. 1) contained 0% of zeolitic waste which will help to analyse its influence in the results.

Table 6. Sample composition of Type-1 composites with zeolitic waste and untreated wood fibres

Sample No.	Portland Cement CEM I 52.5R, g	Wood fibres untreated, g	Zeolitic waste, g / %	Water, ml	W/C ratio
1	500	25	0 (0%)	330	0.66
2	500	25	5 (1%)	330	0.66
3	500	25	10 (2%)	330	0.66
4	500	25	25 (5%)	330	0.66
5	500	25	50 (10%)	330	0.66

Firstly, to prepare the samples all the raw materials were weighed and added to separate containers. Then the dry materials, Portland cement (CEM I 52.5R) and zeolitic waste (FCC) were mixed uniformly, to which the untreated wood fibres were incorporated and then the required amount of water was added and mixed with a shovel until a homogenous cement composite paste was made as shown in Fig. 16.

Fig. 16. Homogeneous wood-fibres cement composite paste

The prepared composite paste was added to the appropriate moulds then compacted, covered with a thin plastic film and stored for about 48 h. After that the samples were de-moulded, added to a container underwater and left for curing. After 28 days the hydration process of samples was concluded, and the properties were tested.

3.2. Preparation of Type-2 wood fibres-cement composites

The second type of composite samples consists of Portland cement, zeolitic waste, ultrasonically treated wood fibres and water. Here, zeolitic waste was added in the same percentages as the first type of composites: 0%, 1%, 2%, 5% and 10%. The water/cement ratio for all samples was also maintained as 0.66. The reference sample (Table 7, No. 1) contained 0% of zeolitic waste and ultrasonically treated wood fibres which will be beneficial to be compared with the reference of the first type composite.

Table 7. Sample composition of Type-2 composites with zeolitic waste and treated wood fibres

Sample No.	Portland Cement CEM I 52.5R, g	Wood fibres treated, g	Zeolitic waste, g / %	Water, ml	W/C ratio
1	500	25	0 (0%)	330	0.66
2	500	25	5 (1%)	330	0.66
3	500	25	10 (2%)	330	0.66
4	500	25	25 (5%)	330	0.66
5	500	25	50 (10%)	330	0.66

The second type of wood fibres-cement composites are prepared following the same steps as the first type of composites, the only difference in the two is that the wood fibres in the second type of composites are added to the composite paste matrix after ultrasonic treatment. The quantities of added raw materials remains unchanged as tabulated above (Table 7), this approach is different from the previous work [45-46 p. 45] in which the quantities of treated and untreated wood fibres and other materials varied, therefore having equal quantities in this study will explain further the dependence of materials and will be useful to draw clear conclusions from results. The properties of the second type of composites are also tested after 28 days.

3.2.1. Ultrasonic treatment of wood fibres

As published in my previous work [45-46 p. 45], wood fibres are treated in the same manner by using ultrasonic dispersion equipment “Bandelin - UW3400”.

Fig. 17. Treated wood fibres after ultrasonic dispersion

The procedure of treating wood fibres follows the immersion of wood fibres into a glass beaker which is mixed with the required amount of water to submerge all the fibres. Then this solution is subjected to ultrasonic dispersion using 20 kHz frequency and 250 W power ultrasonic waves for a duration of

about 1 min. After that, the wood fibres are filtered out from the solution and squeezed dry, these wood fibres shown in Fig. 17 are ready to be added to the cement composite paste.

3.3. Preparation of Type-3 wood fibres-cement composites

The third type of composite samples consists of Portland cement, synthetic zeolite (hydro-sodalite), ultrasonically treated wood fibres and water. Here, zeolite (hydro-sodalite) was added by percentage of cement weight: 0%, 1%, 2%, 5% and 10% increments. The water/cement ratio for all samples was maintained as 0.53. The reference sample (Table 8, No. 1) contained 0% of zeolite synthesized in laboratory (hydro-sodalite) and 5% of ultrasonically treated wood fibres.

Table 8. Sample composition of Type-3 composites with synthetic zeolite and treated wood fibres

Sample No.	Portland Cement CEM I 52.5R, g	Wood fibres treated, g	Zeolite, g / %	Water, ml	W/C ratio
1	500	25	0 (0%)	265	0.53
2	500	25	5 (1%)	265	0.53
3	500	25	10 (2%)	265	0.53
4	500	25	25 (5%)	265	0.53
5	500	25	50 (10%)	265	0.53

For preparing the third type of composite samples the same procedure as the second type of composites was adopted. The only difference between the two types of composites was that synthetic zeolite was incorporated in the third type of composite matrix instead of zeolitic waste. The amounts of raw materials used for preparation remained unchanged. The ultrasonic treatment of wood fibres followed the same technique as described previously. Here, the dry materials, Portland cement (CEM I 52.5R) and zeolite (hydro-sodalite) were mixed uniformly, to which the ultrasonically treated wood fibres were incorporated. After that the required amount of water was added and mixed with a shovel until the consistency of the cement composite paste as shown in Fig. 16 was obtained. The composite mix was added to moulds. Then compacted to remove excess air pockets, covered with a plastic film and stored on an even surface as shown in Fig. 18. After the period of 28 days these samples were tested.

Fig. 18. Storage of samples for initial set

4. Results and discussion

In this chapter, the experimental results obtained from the investigation methods are discussed. The X-ray diffraction pattern (XRD), scanning electron micrograph (SEM), hydration characteristics, other physical and mechanical properties of three different types of hardened wood fibres cement composites are examined.

4.1. Hardened cement composites with zeolitic waste and untreated wood fibres (Type-1 composites)

In the hardened composite samples with untreated wood fibres and zeolitic waste, it is observed that there is an agglomeration of wood fibres in the cement matrix. This aggregation leads to inhomogeneous scattering of wood fibres in the samples as reflected in Fig. 19, creating weak points in the pores.

Fig. 19. Macroscopic structure of samples with untreated wood fibres and zeolitic waste

– **Analysis of compressive strength**

The compressive strength of hardened Type-1 cement composites with 5% untreated wood fibres and varying amounts of zeolitic waste was tested after 28 days of hydration. The compressive strength of the samples was then calculated according to Formula (1). In a study [33] performed on the properties of wood fibres-cement matrix, the highest compressive strength of the samples with untreated wood fibres (i.e. without chemical treatment) was found to be 1.95 MPa, tested after 28 days of curing. Here, the wood/cement ratio was taken as 40:60 with no zeolite or zeolitic waste added to the composite system. The low value of the compressive strength might be related to the presence of high wood/cement ratio and absence of pozzolanic substance. Comparing this research with the above study [33], even the samples with untreated wood fibres and no zeolitic waste resulted in much higher strength with only 5% of untreated wood fibres in the matrix. Blankenhorn et al. [47] published a study on the effects of surface (chemical) treatments on three types of wood fibres with water/cement ratio of 0.37, where the 28^{th} day compressive strength of 4% untreated hardwood fibre composites was found to be 49.3 MPa; 4% untreated kraft softwood fibre composites was found to be 51.7 MPa; and 4% untreated newsprint fibre composites was found to be 43.3 MPa. The authors concluded that when the wood fibres were chemically treated, their compressive strength decreased. Different types of fibres and their respective characteristic features influenced the mechanical properties of these

wood fibres-cement composites. In another study [48], the authors recorded the compressive strength of 4% untreated kraft wood fibres cement composites to be equal to approximately 32.8 MPa (4750 psi) with the same water/cement ratio of 0.37 as the previous study [47]. This value of compressive strength is closer to the values obtained in this study.

The calculated compressive strength of Type-1 wood fibres cement composites is presented in Table 9. According to the obtained results, the maximum compressive strength of 29.6 MPa was noted for the samples with 5% zeolitic waste. Whereas, the reference samples with 0% zeolitic waste had a compressive strength of 25.9 MPa. From which it can be inferred that the samples with 5% zeolitic waste had 3.7 MPa higher strength, that is approximately 14.3% more compressive strength than the reference samples.

Table 9. Results of compressive strength of hardened cement composites with zeolitic waste and untreated wood fibres

Sample No.	Weight, g	Length l, mm	Breadth b, mm	Height h, mm	Force F, kN	Compressive strength, MPa	Average compressive strength, MPa
1.11	321.4	60	40	40	64.3	26.79	25.9
1.12	321.4	60	40	40	59.9	24.96	
1.21	336.9	60	40	40	63.5	26.46	
1.22	336.9	60	40	40	61.1	25.46	
2.11	340.5	60	40	40	59	24.58	26.1
2.12	340.5	60	40	40	68.2	28.42	
2.21	335.9	60	40	40	58.1	24.21	
2.22	335.9	60	40	40	65.1	27.13	
3.11	336.3	60	40	40	67.5	28.13	27.1
3.12	336.3	60	40	40	64	26.67	
3.21	341.5	60	40	40	61.6	25.67	
3.22	341.5	60	40	40	67.2	28.00	
4.11	350.2	60	40	40	71.6	29.83	29.6
4.12	350.2	60	40	40	68.1	28.38	
4.21	347.8	60	40	40	71.5	29.79	
4.22	347.8	60	40	40	73.3	30.54	
5.11	336.2	60	40	40	63.2	26.33	25.9
5.12	336.2	60	40	40	58.4	24.33	
5.21	331.1	60	40	40	66.1	27.54	
5.22	331.1	60	40	40	60.9	25.38	

The average values of the compressive strength of Type-1 composites are plotted in Fig. 20. The development of compressive strength as shown in graph (Fig. 20) indicates an increase in compressive strength with the increasing amount of zeolitic waste from 1% until 5%; further, samples with 10% zeolitic waste reached compressive strength equal to that of reference (0% zeolitic waste). It can be implied that the addition of zeolitic waste on the samples with untreated wood fibres has a positive influence on the compressive strength of these samples.

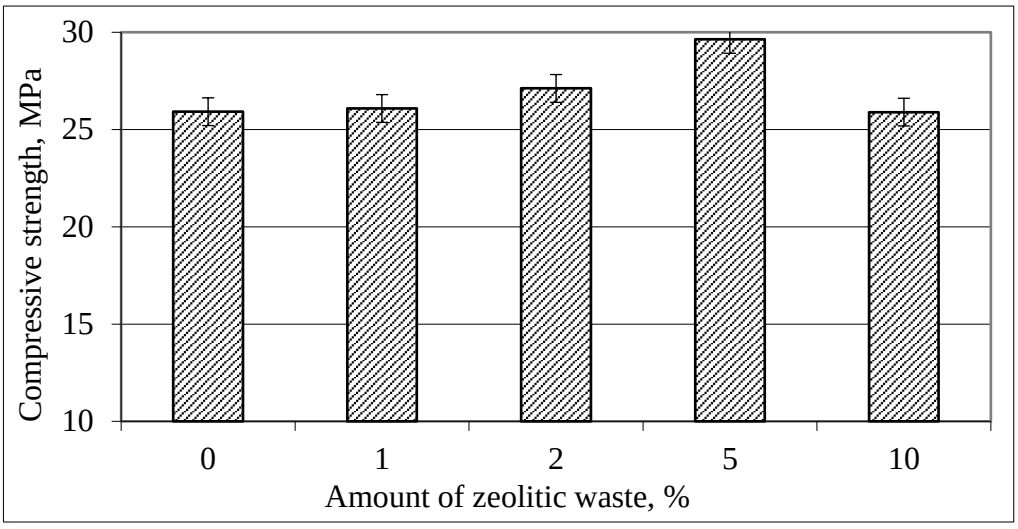


Fig. 20. Compressive strength of hardened cement composites with zeolitic waste and untreated wood fibres

– **Analysis of flexural strength and density**

The flexural strength and density of hardened Type-1 cement composites was calculated according to Formula (2) and Formula (3), respectively. From the calculated results of the flexural strength (Table 10), the reference samples had a strength of 4.8 MPa while the maximum strength was noted to be 5.5 MPa of samples with 5% zeolitic waste. The flexural strength of samples with 5% zeolitic waste indicated an increase of 14.6% from the reference samples. The average values of flexural strength of samples with zeolitic waste was found to be higher than the reference. However, there was no significant relationship between the flexural strength and the amount of zeolitic waste, this could be due to the agglomerated wood fibres pockets present in the samples.

Table 10. Results of flexural strength and density of hardened cement composites with untreated wood fibres and zeolitic waste

Sample No.	Weight, g	Length l, mm	Breadth b, mm	Height h, mm	Force F, kN	Flexural strength, MPa	Average Flexural strength, MPa	Average Density, kg/m^3
1.1	321.4	159	40	40	2.069	4.85	4.8	1290
1.2	336.9	160	40	40	2.040	4.78		
2.1	340.5	160	40	40	2.145	5.03	5.4	1325
2.2	335.9	159	40	40	2.480	5.81		
3.1	336.3	160	40	40	2.216	5.19	5.0	1324
3.2	341.5	160	40	40	2.075	4.86		
4.1	350.2	160	40	40	2.458	5.76	5.5	1359
4.2	347.8	161	40	40	2.274	5.33		
5.1	336.2	160	40	40	2.140	5.02	5.2	1303
5.2	331.1	160	40	40	2.309	5.41		

Through experimental research, Blankenhorn et al. [47] observed the 28th day average bending strength of 4% untreated hardwood fibre composites to be equal to 11.3 MPa; 4% untreated kraft softwood fibre composites to be equal to 11.5 MPa; and 4% untreated newsprint fibre composites to

be equal to 9.6 MPa. The authors noted that the untreated wood fibres–cement composites average flexural strength values were higher than that of the control specimens. In this paper, the samples did not contain any zeolites, but superplasticizer was added to reduce the water content for sample compositions. Pehanich et al. [48] studied the flexural strength properties of two types of fibre composites, the flexural strength of untreated kraft wood fibres composite was found to be 8.6 MPa (1250 psi) whereas, the flexural strength of untreated newspaper fibres composite was found to be 10.3 MPa (1490 psi). The study concluded that the chemical treatment of wood fibre cement composites enhanced their flexural strength.

According to the plotted graph (Fig. 21), the development of flexural strength in the samples with untreated wood fibres found to have an uneven distribution. The reference samples had a strength of 4.8 MPa, then it increased till 5.4 MPa after that the values went down, then went up again to reach maximum flexural strength of 5.5 MPa and finally went down to 5.2 MPa. This trend of development could be related to the inhomogeneous scattering of the wood fibres in the cement matrix.

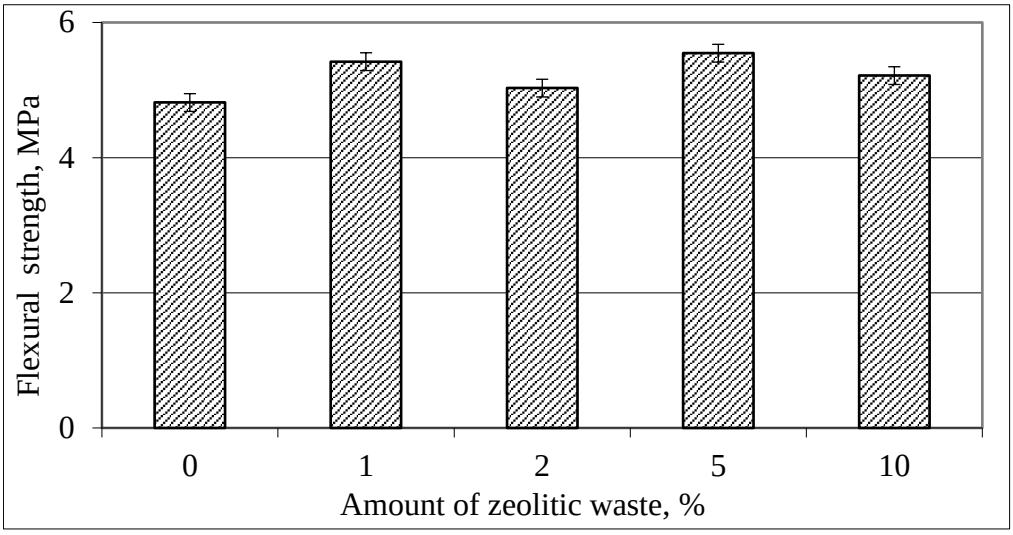


Fig. 21. Flexural strength of hardened cement composites with zeolitic waste and untreated wood fibres

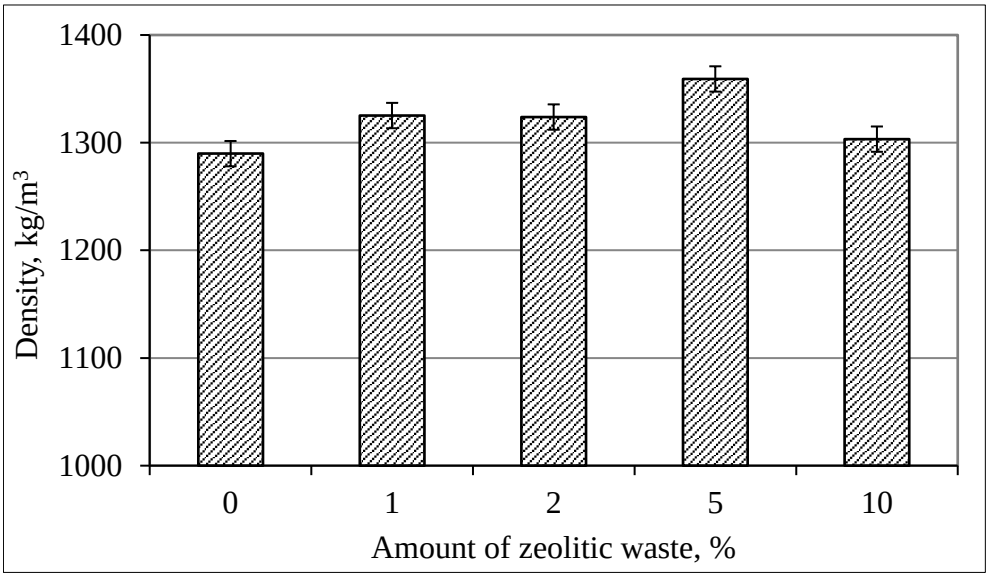


Fig. 22. Density of hardened cement composites with zeolitic waste and untreated wood fibres

The density of the samples with untreated wood fibres as plotted in Fig. 22 showed that all the samples had low densities around 1300 kg/m^3. The density of the reference was found to be 1290 kg/m^3, which was the lowest among the other samples. The highest density of 1359 kg/m^3 was noted for the sample with 5% zeolitic waste. By increasing the amount of zeolitic waste from 1% to 5% the density slightly increased too, from 1325 kg/m^3 to 1359 kg/m^3, respectively. When the amount of zeolitic waste reached 10% the density went down till 1303 kg/m^3. This indicated that when the highest amount of zeolitic waste was added to the composites, it reduced the density as well as the mechanical strength of the composites. From the obtained results, it can be observed that the highest compressive strength, flexural strength and density of Type-1 composites was achieved by the samples with 5% zeolitic waste. However, due to the agglomeration of untreated wood fibres in the hardened cement matrix. Such composite system cannot be suitable for future application. To negate this influence, wood fibres were treated ultrasonically in Type-2 and Type-3 composites. Therefore, further tests were not conducted on these samples.

4.2. Hardened cement composites with zeolitic waste and ultrasonically treated wood fibres (Type-2 composites)

The ultrasonic treatment of wood fibres in water was used for even distribution of wood fibres in the matrix of hardened cement paste. In the composite specimens with ultrasonically treated wood fibres and zeolitic waste, macroscopic observation showed a homogenous distribution of the wood fibres in the cement matrix. This uniformity is contributed to the ultrasonic dispersion of wood fibres prior to the addition to the cement composite system. When compared with the samples that contain untreated wood fibres, the samples with treated wood fibres do not exhibit segregation of wood fibres in the hardened composites, as seen in Fig. 23.

Fig. 23. Macroscopic structure of samples with treated wood fibres and zeolitic waste

– **Analysis of compressive strength**

Currently, there is a lack of scientific research where ultrasonically treated wood fibres and zeolitic waste/zeolite can be used to make wood fibres cement composites. This research will be the first in this field, therefore the investigated properties of this study cannot be fully compared with that of the existing scientific studies.

After 28 days of hydration, the compressive strength of the hardened wood fibres-cement composites with 5% of ultrasonically treated wood fibres and different measures of zeolitic waste was tested. The compressive strength of the samples was also calculated according to Formula (1). From the

calculated results (Table 11), the highest compressive strength of 19.2 MPa was noted for the samples with 10% zeolitic waste whereas, the reference samples with 0% zeolitic waste had a compressive strength of 13.3 MPa. The specimens with maximum compressive strength had approximately 44% more compressive strength than the reference samples.

Table 11. Results of compressive strength of hardened cement composites with zeolitic waste and treated wood fibres

Sample No.	Weight, g	Length l, mm	Breadth b, mm	Height h, mm	Force F, kN	Compressive strength, MPa	Average compressive strength, MPa
1.11	237	60	40	40.1	31.31	13.05	13.3
1.12	237	60	40	40.1	31.71	13.21	
1.21	250.8	60	40	41.5	35.11	14.63	
1.22	250.8	60	40	41.5	29.18	12.16	
2.11	233.8	60	40	41	30.66	12.78	12.5
2.12	233.8	60	40	41	31.9	13.29	
2.21	229.2	60	40	40.2	29.15	12.15	
2.22	229.2	60	40	40.2	27.9	11.63	
3.11	257.1	60	40	39.6	36	15.00	15.1
3.12	257.1	60	40	39.6	34.46	14.36	
3.21	255	60	40	39.5	36.81	15.34	
3.22	255	60	40	39.5	37.93	15.80	
4.11	237.5	60	40	39.7	32.08	13.37	13.4
4.12	237.5	60	40	39.7	31.44	13.10	
4.21	243.1	60	40	39.6	33.27	13.86	
4.22	243.1	60	40	39.6	31.77	13.24	
5.11	287.8	60	40	39.2	46.99	19.58	19.2
5.12	287.8	60	40	39.2	46.47	19.36	
5.21	293	60	40	40	44.29	18.45	
5.22	293	60	40	40	46.75	19.48	

The average values of the compressive strength of Type-2 composites are plotted in Fig. 24. From the graph of compressive strength, it was noted that the samples with 2% and 10% zeolitic waste presented better compressive strength of 15.1 MPa and 19.2 MPa, respectively than compared with the other samples. The development of compressive strength of all the samples revealed that as the amount of zeolitic waste increased in Type-2 composites, the compressive strength also increased slightly. It was also observed that there is a rapid growth in compressive strength of the samples containing 10% of zeolitic waste. This improvement in strength can be explained due to the generation of additional hydration products (like calcium silicate hydrates and ettringite) in the cementitious composite matrix. When comparing the compressive strength results of Type-1 and Type-2 composites, Type-1 composites showed higher compressive strength. However, due to their uneven scattering of wood fibres, Type-2 composites with treated wood fibres will be preferred over them.

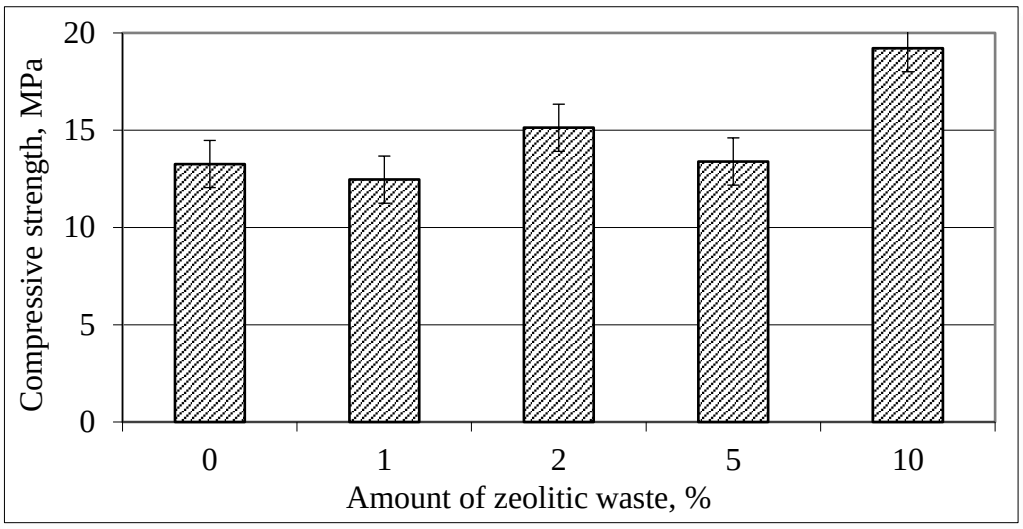


Fig. 24. Compressive strength of hardened cement composites with zeolitic waste and treated wood fibres

– **Analysis of flexural strength and density**

The investigation of flexural strength and density for the samples with zeolitic waste and ultrasonically treated wood fibres are represented in Fig. 25 and Fig. 26, respectively. From the experimental results of the flexural strength (Table 12), the reference samples had a strength of 4.6 MPa while the maximum strength was noted to be 5.5 MPa of samples with 10% zeolitic waste, which indicated an increase of 19.6% from the reference samples.

Table 12. Results of flexural strength and density of hardened cement composites with treated wood fibres and zeolitic waste

Sample No.	Weight, g	Length l, mm	Breadth b, mm	Height h, mm	Force F, kN	Flexural strength, MPa	Average Flexural strength, MPa	Average Density, kg/m^3
1.1	237	158	36.2	40.1	1.85	4.77	4.6	1045
1.2	250.8	159	36	41.5	1.85	4.48		
2.1	233.8	159	35.2	41	1.72	4.36	4.7	1022
2.2	229.2	159	35	40.2	1.88	4.99		
3.1	257.1	159	39.2	39.6	1.59	3.88	4.5	1065
3.2	255	159	37.34	39.5	2.02	5.20		
4.1	237.5	158	36	39.7	1.62	4.28	4.4	1059
4.2	243.1	159	36.2	39.6	1.69	4.47		
5.1	287.8	159	39	39.2	2.16	5.41	5.5	1183
5.2	293	158	39.2	40	2.34	5.60		

From the graph (Fig. 25) of flexural strength, it can be concluded that the flexural strength of Type-2 composite samples decreased with the addition of zeolitic waste from 1% until 5%, then when 10% zeolitic waste was added there was a significant increase in the flexural strength similar to the growth of compressive strength. The values of flexural strength of Type-1 and Type-2 composites are very similar.

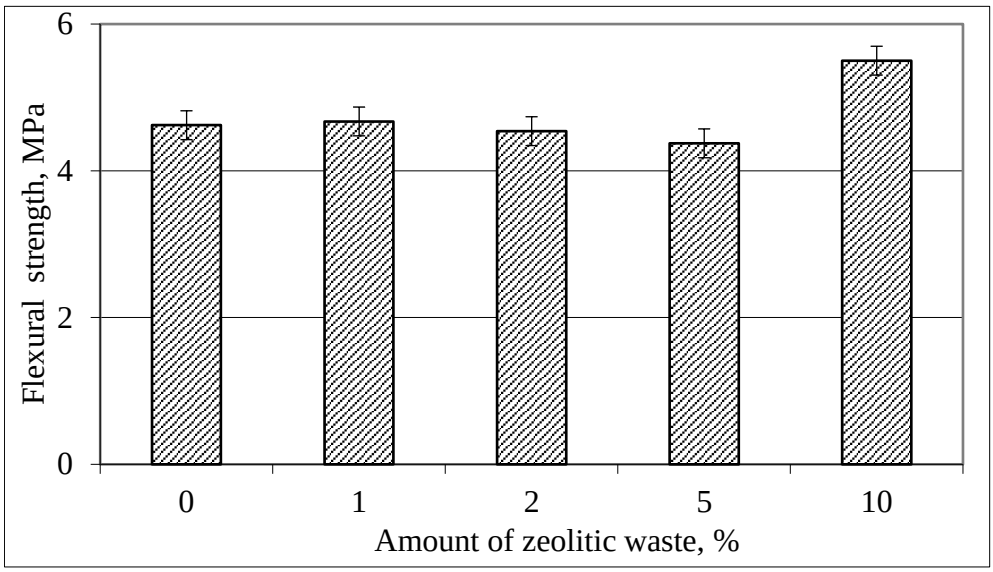


Fig. 25. Flexural strength of hardened cement composites with zeolitic waste and treated wood fibres

From the calculated density results, all the samples with treated wood fibres and zeolitic waste had relatively low densities ranging from approximately 1000 to 1200 kg/m^3. The reference sample had a density of 1045 kg/m^3 and the highest density of 1183 kg/m^3 was noted for the samples with 10% zeolitic waste which was approximately 13% more than the reference samples. From Fig. 26 it can be observed that by increasing the amount of zeolitic waste the density of Type-2 composites slightly increased. The investigated density results pointed out that the densities of the samples with ultrasonically treated wood fibres was lower than the samples with untreated wood fibres. Low density of the samples can be mainly contributed to the usage of wood fibres in the cement composites and shows future applications of this research to make low density concrete.

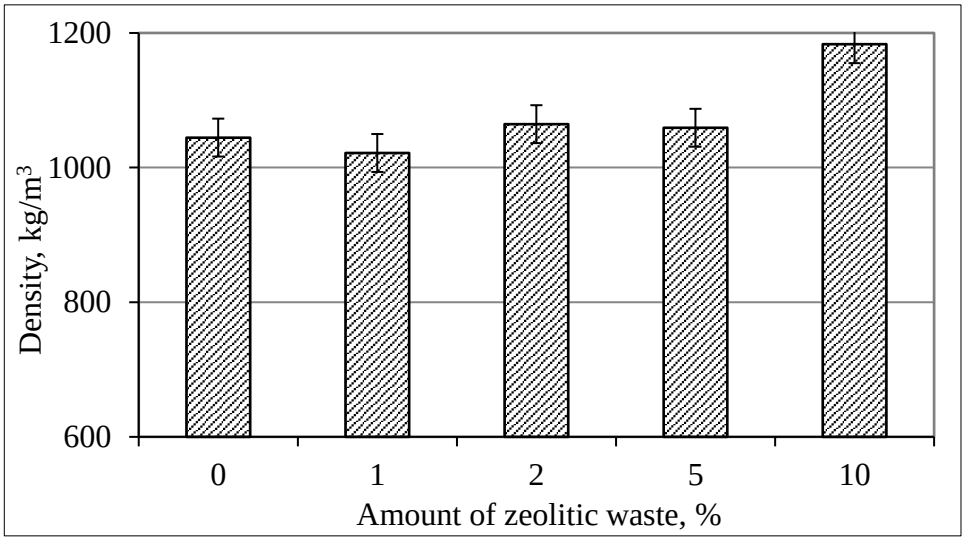


Fig. 26. Density of hardened cement composites with zeolitic waste and treated wood fibres

Since, the compressive strength, flexural strength and density of the samples with 10% zeolitic waste was the highest among all the other Type- 2 composites, it can be assumed that the amount of 10% zeolitic waste had a positive result on the hydration of wood-fibres cement composite.

– **Analysis of the mineral composition (XRD) of composites reinforced with wood fibres (without zeolitic by-product and with zeolitic by-product)**

The mineral composition was evaluated by using X-Ray diffraction patterns (Fig. 27). Ettringite, portlandite, calcium silicate hydrate (C-S-H) and calcite were identified as the main hydration products of the wood fibres-cement composites. Both the curves with 0% zeolitic waste and 10% zeolitic waste had the same peaks, but the intensity of these peaks slightly differed. The incorporation of zeolitic by-product had a positive influence on the significant increase of the peaks of ettringite. This zeolitic by-product also led to the formation of additional hydration product - calcium aluminate hydrates, calcium aluminosilicate hydrates and calcium sulfoaluminate hydrates. The generation of the additional hydration products could be related with the higher compressive strength of samples with zeolitic waste compared with the reference samples (Fig. 24).

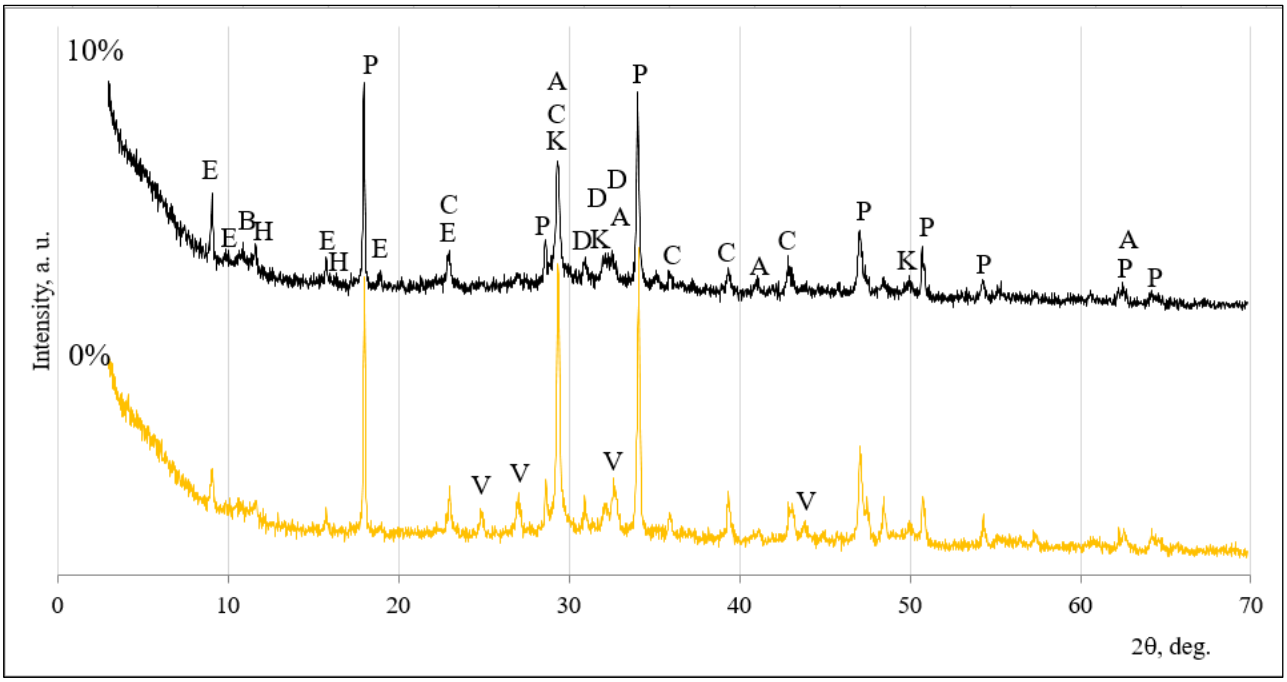


Fig. 27. X-Ray diffraction patterns of composites reinforced with wood fibres without zeolitic by-product and with zeolitic by-product, after 28 days. Notes: E – ettringite $Ca_6Al_2(SO_4)_3(OH)_{12}\cdot 26H_2O$ (41–1451), P – portlandite $Ca(OH)_2$ (44-1481); A – $Ca_{54}MgAl_2Si_{16}O_{90}$ (13-272); D – larnite $Ca_2(SiO_4)$ (83-461); K – calcium silicate hydrate $Ca_{1.5}SiO_{3.5}\cdot x\ H_2O$ (33-306); C – calcite $CaCO_3$ (72-1651); B – brownmillerite $Ca_2(Al,Fe)_2O_5$ (30-226); H – hydrotalcite $((Mg_4Al_2)(OH)_{12}CO_3(H_2O)_3)_{0.5}$ (70-2151); V – vaterite $CaCO_3$ (25-127)

– **Analysis of the microstructure (SEM) of composites reinforced with wood fibres (without zeolitic by-product and with zeolitic by-product)**

The microstructure investigation of composites provides information about the influence of zeolitic by-product on the particles shape and the contact zones of the new formed crystalline phases (Fig. 28). Composites reinforced with 5.0% of treated wood fibres without zeolitic by-product are shown in the Figure 28, a and b. The observable contact zone reveals less adhesion between wood fibres and hardened cement paste. This weak contact could be related with the lower compressive and flexural strength values of the samples without zeolitic waste (Fig. 24 and 25).

Figure 28, c and d represent the microstructure of samples with the incorporation of 5.0% treated wood fibres and 10% zeolitic by-product. In this case, it is possible to detect better adhesion between wood fibres and cementitious products in the matrix of hardened cement paste. In the Fig. 28, c the particles of C-S-H and ettringite cover the wood fibres completely, which provides better contact zone. These two minerals have a positive effect on the mechanical properties of the composites. This type of compact contact zone increased the values of compressive strength, flexural strength as well as the density of the samples. The $CaO–Al_2O_3–CaSO_4–H_2O$ system primarily forms ettringite at the early stages of Portland cement hydration, which subsequently transforms into monosulfate (group of calcium aluminate hydrates - AFm). Zeolitic waste stabilized ettringite in the composite system. However, this study does not include long terms effect of such ettringite phases. The particles of C-S-H has fibres, threads or needles like shape. While the ettringite phases crystallised as acicular-shaped crystals [46].

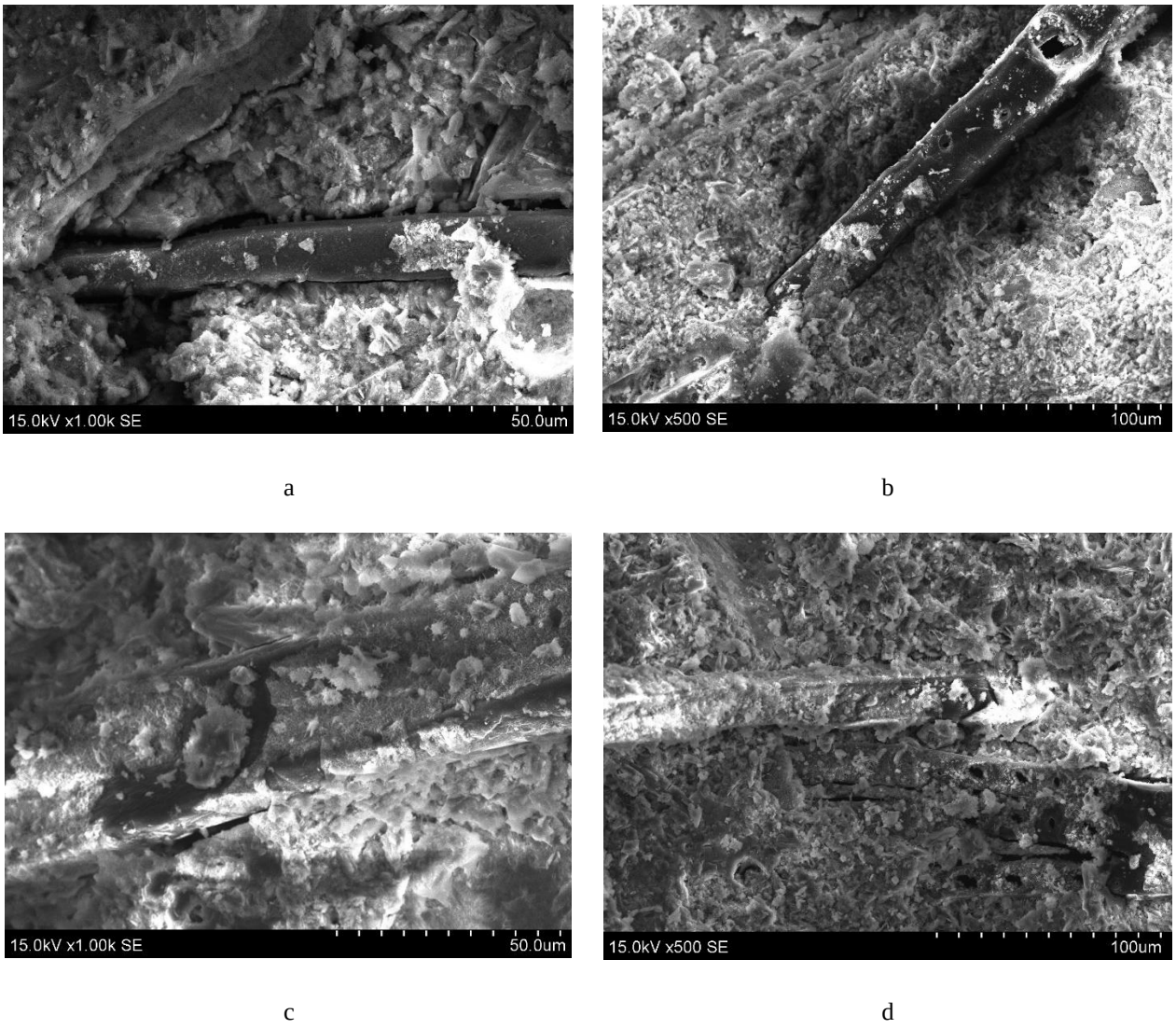


Fig. 28. Microstructure of composites reinforced with wood fibres without zeolitic by-product (a and b) and with zeolitic by-product (c and d), after 28 days.

– **Analysis of semi-adiabatic calorimetry**

From the results of hydration, it can be deduced that all compositions reached hydration at similar intervals when compared with the reference. The maximum temperature of hydration was noted at

37.4°C which was reached by the sample with 1% zeolitic waste, that was 1.5°C higher than the maximum temperature of hydration of the control sample with 0% zeolitic waste. Maximum delay in hydration was recorded for the composition with 5% zeolitic waste with a delay in peak time of 83 min from the reference; reached peak temperature of about 34°C that was 1.9°C lower than the control composition.

Table 13. Table of results of hydration of cement composite paste with treated wood fibres and zeolitic waste

Sample No.	Sample Composition	Time of initial set, min	Temperature at initial set, °C	Time of final set, min	Temperature at final set, °C
1	CEM 1 52.5R + 0% Zeolitic waste	192	27.3	700	35.9
2	CEM 1 52.5R + 1% Zeolitic waste	181	28.2	692	37.4
3	CEM 1 52.5R + 2% Zeolitic waste	202	27.9	686	36.4
4	CEM 1 52.5R + 5% Zeolitic waste	201	27.6	783	34.0
5	CEM 1 52.5R + 10% Zeolitic waste	199	29.1	709	33.0

Analogous tendencies can be observed from the plotted graph of hydration curves (Fig. 29). When the composite system was incorporated with 5% and 10% zeolitic waste, the hydration temperature was lower than in the sample without zeolitic waste, this can be closely related with the reaction of ordinary Portland cement in the system. When lower amount of Portland cement reacts in the composite the lower hydration temperature was reached, due to the dilution effect (Fig. 29).

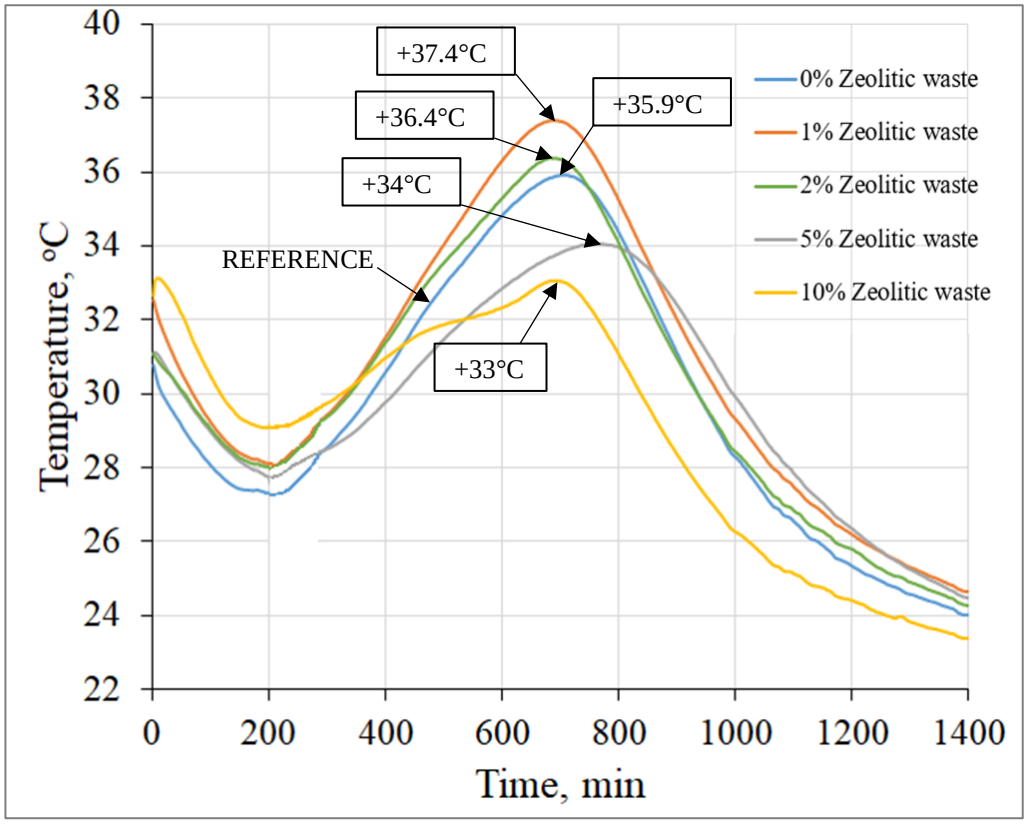


Fig. 29. Hydration temperature graph of composites with treated wood fibres and zeolitic waste

– **pH analysis of composites with treated wood fibres and zeolitic waste**

The pH values (Table 14) of hardened composites with treated wood fibres and zeolitic waste shows identical values, but all values are less than 12.5. Cement systems are alkaline in nature and their values of pH range are usually above 12.5. The sample with 0% zeolitic waste has the highest pH value compared to the other samples. There is a gradual decrease in the pH of samples with an increase in the content of zeolitic waste. This reduction of pH decelerates the hydration process and prolongs the setting time of composites. During the experiment, the pH of the treated wood fibres was measured and found to be 6.8 making it a slightly acidic compound in the composite system.

Table 14. pH values of hardened cement composites with zeolitic waste and treated wood fibres

Sample No.	Percentage of zeolitic waste	pH
1	0%	12.18
2	1%	12.10
3	2%	12.04
4	5%	12.00
5	10%	11.99

4.3. Hardened cement composites with synthetic zeolite and ultrasonically treated wood fibres (Type-3 composites)

In the hardened composites with ultrasonically treated wood fibres and synthetic zeolite, macroscopic observation shows homogenous distribution of wood fibres in the cement matrix, which resembles the distribution of wood fibres in composites with treated wood fibres and zeolitic waste. This homogeneity is maintained due to the ultrasonic dispersion of wood fibres prior to the addition to the cement system. When compared with the samples that contain untreated wood fibres, the samples with treated wood fibres do not exhibit agglomeration of wood fibres in hardened composites, as seen in Fig. 30.

Fig. 30. Macroscopic structure of samples with treated wood fibres and zeolite (hydrosodalite)

– **Analysis of compressive strength**

The compressive strength of samples with 5% ultrasonically treated wood fibres and synthetic zeolite (hydrosodalite) was tested after 28 days of curing. The compressive strength of the samples was then calculated according to Formula (1). From the obtained results (Table 15), the maximum compressive

strength of 15.6 MPa was noted for the samples with 1% synthetic zeolite. The reference samples with 0% synthetic zeolite had a compressive strength of 13.3 MPa, from which it can be deduced that the reference sample had approximately 17% less compressive strength than the samples with 1% synthetic zeolite.

Table 15. Results of compressive strength of hardened cement composites with synthetic zeolite and treated wood fibres

Sample No.	Weight, g	Length l, mm	Breadth b, mm	Height h, mm	Force F, kN	Compressive strength, MPa	Average compressive strength, MPa
1.11	237	60	40	40.1	31.31	13.05	13.3
1.12	237	60	40	40.1	31.71	13.21	
1.21	250.8	60	40	41.5	35.11	14.63	
1.22	250.8	60	40	41.5	29.18	12.16	
2.11	291.8	60	40	40	38	15.83	15.6
2.12	291.8	60	40	40	38.9	16.21	
2.21	291.5	60	40	38.9	37	15.42	
2.22	291.5	60	40	38.9	36.1	15.04	
3.11	301	60	40	40	33.7	14.04	14.7
3.12	301	60	40	40	35.6	14.83	
3.21	295.4	60	40	39.4	35.6	14.83	
3.22	295.4	60	40	39.4	36.2	15.08	
4.11	296.1	60	40	40.4	34.3	14.29	14.6
4.12	296.1	60	40	40.4	35.7	14.88	
4.21	299.8	60	40	39.5	35.4	14.75	
4.22	299.8	60	40	39.5	35.1	14.63	
5.11	288.9	60	40	39.8	31.6	13.17	13.3
5.12	288.9	60	40	39.8	34.1	14.21	
5.21	287.1	60	40	39.6	31.2	13.00	
5.22	287.1	60	40	39.6	30.8	12.83	

The average values of the compressive strength of Type-3 composites are plotted in Fig. 31. The development of compressive strength followed that with the increase of the amount of synthetic zeolite in the composites the compressive strength of the samples significantly decreased. All samples with synthetic zeolite had compressive strength not less than that of the reference samples. The highest compressive strength was noted for the samples with 1% synthetic zeolite. From the examination of compressive strength results, it can be concluded that higher compressive strength can be reached with the incorporation of lower amount of synthetic zeolite in wood fibres cement composites.

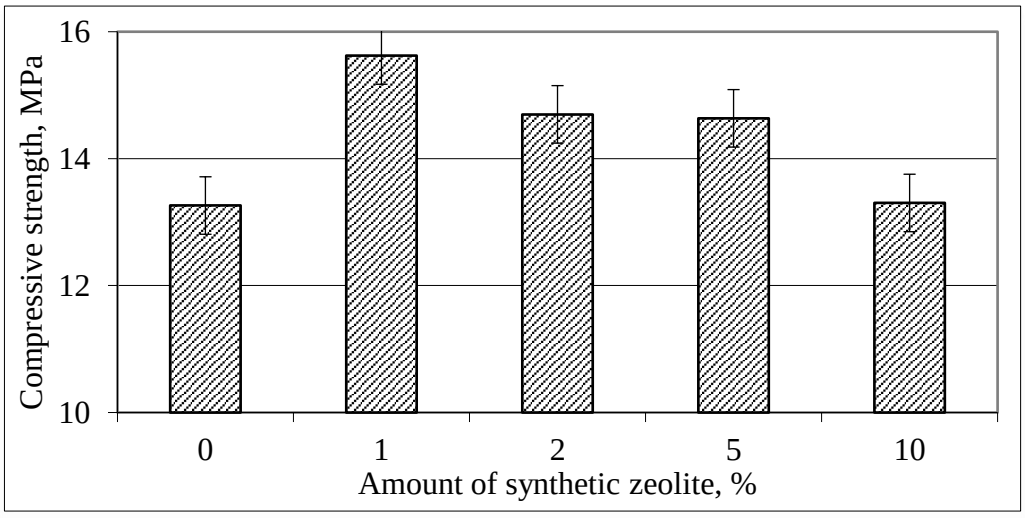


Fig. 31. Compressive strength of hardened cement composites with synthetic zeolite and treated wood fibres

– **Analysis of flexural strength and density**

The flexural strength and density of samples with ultrasonically treated wood fibres and zeolite (hydrosodalite) was tested and graphically represented in Fig. 32 and Fig. 33, respectively. The flexural strength of samples was calculated and presented in Table 16, that indicated that all the samples have similar values of flexural strength. Among all the samples, the highest flexural strength of 4.8 MPa was noted for the sample with 1% synthetic zeolite. The reference sample had a strength of 4.6 MPa which was approximately 4.4% less than the samples with the highest flexural strength.

Table 16. Results of flexural strength and density of hardened cement composites with treated wood fibres and synthetic zeolite

Sample No.	Weight, g	Length l, mm	Breadth b, mm	Height h, mm	Force F, kN	Flexural strength, MPa	Average Flexural strength, MPa	Average Density, kg/m^3
1.1	237	158	36.2	40.1	1.85	4.77	4.6	1045
1.2	250.8	159	36	41.5	1.85	4.48		
2.1	291.8	160	38	40	1.90	4.70	4.8	1218
2.2	291.5	160	37.9	38.9	1.89	4.94		
3.1	301	160.4	38.2	40	2.05	5.03	4.4	1213
3.2	295.4	160.4	39	39.4	1.49	3.69		
4.1	296.1	160.5	39	40.4	1.51	3.57	3.7	1184
4.2	299.8	160.5	39.5	39.5	1.59	3.86		
5.1	288.9	160.2	38.1	39.8	1.67	4.15	3.9	1179
5.2	287.1	160	38.8	39.6	1.49	3.68		

From the plotted graph of flexural strength, it was observed that the flexural strength of Type-3 composites decreased as the amount of synthetic zeolite increased in the composites. The results of flexural strength are similar to the results of compressive strength of Type-3 composites.

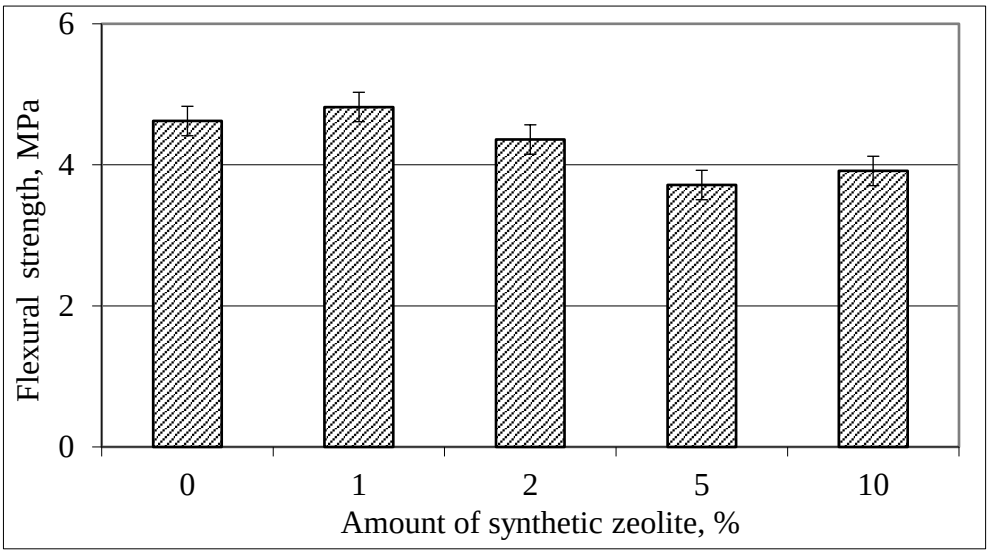


Fig. 32. Flexural strength of hardened cement composites with synthetic zeolite and treated wood fibres

From the calculated density results, all the samples with treated wood fibres and synthetic zeolite had relatively low densities ranging from approximately 1000 to 1220 kg/m^3, the reference sample had a density of 1045 kg/m^3 and the highest density of 1218 kg/m^3 was noted for the sample with 1% zeolite which was approximately 16% more than the reference sample. The samples containing synthetic zeolite have similar values of density, among them the lowest density was noted for the sample with 10% synthetic zeolite. The density of both Type-2 and Type-3 composites was lower than the density of samples with untreated wood fibres. From the obtained results, it can be observed that the samples with 1% synthetic zeolite had the highest compressive strength, flexural strength and density compared to all other Type-3 composite samples.

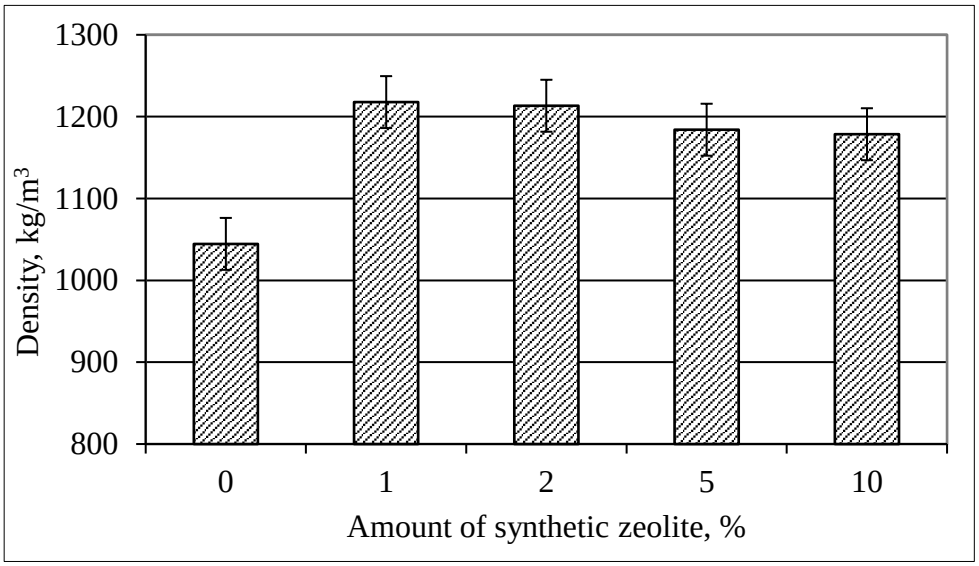


Fig. 33. Density of hardened cement composites with synthetic zeolite and treated wood fibres

– **Analysis of semi-adiabatic calorimetry**

From the results of hydration, it can be deduced that all compositions that contained synthetic zeolite (hydrosodalite) had a delayed hydration when compared with the reference sample. The maximum

temperature was noted at 27.6°C for the samples with 2% synthetic zeolite. Maximum delay in hydration was recorded for the composition with 10% synthetic zeolite with a delay in peak time of 326 min; it reached temperature of about 26.67°C which was approximately 0.1°C lower than the peak temperature of the reference samples.

Table 17. Table of results of hydration of cement composite paste with treated wood fibres and synthetic zeolite

Sample No	Sample composition	Time of initial set, min	Temperature at initial set, °C	Time of final set, min	Temperature at final set, °C
1	CEM 1 52.5R + 0% Zeolite	105	23.8	475	26.78
2	CEM 1 52.5R + 1% Zeolite	175	23.03	648	27.58
3	CEM 1 52.5R + 2% Zeolite	299	22.59	744	27.6
4	CEM 1 52.5R + 5% Zeolite	315	22.01	745	27.24
5	CEM 1 52.5R + 10% Zeolite	337	21.65	801	26.67

From Fig. 34, it is evident that all composites containing synthetic zeolite (hydrosodalite) have prolonged hydration time but the peaks of final set reached higher temperatures. The delayed hydration of Type-3 composites can be related to the degradation of cellulose and lignin in the presence of hydrosodalite. The samples that reached higher temperatures also had increased mechanical strength properties in Type-3 composites.

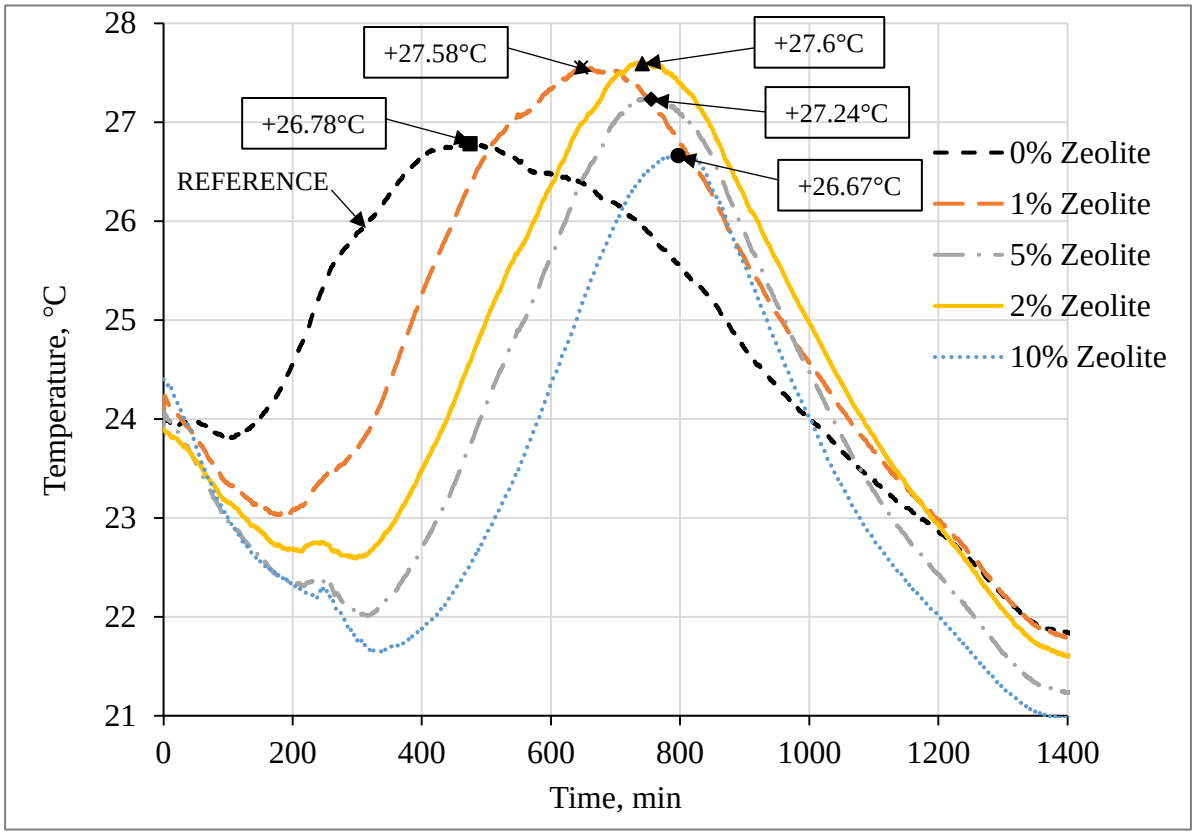


Fig. 34. Hydration temperature graph of treated wood fibres and synthetic zeolite (hydro-sodalite)

– **pH analysis of composites with treated wood fibres and synthetic zeolite**

The pH values (Table 18) of the hardened wood fibres-cement composites with treated wood fibres and synthetic zeolite displayed identical values, but all values are less than 12.5. Cement systems are usually alkaline in nature and the values of pH range is above 12.5. The sample with 10% zeolitic waste has the highest pH value compared to the other samples. The value of pH slightly increases with an increase in the amount of synthetic zeolite present in the samples, from 1% until 10%. During the experiment, the pH of treated wood fibres present in samples was also measured separately and found to be 6.8.

Table 18. pH values of hardened cement composites with zeolite and treated wood fibres

Sample No.	Percentage of zeolitic waste	pH
1	0%	12.18
2	1%	12.27
3	2%	12.28
4	5%	12.29
5	10%	12.30

– **Water absorption analysis of Type-2 and Type-3 composites**

The water-absorption capacity of the wood fibres cement composites with treated wood fibres and pozzolanic agents (zeolitic waste and synthetic zeolite) was measured by soaking oven-dried hardened composites in a static water bath for a period of 48 hr. To determine the water absorption of samples, the difference in the mass of samples before water bath and after water bath was calculated and plotted in a graph.

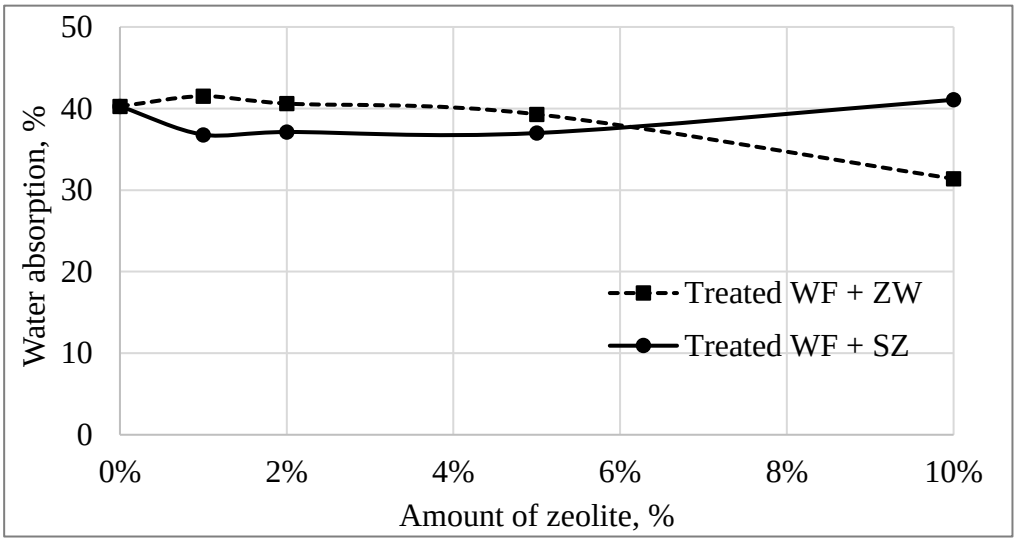


Fig. 35. Graph of water absorption of Type-2 and Type-3 composites. Here WF - wood fibres, ZW - zeolitic waste and SZ - synthetic zeolite (hydrosodalite)

From the graph (Fig. 35), It can be concluded that, when the composites were incorporated with synthetic zeolite from 1% until 5% there was a decrease in the amount of water absorption but when the amount reached 10% the water absorption significantly increased. This can be related with the

higher density and the mechanical properties of Type-3 composites with lower water absorption values. Whereas, when the composites were incorporated with zeolitic waste from 1% until 2% there was an increase in the amount of water absorption in samples, but when the amount reached 5% until 10% the water absorption significantly decreased. Correspondingly, this effect can be related with the higher density and the mechanical properties of Type-2 composites with lower water absorption values. The highest water absorption of 41% was noted for samples with 10% synthetic zeolite. While, the lowest water absorption of 31% was noted for samples with 10% zeolitic waste. Mahzabin et al. [33] in their research with wood fibres-cement composites recorded similar values of water absorption for chemically treated wood fibres without pozzolanic materials, the water absorption values were in the range of 25-35% approximately.

Conclusions

1. Type-1 composites with untreated wood fibres and zeolitic waste indicated agglomeration of wood fibres in the structure of the samples. The compressive strength, flexural strength and density of the composites varied with the addition of different percentages of zeolitic waste, which could be related to the inhomogeneous scattering of wood fibres in the hardened composites. It has been found that composites with ultrasonically treated wood fibres did not exhibit agglomeration and maintained homogenous distribution of wood fibres in the matrix of hardened cement paste.
2. When 10% of zeolitic waste was added to the cement composite with treated wood fibres, the maximum compressive strength of 19.2 MPa was reached which was 44% more than the reference; the maximum flexural strength of 5.5 MPa was reached which was found to be 19.6% more than the reference; and the density of 1183 kg/m^3 was reached among Type-2 composites.
3. The mineral composition evaluated by XRD of Type-2 composites with 0% and 10% zeolitic waste revealed that the addition of zeolitic waste into the wood fibre cement system increased the formation of hydration products primarily, calcium silicate hydrate (C-S-H) and ettringite, this is directly correlated with the higher compressive strength obtained for these composites.
4. The microstructure analysis of Type-2 composites detected particles of C-S-H and ettringite phases. It was also noted that the samples without zeolitic waste showed no interaction between wood fibres and hydration products whereas, in the presence of zeolitic waste the compactness of the contact zone between wood fibres and cementitious hydration products was observed with better adhesion.
5. From the hydrothermal synthesis of silica-gel waste, synthetic zeolite was produced which mainly constituted of hydrosodalite. Type-3 composites were prepared by incorporating these produced synthetic zeolites into the wood fibres cement paste, the maximum compressive strength, flexural strength and density was reached by the samples which contained 1% of synthetic zeolite and treated wood fibres.
6. Comparing the three types of composites, wood fibres cement composite with ultrasonically treated wood fibres and 10% zeolitic waste (Type-2 composites) showed the most feasible results in terms of mechanical strength and physical properties. The investigation of the density of samples with treated wood fibres revealed that it was possible to produce lightweight composites with the densities ranging from 1000-1200 kg/m^3 in this research.

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