UDK 62-73:666.965
Original scientific article/Izvirni znanstveni članek

ISSN 1580-2949
MTAEC9, 58(1)53(2024)

SYNTHESIS OF CALCIUM SILICATE MATERIALS FROM THE RESIDUAL WASTE SLUDGE OF A WATER-PURIFICATION PLANT

SINTEZA KALCIJ-SILIKATNIH MATERIALOV IZ OSTANKOV ODPADNEGA BLATA, NASTALEGA V NAPRAVI ZA ČIŠČENJE ODPADNIH VOD

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Prejem rokopisa – received: 2023-05-15; sprejem za objavo – accepted for publication: 2023-12-15

doi:10.17222/mit.2023.876

Calcium silicate is produced from a mixture of silica sand powder, lime, paper pulp, and Portland cement hydrothermally steamed at 180 °C for about 16 h. This material is considered environmentally friendly and is popular in countries around the world. In this study, quartz sand was replaced with residual waste sludge from water-filtration plants to produce calcium silicate materials. Nowadays, the residual waste sludge from water-filtration plants is an environmental problem that needs to be treated. The results of determining the properties showed that a sample using 10 *w*/% residual waste sludge gave the best replacement. This sample had a bending strength of 10.95 MPa, a volumetric density of 1.57 g/cm^3, and water absorption of 23.67 %. The results of the analysis of the mineral composition (by X-ray diffraction analysis and Fourier-transform infrared spectroscopy) and microstructure (by scanning electron microscopy) showed that all samples formed tobermorite and xonotlite minerals. The tobermorite and the xonotlite are hydro-silicate-calcium minerals characteristic of calcium silicate materials, which are the synthesis products of chemical reactions of SiO_2, CaO, and H_2O under hydrothermal conditions. Samples using 5–10 *w*/% of waste residual sludge have even higher mechanical strength than samples without. Therefore, using waste residual sludge from water-filtration plants to replace part of the sand in producing calcium silicate materials can be considered an effective method to treat environmental problems caused by waste residual sludge.

Keywords: residual waste sludge, water-purification plant, hydrothermal reactions, tobermorite and xonotlite, Al-tobermorite

Kalcij-silikatne materiale se običajno proizvaja iz mešanice kremenovega (SiO_2)peska oz. prahu, kalcijevega hidroksida ($Ca(OH)_2$), papirne pulpe oz. kaše in hidrotermalno pri 180 °C in 16 urah s paro obdelanega Portland cementa. Tako izdelan material se ocenjuje kot zelo okolju prijazen in popularen material v večini držav tega sveta. V tem članku avtorji opisujejo postopek izdelave kalcij-silikatnega materiala pri katerem so kremenčev prah zamenjali z odpadnim blatom nastalim kot ostanek pri filtraciji odpadne vode. Dandanes odpadno blato, ki nastaja v obratih za filtracijo vode oz. odplak postaja okoljski problem, ki ga je potrebno rešiti. Rezultati raziskave so pokazali, da je optimalni dodatek odpadnega blata 10 *w*/%. Vzorec iz takšne mešanice je imel upogibno trdnost 10,95 MPa pri volumski gostoti 1,57 g/cm^3 in absorpciji vode 23,67 %. Rezultati analiz mineralne sestave in mikrostrukture s pomočjo rentgenskega difraktometra (XRD), Fourierjeve transformacijske infrardeče spektroskopije (FTIR) ter vrstične elektronske mikroskopije (SEM) so pokazali, da vsi vzorci vsebujejo tobermoritne in ksonotlitne minerale. Tobermorit in ksonotlit sta hidro-silikatno-kalcijeva minerala značilna za silikatne materale, ki nastajajo kot produkti kemijskih reakcij med SiO_2, CaO in H_2O v hidrotermalnih pogojih. Vzorci s 5 do 10 *w*/% odpadnega blata so imeli celo višje mehansko trdnost kot vzorci brez le-tega. Zato avtorji v članku povdarjajo, da preostanek odpadnega blata iz obratov za filtracijo vode lahko predstavlja delni nadomestk za kremenčev prah. Takšen način izdelave Ca-Si materialov se lahko smatra kot učinkovita metoda za rešitev okoljskih problemov zaradi ostankov odpadnega blata.

Ključne besede: ostanek odpadnega blata, obrat oziroma tovarna za čiščenje vode, hidrotermalne reakcije, tobermorit in ksonotlit, Al-tobermorit

1 INTRODUCTION

Residual sewage sludge (RWS) is a byproduct of water-filtration plants. It is not hazardous waste, but it occupies a large area of urban land for storage and landfill. Therefore, this waste needs to be treated. If handled properly, it will provide better economic and environmental performance. The RWS is aluminosilicate with fine particle size and consists of minerals like kaolinite, quartz, and hematite, which can be suitable raw materials for producing building materials. Many studies have looked at reusing RWS, such as unburnt construction materials[1] or treatment by polymerization.[2] In particular, the reuse of industrial waste as an additive for calcium silicate by hydrothermal autoclaving technology has been studied.[3] Building materials made in hydrothermal conditions are relatively lightweight, have lower thermal conductivity, higher heat resistance, lower shrinkage, and structure faster than conventional concrete.

The raw materials for hydrothermal solidification are usually natural aluminosilicate resources such as sand, felspar, clay minerals, $Ca(OH)_2$, and Portland cement.

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Hydrothermal solidification is carried out at 140–180 °C for 2–40 h. Under the hydrothermal reaction conditions, minerals like xonotlite and tobermorite will be formed. The solids' flexural strength will increase as the processing temperature and processing time increase, reaching values up to 20 MPa.[4]

The temperature and pressure conditions considered optimal for the hydrothermal process to fabricate industrial products such as autoclaved aerated concrete (AAC) and calcium silicate sheets are 180 °C, corresponding to a pressure of about 1 MPa.[5] In terms of microstructure, the tobermorite mineral ($Ca_5Si_6O_{16}(OH)_2.4H_2O$) is a significant contributor to the strength of these materials. The tobermorite is formed in the hydrothermal and strongly alkaline environment of the $CaO–SiO_2–Al_2O_3–H_2O$ system.[6] Co-forming with the tobermorite under hydrothermal conditions is xonotlite ($Ca_6Si_6O_{17}(OH)_2$).[6] However, tobermorite does not produce as high strength as xonotlite.

Hydrothermal materials are becoming more and more attractive due to their excellent properties and environmental friendliness. AAC is a combination of silica sand, Portland cement, gypsum, lime, water, and an expander. To improve the mechanical properties and reduce production costs, waste materials were used as raw materials.[7]

With AAC bricks, part of the raw gypsum is replaced with waste up to 30 % without affecting the functional and structural properties of the final product. Physical properties and sound absorption data confirmed that the addition of waste did not significantly alter the typical porosity of the AAC. It is even suggested that the optimum ratio of the raw-material mix consists of 55–65 % of construction waste.[8] The formation of tobermorite when $Ca(OH)_2$ reacts with kaolinite has also been investigated. Adding Al-containing compounds such as kaolinite to the mix also changed the reaction. Al^{3+} ions can replace Si^{4+} and produce Al-tobermorite. The formation of tobermorite will be increased, thus preventing the appearance of xonotlite.[9] Some other studies also indicated that the presence of Al^{3+} accelerates the tobermorite formation from CSH and inhibits its conversion to xonotlite,[10] even forming at lower temperatures.[11] Thus, wastes containing many Al^{3+} ions, such as the residual waste sludge, can enhance the mechanical strength of the material's alkaline $Ca(OH)_2$ activities under hydrothermal conditions.

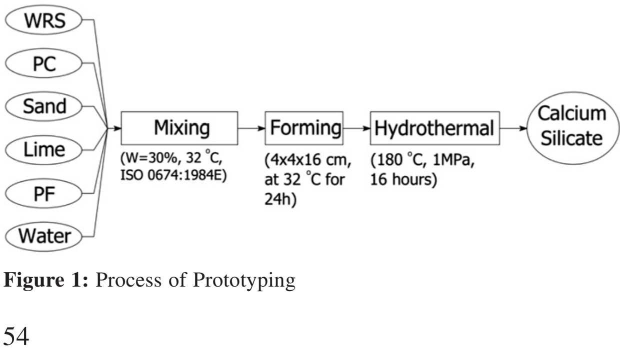

Figure 1: Process of Prototyping

In this study, the RWS from the Thu Duc water-filtration plant was used to partially replace the silica sand to produce calcium silicate sheets – the material for ceiling and partition panels. The method used to synthesize the calcium silicate is the hydrothermal method in a stainless-steel autoclave. The tobermorite crystals are formed from a mixture of Portland cement, lime, sand, paper fiber, and hydrothermal reaction in the autoclave at 180 °C and 1 MPa pressure for 16 h.

2 EXPERIMENTAL METHODOLOGY

The raw materials were used, including RWS, sand, lime, Portland cement (PC), paper fiber (PF), and water. The prototyping process is shown in **Figure 1**. They were mixed in different proportions, in which the residual waste sludge partially replaced sand in the range 0–25 *w*/%.

Samples were mixed with a moisture content of 30 % by mass at room temperature. The mixing equipment is a planetary machine model JJ-5. The mixing process is carried out according to ISO 0674:1984E standards. The mixture compounds were shaped in a steel mold with dimensions of 4 cm × 4 cm × 16 cm. The post-formed samples were cured in the air for 24 h. Finally, the samples were hydrothermally autoclaved to form calcium silicate at 180 °C and 1 MPa for 16 hours. These parameters, such as the processing temperatures and times, correspond to the production of calcium silicate sheets today. After hydrothermal treatment processing, the samples were determined to have various properties, including bending strength, volumetric density, and water absorption. The microstructure was determined through phase composition, functional group composition, and morphology.

The bending strength was determined on the Matest E183N device by the 3-point bending method with a loading speed of 25 N/s. Volumetric mass and water absorption were determined according to the ASTM C830 standard.

The phase composition was determined by X-ray diffraction (XRD) on the D8 Advance – Bruker instrument and using Cu–K_α radiation (λ = 0.154 nm). The analytical sample was a powder sample, scanning angle 2θ from 10° to 60°, scanning step 0.019°. An XRD pattern was also used to calculate the formed crystal composition. The calculation expression is shown in Equation (1), where, %C is the crystal content, S_c is the total area on the XRD pattern of the crystal, and ΣS is the total area of the XRD pattern.

$$\%C = 100\% \cdot S_c / \Sigma S \tag{1}$$

The functional group composition was determined by Fourier-transform infrared spectroscopy (FT-IR) on a Nicolet 6700 – Thermo Scientific instrument. Analytical conditions include KBr binder, scan angle from 450 cm^{-1} to 4000 cm^{-1}, and scan step at 0.964 cm^{-1}.

The morphology was determined by scanning electron microscopy (SEM) on a Hitachi S-4800 instrument. Analytical conditions were a voltage of 10 kV, low electronic mode SE(M), and magnification of 20,000 times.

3 RESULTS AND DISCUSSION

3.1 Raw materials and the mix compositions of the samples

The waste used to make the calcium silicate materials in this study was RWS. The RWS was taken from the Thu Duc water-purification plant in Ho Chi Minh City, Vietnam. The particle size of RWS was in the range of 6.720–77.339 µm. The average diameter was 14.259 µm. The chemical compositions (*w*/%) were 32.8 % SiO_2, 26.3 % Al_2O_3, 33.7 % Fe_2O_3, 0.22 % P_2O_3, 1.91 % K_2O, 0.38 % CaO, 0.59 % TiO_2, and 6.38% lost on ignition. Besides RWS, the remaining raw materials used to make the calcium silicate included sand, lime, and water. The mixed compositions of the samples are shown in **Table 1**. These samples were denoted by S0, S5, S10, S15, S20, and S25, corresponding to the amount of sand replacement by RWS from 0 *w*/% to 25 *w*/%.

The XRD pattern in **Figure 2** shows that the mineral compositions of the RWS were nacrite ($Al_2Si_2O_5(OH)_4$),[12] quartz (SiO_2),[13] diaspore (AlO(OH)),[14] and kaolinite ($Al_2Si_2O_5(OH)_4$).[15] Nacrite and kaolinite have the same chemical formula, but nacrite crystallizes in the monoclinic, while kaolinite is in the triclinic system. These are minerals commonly found in the composition of clay mud. In addition, the background of the XRD pattern in **Figure 2** also shows that the RWS pattern contains several components in the amorphous state. This composition may contain amorphous SiO_2, which enhances the RWS activity.[16] As a result, using RWS can increase the ability to form calcium silicate hydrate minerals, which helps to increase the strength of the calcium silicate materials after the hydrothermal autoclave.[17]

Table 1: Compositions of raw materials

Raw materials	Samples (*w*/%)					
	S0	S5	S10	S15	S20	S25
RWS	–	5	10	15	20	25
PC	39	39	39	39	39	39
Sand	56	51	46	41	36	31
Lime $Ca(OH)_2$	5	5	5	5	5	5
PF(*)	4.76	4.76	4.76	4.76	4.76	4.76

(*)Paper fibre was used for 4.76% of the solid material mass.

Some other materials were used in the experiments, such as sand with a composition of 99 % SiO_2, crushed to a particle size of less than 45 µm, and the Portland cement used was PC40.

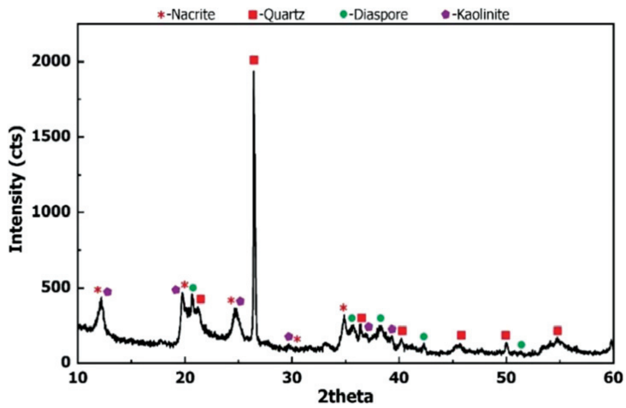

Figure 2: XRD pattern of RWS

3.2. Mechanical and physical characteristics

The mechanical and physical characteristics, such as bending strength, volumetric density, and water absorption, are presented in **Figure 3** and **Figure 4**.

The results of **Figure 3** show that the volumetric density decreased, and the water absorption increased when replacing the sand with RWS. Since sand and sludge

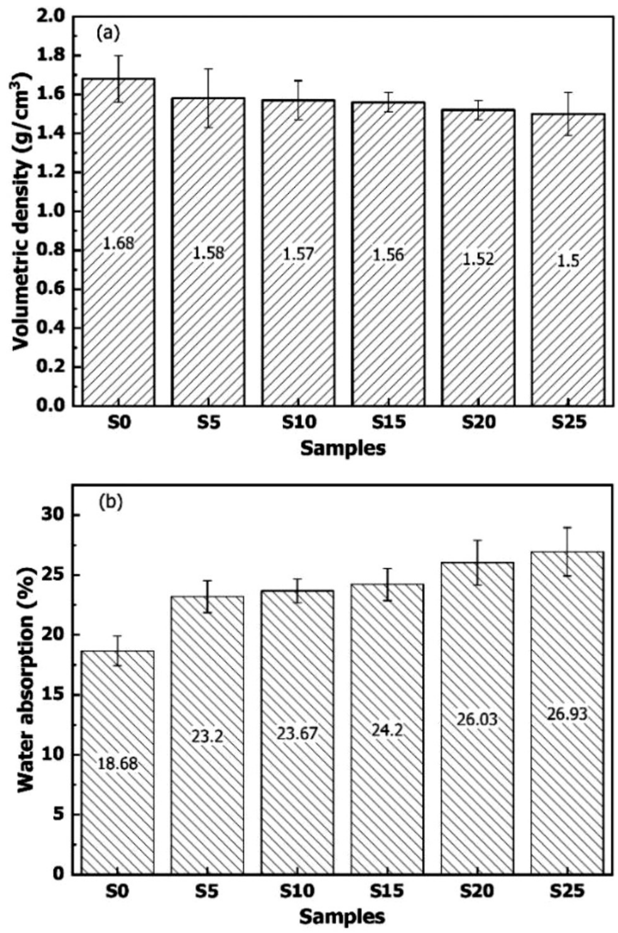

Figure 3: a) Volumetric density and b) water absorption of the samples

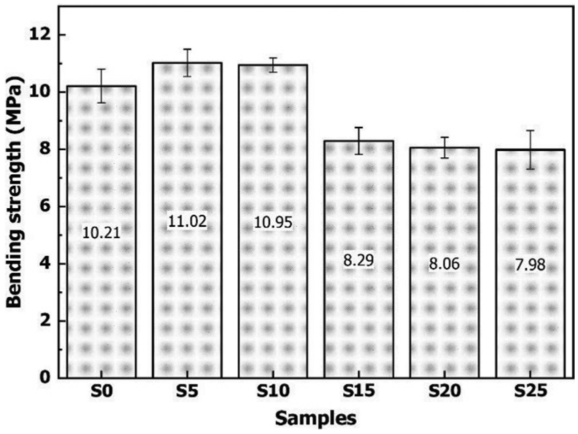

Figure 4: Bending strength of the samples

side would be washed away in water when determining the volumetric weight and water absorption according to ASTM C830. This decreased the volumetric density and increased the water absorption of the samples.

have different particle sizes, the replacement of sand with RWS also contributes to a change in the density of the particles in the aggregate composition. Therefore, the volumetric mass of the calcium silicate will also change. With an average sand grain size of 45 µm and an average sludge particle size of 14.259 µm, when replacing sand with RWS, the samples would have a tighter arrangement. As a result, the volumetric density of the sample increases, and the water absorption decreases. However, **Figure 3** shows that the volumetric density of the sample decreased, and the water absorption of the sample increased. As the results of the XRD pattern in **Figure 2** show, the composition of RWS contains a quantity of clay mud and impurities. When the amount of RWS used was too much, clay mud and fine impurities would interfere with the bonding between the aggregate particles. Therefore, in the calcium silicate samples after autoclaving, the clay particles and impurities on the outside would be washed away in water when determining the volumetric weight and water absorption according to ASTM C830. This decreased the volumetric density and increased the water absorption of the samples.

Figure 4 shows that the bending strength increased with the S5 and S10 samples, corresponding to the amount of sludge from 0 % to 10 % by weight. However, if the amount of used RWS was more than 10 %, the bending strength would be reduced compared to the S0 sample. The results show that the strength of the calcium silicate samples increased when replacing the sand with the RWS using a reasonable amount. The bending strength of the samples achieved is similar to the research conducted by M. Takagi et al.[18]

As shown in **Figure 2**, the composition of RWS can contain SiO_2, which is amorphous. This amorphous SiO_2 reacts more readily with lime than the crystalline SiO_2 of sand in a hydrothermal reaction. These reactions helped to form minerals that strengthen the calcium silicate. However, besides amorphous SiO_2, the sludge contains many clay mud and other fine impurities.[19] When using too much RWS, fine particles interfered with the components' bonding. Therefore, the strength of the sample after the hydrothermal process would be reduced when using RWS of more than 10 %.

3.3 The XRD and FT-IR

Figure 5 shows the FT-IR analysis results of the calcium silicate samples. The analysis of the samples showed the appearance of two groups of wave numbers. The wave-number group characterized the molecular vibrations of the O-H group. The other group represented the molecular vibrations of the silicate tetrahedra.

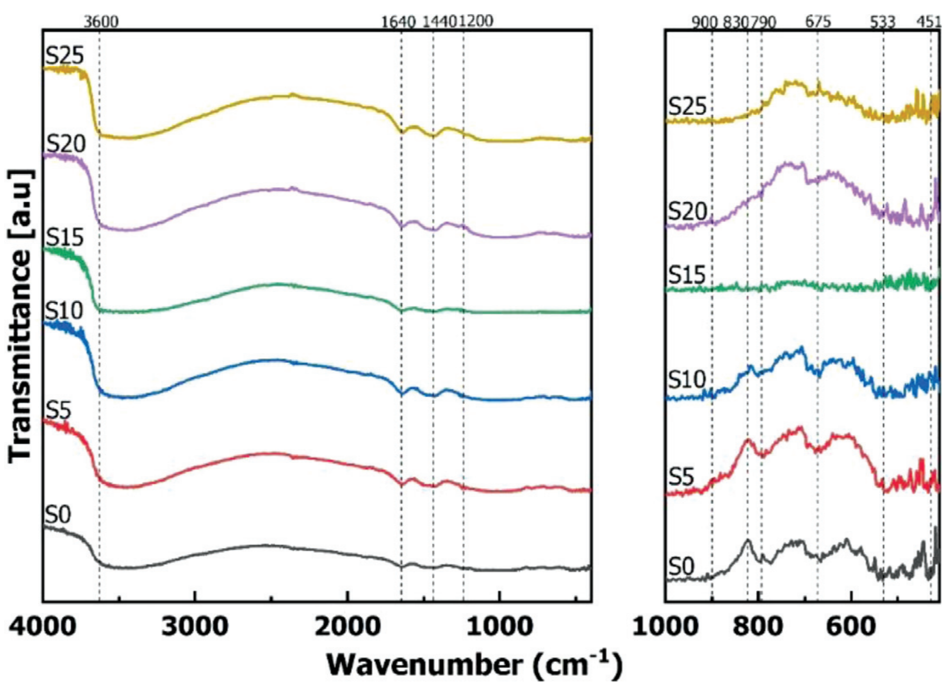

Figure 5: FT-IR spectra of the samples

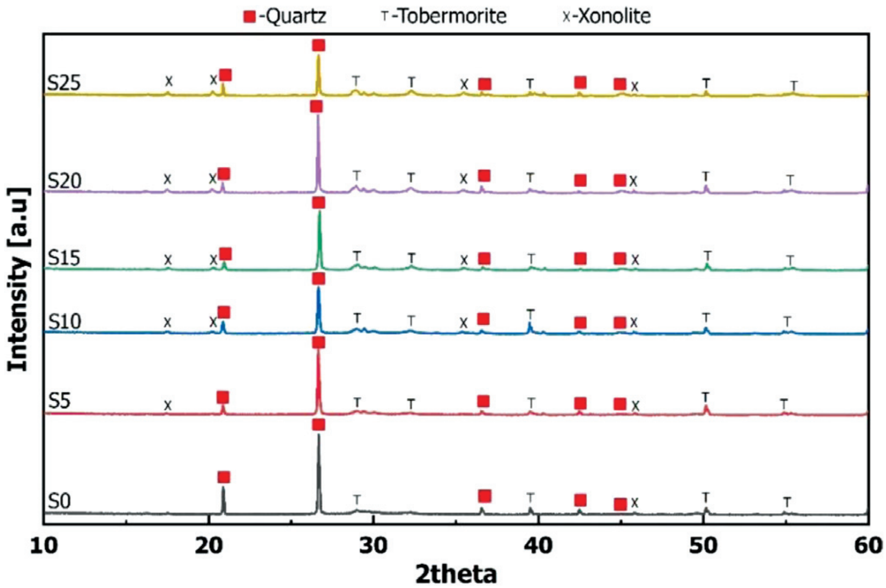

Figure 6: XRD patterns of the samples

The peak at 1640 cm^{-1} was attributed to the -OH vibration. It is the free-water molecules present in the sample. The second vibration from 3400 cm^{-1} to 3600 cm^{-1} is characteristic of the chemical water that existed in the structure of the mineral.[20] Combined with the bending vibration of the CaO-H group at wavenumber of 629–631 cm^{-1}, the chemical water formed was the hydroxyl group existing in the structure of the calcium silicate hydrate mineral.[21] The vibration at 675 cm^{-1} was also a characteristic of the bonding oxygen bridge between the silicate tetrahedra and the CaO polyhedron (Q2).[21]

With the silicate tetrahedra, the FT-IR spectra of all the samples showed the characteristic vibrations of $[SiO4]^{4-}$ in the structure of calcium silicate hydrate. In **Figure 5**, all the samples appeared to vibrate at wavenumber 1200–1220 cm^{-1}. This band is typical for the stretching vibration of Si–O at the junction between two silicate tetrahedra through an oxygen bridge (Q3)[22]. The stretching vibration of Si–O in the range 900–1000 cm^{-1}, the bending vibration of O–Si–O in the range 451 533 cm^{-1}, the bending vibration of Si–O–Si in the range 675–790 cm^{-1}, and the stretching vibrations of Si–O of the last tetrahedron in the bond chain $[SiO4]^{4-}$ in the range 800–830 cm^{-1} (Q1)[22] also showed the appearance of the silicate tetrahedra.[23]

The results of the FT-IR spectra showed that there was structure formation of calcium silicate hydrate in the samples after the hydrothermal autoclave. The XRD results in **Figure 6** indicate the formation of these calcium silicate hydrate minerals.

The results of the XRD patterns analysis showed the appearance of peaks characteristic of quartz, tobermorite, and xonolile minerals. Quartz[23] was the mineral in the material remaining in the samples after the reaction. The results also showed that crystalline SiO_2 hardly reacted with CaO to form calcium silicate hydrate under hydrothermal conditions. Meanwhile, tobermorite[24] and xonotlite[25] were two minerals formed after the hydrothermal treatment. These two minerals were the calcium silicate hydrate minerals shown on the FT-IR spectrum. Tobermotite and xonotlite would help increase the mechanical strength of the product. The XRD results in **Figure 6** also showed that the diffraction peaks of tobermorite and xonotlite on the S5–S25 samples appear more obvious than those of the S0 sample. This result proves that the sludge contains amorphous SiO_2. Amorphous SiO_2 readily reacts with CaO to form tobermorite and xonotlite. The highly developed tobermorite and xonotlite crystals make the diffraction peaks of the S5 and S10 samples more obvious than those of the S0 sample.

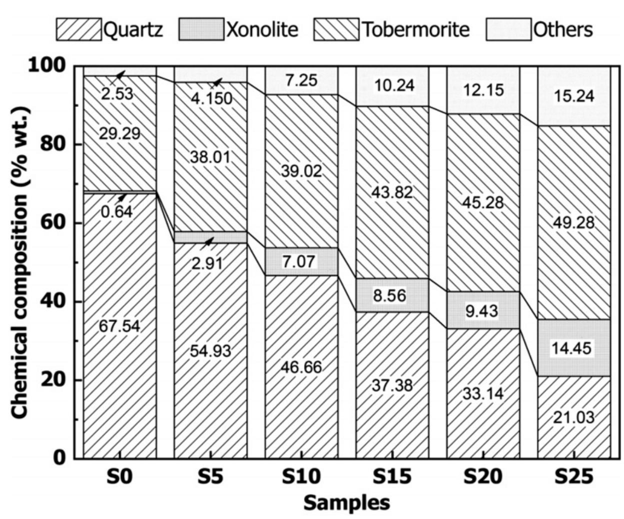

Figure 7: Chemical composition of the samples

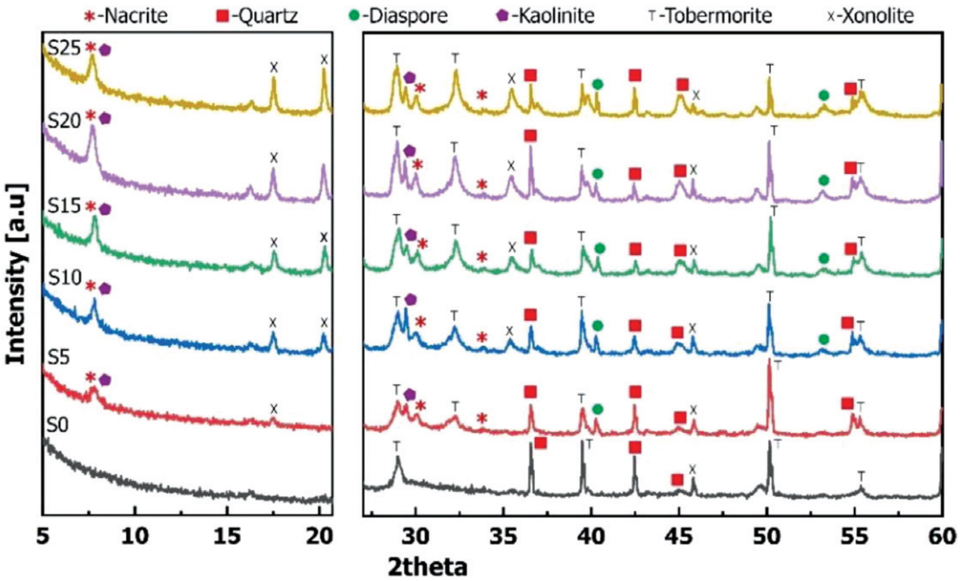

Figure 8: XRD patterns of the samples at 5°–20° and 27°–60°

The formation of minerals was also calculated from the XRD patterns (**Figure 6**) and Equation (1). **Figure 7** is the result of calculating the mineral composition. The decrease in quartz content shows that the quartz is mainly located in sand. When the sand content in the mixture is reduced, the quartz in the product is also reduced. The calcium silicate minerals (including xonotlite and tobermorite) in the product gradually increase as the RWS increases. For a sample containing 25 % RWS, the content of xonotlite and tobermorite accounts for 63.73 %, an increase of 112.93 % compared to the sample without RWS. This result shows the role of RWS in forming calcium silicate minerals by hydrothermal method. The RWS contains active substances that help make forming the calcium silicate easier. In addition, the tobermorite content was also higher than the xonotlite in all samples. This result is similar to the conclusions in previous studies. M. Diez-Garcia et al. also demonstrated that the main mineral formed would be tobermorite when the calcium silicate material is formed by the hydrothermal method at 170–200 °C and 1 MPa.[26] Besides, the composition of other minerals also increases when the amount of RWS increases. These minerals are usually minerals of clay mud. They reduce the strength of calcium silicate materials (**Figure 4**). The existence of clay minerals was shown on the XRD patterns in **Figure 8**.

Because the intensity of quartz mineral at position 26.62° is too high, it can mask other diffraction peaks. Therefore, **Figure 8** shows the samples' XRD result after cutting this diffraction peak. **Figure 8** showed that in the samples using RWS, besides quartz, tobermorite, and xonotlite, minerals such as nacrite, diaspore, and

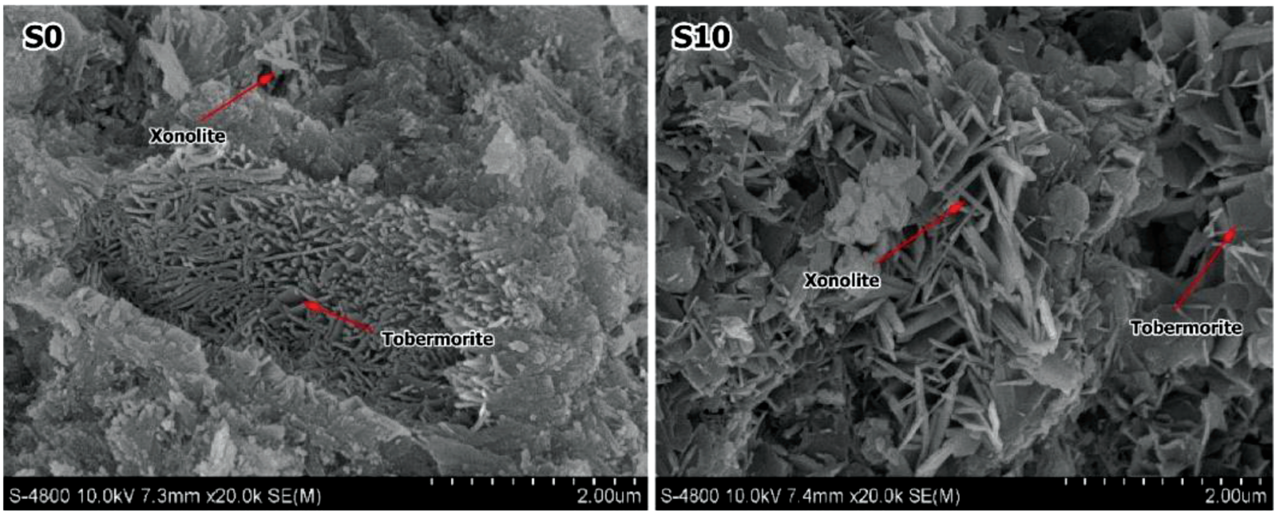

Figure 9: SEM images of the S0 and S10 samples

kaolinite also appeared. Nacrite, diaspore, and kaolinite were components of the clay mud present in RWS (XRD pattern in **Figure 2**). These minerals did not participate in forming calcium silicate hydrate and remained in the sample after the hydrothermal process. The existence of nacrite, diaspore, and kaolinite in the XRD results of the S5–S25 samples also explained the decreased bending strength (S15–S25 samples), decreased volumetric density, and increased water absorption, as shown in **Figure 3** and **Figure 4**.

3.4 Microstructure by SEM

The microstructures of the samples were also observed through scanning electron microscopy. **Figure 9** shows the SEM results of the samples.

The SEM images show that the samples were typical tobermorite and xonotlite crystals. Crystalline tobermorite was plate-shaped, and crystalline xonotlite was needle-shaped.[27] The crystal shapes of tobermorite and xonotlite are similar to those shown in the study of K. Luke et al. However, at the same magnification (20,000 times), the size and density of the tobermorite and xonotlite crystals of sample S10 were clearer and larger than the S0 sample. It again proved that the mineralization capacity of the RWS was higher than sand.

5 CONCLUSIONS

The results show that using 5–10 % of the RWS was an appropriate ratio to create calcium silicate materials in hydrothermal at 180 °C, 1 MPa for 16 h. The formed sample had a bending strength of 10.95 MPa, a volumetric density of 1.57 g/cm^3, and a water absorption of 23.67 %. The results of the mineral composition analysis by XRD and FT-IR showed that the primary minerals formed were quartz, tobermorite, and xonotlite. The tobermorite and xonotlite contributed to the mechanical strength of the calcium silicate materials. In addition, the XRD patterns of the samples that used RWS appeared as nacrite, diaspore, and kaolinite minerals. They were components of the clay mud present in the RWS. Therefore, if the amount of residual waste sludge were used too much (over 10 *w*/%), clay mud and fine impurities in the sludge would hinder the association between particles and reduce the mechanical strength of the product. SEM images also demonstrated that the sample using the RWS has a larger size and density of the tobermorite and xonotlite crystals than the untreated sample. The results show that RWS can enhance the formation of tobermorite and xonotlite minerals under hydrothermal conditions. Therefore, a hydrothermal process can be considered an effective method to treat the RWS of water-filtration plants.

Acknowledgment

We acknowledge Ho Chi Minh City University of Technology (HCMUT), VNU – HCM for supporting this study.

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