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DEVELOPMENT OF SUPPLEMENTARY CEMENTITIOUS MATERIALS BY MECHANOCHEMICAL ACTIVATION USING RAW MATERIALS FROM HUNGARY

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Supplementary cementitious materials (SCMs) are currently at the forefront of research due to the demand for cement in the rapidly growing construction industry and strict environmental regulations. Their application can efficiently reduce the energy required for production and CO_2 emissions. Trass and thermally activated kaolin (metakaolin) exhibit pozzolanic reactivity because of their high active silica and alumina content. Their application reduces the amount of cement clinker and the energy required for cement production, while providing beneficial properties to concrete. As a viable alternative to thermal activation for the production of SCMs, mechanochemical activation (MCA) is currently the subject of extensive research. In this work, MCA was performed by high-energy dry grinding mixtures of locally available kaolin and trass in the mass ratios of 25:75, 50:50, 75:25 and 100:0. X-ray diffraction, thermal analysis, infrared spectroscopy, scanning electron microscopy and specific surface area measurements were used to examine the structural as well as morphological changes that occurred during MCA. To characterize the pozzolanic reactivity, the compressive strength of binders was studied, in which 10% w/w of Ordinary Portland Cement (OPC) was replaced by activated mixtures. It was found that the addition of 25, 50 and 75% w/w of trass reduced the grinding time of the complete amorphization of the kaolinite from 90 mins to 75, 45 and 30 mins, respectively. Meanwhile, almost complete (~90%) amorphization was achieved for all mixtures by halving the grinding times. The 28-day-long compressive strength of the binders containing activated mixtures reached that of the OPC reference. Overall, it was concluded that the addition of trass positively reduced the grinding time and energy required for the amorphization of kaolinite, moreover, that SCMs with good levels of pozzolanic reactivity can be produced with 90% amorphization from local raw materials.

Keywords: kaolin, trass, mechanochemical, pozzolanic, local

1. Introduction

A variety of natural and calcined raw materials, e.g. trass and metakaolin, as well as industrial by-products or wastes like granulated blast furnace slag (GBFS) and concrete demolition waste can be utilized as partial replacements for Ordinary Portland Cement (OPC) in mortar and concrete [1],[2]. These replacements are one of the most effective ways to reduce annual global anthropogenic CO_2 emissions by around 7% resulting from cement production [3]. The availability of industrial by-products, e.g. fly ash, which is one of the most commonly used supplementary cementitious materials (SCMs), is strongly influenced by the output of the manufacturing industry. In regions where fly ash or GBFS is scarce or of low quality, the use of locally available pozzolans like trass should be considered to avoid the need for long-distance transportation and promote sustainability [3]. Trass, also known as volcanic tuff, is a naturally occurring pozzolan, which can be found in large quantities throughout Asia, Europe and America.

Metakaolin is a commonly used pozzolanic SCM produced by the thermal activation (TA) of kaolin [1]. According to industrial data, the amount of energy required for the TA of kaolin is around 1600 kWh/t [4]. Therefore, alternative methods such as mechanochemical activation (MCA) should be considered. The MCA of kaolin can break the O-H, Al-OH, Al-O-Si and Si-O bonds of kaolinite, resulting in an amorphous phase and weakly bound coordinated water [5]. The mechanochemically activated kaolin has been shown to exhibit advantageous pozzolanic reactivity, which is competitive with that of the thermally activated equivalent [4],[6]. However, MCA can fundamentally alter the plate-like morphology of kaolinite, resulting in aggregates or agglomerates of nano-sized spherical particles [6]. Compared to TA, one advantage of MCA is that its energy demand is only between 200-1000 kWh/t [4]. MCA can be carried out in high-energy mills such as stirred media and planetary ball mills, which are suitable

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Table 1: Chemical composition of the kaolin and trass (LOI: loss on ignition)

Component	Kaolin (% w/w)	Trass (% w/w)
SiO_2	57.91	69.76
Al_2O_3	26.31	11.18
Fe_2O_3	2.12	0.20
CaO	0.59	1.20
K_2O	1.09	4.23
MgO	0.32	0.49
Na_2O	<0.01	3.18
TiO_2	0.93	0.90
P_2O_5	<0.01	3.49
SO_3	0.06	1.25
LOI	10.62	3.12

for laboratory and even industrial applications [7],[8]. Planetary ball mills are frequently used for MCA and their effectiveness depends on several factors such as the material the grinding balls and jars are composed of, the ball-to-powder ratio, the grinding time as well as the rotational speed [5],[6],[9],[10].

In our previous studies, the MCA of kaolin together with inert quartz and pozzolanic silica fume, trass as well as diatomaceous earth effectively accelerated the amorphization of kaolinite, reducing the energy consumption [5],[9],[10]. Nowadays, the production of high-activity SCMs using local raw materials is of great importance to reduce the environmental impact of transportation [3]. In this work, the MCA of mixtures of locally available kaolin and trass is investigated to produce amorphized materials with high pozzolanic reactivity over reduced grinding times while consuming less energy. In the present study, the effect of the grinding time and trass content on the amorphization efficiency and pozzolanic reactivity of the amorphized samples using 28-day-long compressive strength tests is studied.

2. Experimental

2.1. Materials

A natural kaolin from Sárisáp in Hungary was used in the experiments. The major crystalline phases of the kaolin were kaolinite (64% w/w) and quartz (24% w/w) but illite (7% w/w) was also present as a minor constituent. An abrasive pozzolanic material, namely naturally occurring trass from Pálháza in Hungary, was also used, 92% of which consisted of a glassy phase. The minor crystalline phases of the trass were cristobalite (1% w/w), quartz (<1% w/w) and albite (6% w/w).

The chemical compositions of kaolin and trass were measured by a Philips Axios PW 4400/24 wavelength dispersive X-ray fluorescence spectrometer, the chemical compositions of which are shown in *Table 1*.

To investigate the amorphization effect of trass, 100, 75, 50 and 25 g of kaolin (Sk) were mixed with 0, 25, 50 and 75 g of trass (Tr). The symbols of the samples containing 100, 75, 50 and 25% w/w of kaolin are Sk, 75Sk, 50Sk and 25Sk, respectively.

OPC (CEM I 42.5 N, Duna-Dráva Cement Kft., Beremend, Hungary) and standard sand (Cemix Hungary Kft., Hungary) were used to prepare samples of standard cement mortar in order to measure the pozzolanic reactivity. The chemical composition of OPC in % w/w was MgO, 1.61; CaO, 61.97; SiO_2, 20.54; Fe_2O_3, 3.26; K_2O, 0.52; Na_2O, 0.11; Al_2O_3, 5.40; TiO_2, 0.27; SO_3, 3.16 and LOI, 2.27. This cement had a Blaine fineness of 3720 cm^2/g.

2.2. Mechanochemical activation

To achieve mechanochemical activation, a Fritsch Pulverisette 5/4 planetary mill was used. According to the previous research [9], the rotational speed of 330 rpm and the ball-to-powder mass ratio (BPR) of 14:1 were used to activate each mixture. To facilitate grinding, 500-cm^3 hardened stainless-steel (Fe-Cr) jars and grinding balls with a diameter of 10 mm were used. As grinding progressed, the amorphization of kaolinite in the activated mixtures was primarily monitored by X-ray diffraction (XRD).

2.3. Thermal activation

The thermal activation (TA) of kaolin was also carried out as a comparison. Before TA, the phase transformation and changes in mass of kaolin during heating were investigated using a MOM Q 1500D Derivatograph. For this thermal analysis, 260 mg of kaolin was measured in a corundum crucible under a static air atmosphere with high-grade corundum as a reference at a maximum heating temperature of 1000 °C and a heating rate of 10 °C/min.

Two mass loss steps were determined on the derivative thermogravimetric (DTG) curve of the raw kaolin in *Figure 1*. The first mass loss step (119 °C, 1.51% w/w) is associated with the removal of physically adsorbed water, while the second (591 °C, 8.31% w/w) corresponds to the dehydroxylation of kaolinite, where

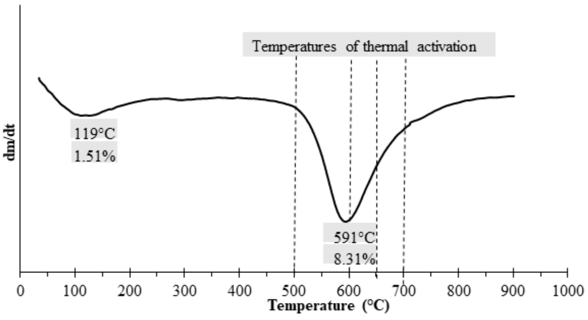

Figure 1: DTG curve of the raw kaolin with DTG peak temperatures, percentage mass losses between 40 and 350°C and treatment temperatures of 350–900°C indicated

the structural OH groups are removed, leading to the formation of metakaolinite. The conversion of kaolinite into metakaolinite is crucial to enhance pozzolanic activity [4],[6]. In *Figure 1*, the vertical dashed lines indicate the temperatures selected for the heat treatment (500, 600, 650 and 700 °C), which cover the range of metakaolinite formation. Considering the DTG results, the TA was carried out by calcining the raw kaolin at 500, 600, 650 and 700 ºC for 0.5, 1.0 and 2.0 h in a programmable electric furnace using a heating rate of 300 °C/h.

2.4. Analytical methods

XRD is one of the most important methods to follow the amorphization of kaolinite during MCA. XRD measurements were performed by a Philips PW 3710 diffractometer with a PW 3020 vertical goniometer and a curved graphite diffracted beam monochromator installed. The XRD patterns were recorded with CuKα radiation (λ=0.1541 nm) generated at 50 kV and 40 mA. In order to characterize phase changes, a continuous scan mode with a step size of 0.02° 2θ and a time per step of 1 s was applied within the range of 5-40° 2θ. X'Pert Data Collector software was used for the controlled measurement of XRD patterns, while X′Pert HighScore Plus software was applied to identify phases and determine the integral intensity of an individual diffraction peak. To characterize the efficiency of MCA, the degree of amorphization (DOA) was determined using the following equation [11]:

$$\mathrm{DOA} = \left(1 - \frac{A_i}{A_0} * 100\right) \qquad (1),$$

where A_0 denotes the area of the 001 reflection of raw kaolinite and A_i stands for the area of the 001 reflection of ground kaolinite.

Fourier transform infrared (FTIR) spectroscopic measurements were taken with a Bruker Vertex 70 spectrometer using a Bruker Platinum ATR adapter without additional sample manipulation. FTIR spectra were recorded with a room temperature DTGS detector at a resolution of 2 cm^{-1}. In each case, an average of 512 scans were recorded to increase the signal-to-noise ratio. The GRAMS/Al 9.0 spectroscopy software (Thermo Fisher Scientific, USA) was used to manage and analyze the spectral data.

The morphology of the kaolin, trass and activated mixture was investigated using a Thermo Fisher Scientific Apreo S scanning electron microscope (SEM). The SEM images with magnifications of 1000x and 20000x were obtained with a secondary electron (SE) detector at an accelerating voltage of 2 kV.

The specific surface area (SSA) of the samples was determined by the Brunauer-Emmett-Teller (BET) method [12] with nitrogen adsorption at −196°C using a Micromeritics 3Flex instrument. The samples were kept under a vacuum at 30 °C for 24 h before measurements were taken.

The pozzolanic activity was characterized by measuring the compressive strength of the binders according to a standard method MSZ EN 196-1:2016 [13], where the binder contained 90% w/w OPC and 10% w/w activated sample. For compressive strength measurements, mortar samples were prepared from 1350 g of standard sand, 450 g of binder and 225 g of water. As a comparison, a reference mortar (Reference) containing only OPC was also tested. The fresh mortar was cast into molds (three 40 x 40 x 160 mm prisms) and compacted using a vibration table. The specimens were kept in the mold at 90% relative humidity for 24 h before being placed in water for 28 days under standard curing conditions. After 28 days, the compressive strength was measured with a CONTROLS AUTOMAX 5 machine according to MSZ EN 196-1:2016.

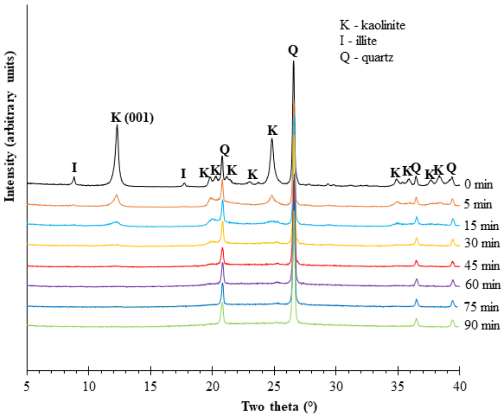

Figure 2: XRD patterns of the kaolin ground for 0, 5, 15, 30, 45, 60, 75 and 90 mins

3. Results and discussion

3.1. XRD analysis

At first, XRD was used to examine the effect of adding trass on the mechanochemical amorphization of kaolinite. XRD patterns of samples containing varying amounts of trass, namely Sk, 75Sk, 50Sk and 25Sk, were recorded over different grinding times. The XRD patterns of the kaolin (Sk) and the mixture containing 25% w/w of kaolin (25Sk) ground over different times are shown in *Figures 2* and *3*, respectively. The XRD pattern of the raw kaolin (*Figure 2*, 0 min) indicates that the reflections of kaolinite (K) and quartz (Q) are strong as they are the major phase constituents, while the reflections of illite (I) are small as it is the minor phase constituent.

In the XRD patterns of the ground kaolin samples (*Figure 2*, 5-90 min), it can be seen that by increasing the grinding time, the kaolinite (K) reflections broadened and their intensity (area) decreased significantly due to the reduction in crystallite size in addition to the delamination and degradation of the crystal structure [4],[6]. Besides the kaolinite reflections, the illite (I) reflections also decreased in intensity and broadened when subjected to MCA, while the quartz (Q) reflections

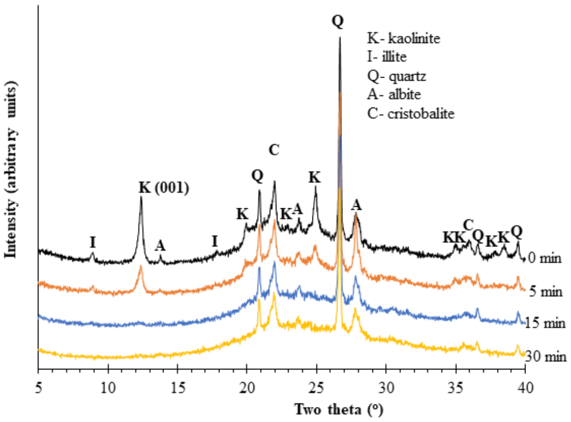

Figure 3: XRD patterns of the 25Sk mixture ground for 0, 5, 15 and 30 mins

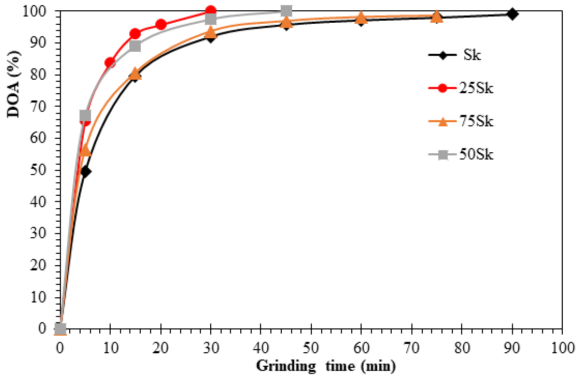

Figure 4: Changes in the DOA of kaolinite as a function of the grinding time for the samples containing different amounts of trass

remained practically unchanged. After 90 mins of grinding, all the kaolinite and illite reflections disappeared, moreover, only a broad peak between 20 and 30° 2θ remained, indicating total amorphization of the kaolinite and illite phases. These results are consistent with previous studies [6],[10]. In *Figure 3*, the XRD patterns of the ground 25Sk mixture also reveal the rapid delamination as well as amorphization of the kaolinite and illite phases, while the degradation of albite (A) and cristobalite (C) is less pronounced. In this case, the kaolinite and illite reflections vanished after only 30 mins of grinding, while the broad peak around 22° 2θ remained, demonstrating a faster amorphization process of the kaolinite and illite phases.

To compare the amorphization process of kaolinite in the samples containing different amounts of trass, the degree of amorphization (DOA) was determined using the 001 reflections of the XRD patterns (*Figure 4*). Over shorter grinding times, the DOA increases rapidly and then more slowly as the grinding progresses. For all samples containing trass, the DOA reaches or exceeds 80% within the first 15 mins of grinding. Analyzing the numerical data, it can be stated that even with a low trass content (25% w/w), the complete a morphization of kaolinite occurred within 75 mins, which is 15 mins less than for the raw kaolin (Sk). However, this low trass content resulted in only a small shift, while the higher trass contents of 50 and 75% w/w resulted in a much larger shift in the DOA curve towards shorter grinding times. At these higher contents of trass, the grinding time required for the complete amorphization of kaolinite was further reduced from 75 to 45 and 30 mins, demonstrating that the addition of hard and abrasive trass can significantly reduce the activation time of the complete amorphization of kaolinite by acting as grinding bodies to enhance the degradation of the kaolinite crystal structure [9]. Our previous study [10] showed that the nearly complete ~90% amorphization of kaolinite was favorable for the production of highly reactive pozzolanic SCM. Looking at the DOA curves in *Figure 4*, it can be seen that with ~90% amorphization, a significant reduction in grinding time, e.g. from 90 to 30 mins for the Sk sample, can be achieved. All in all, MCA with the addition of trass resulted in a similar amorphization process in the Sk (Hungary) as in the previously studied kaolin from Zettlitz (Czech Republic) [9]. However, under the same grinding conditions and with a low trass content, a slightly higher DOA was obtained for Sk than for kaolin from Zettlitz, probably due to the higher quartz content of the Sk.

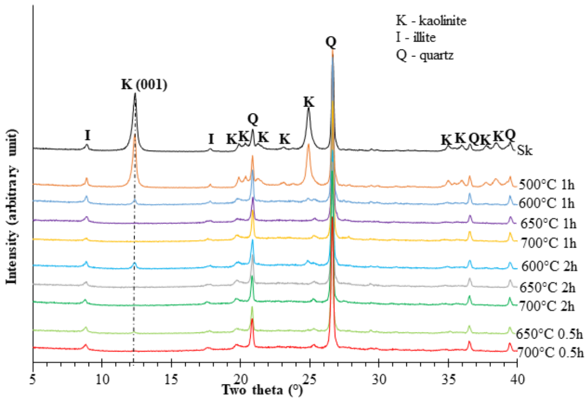

Figure 5: XRD patterns of the thermally treated kaolin at various heat treatment temperatures and after different treatment times

XRD was also used to study the TA of the Sk. In *Figure 5*, the XRD patterns of thermally activated kaolin when treated at 500 and 600 °C for 1 h show that the kaolinite peaks reduced, while a broad peak appeared as well as increased between 15 and 35° 2θ, indicating the dehydroxylation of kaolinite and the formation of metakaolinite [6],[11]. After TA at 650 and 700 °C for 1 h, the kaolinite peaks completely disappeared, confirming the full conversion of kaolinite into metakaolinite. Therefore, changing the temperature of the heat treatment had a significant effect on the dehydroxylation of kaolinite, demonstrating that it is a dominant factor. After a longer treatment at 600 °C for 2 h, the kaolinite peaks remained almost practically the same as over the shorter treatment time of 1 h, suggesting that increasing the duration in this case did not affect dehydroxylation. Varying the treatment time from 0.5,

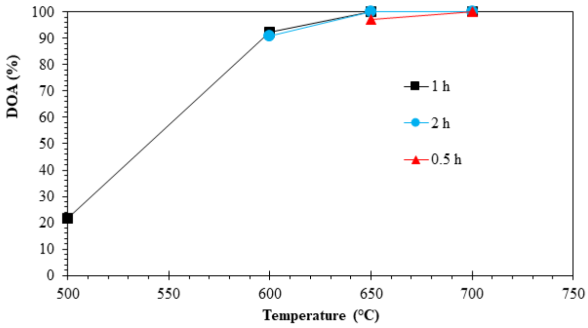

Figure 6: Changes in the DOA of kaolinite as a function of the heating temperature for the Sk sample after different treatment times

1 h to 2 h at both 650 and 700 °C had a minimal effect on the thermal transformation of kaolinite, indicating that the treatment time is a less dominant factor. In the studied TA processes, the quartz and illite peaks remained unchanged, demonstrating that these phases were not transformed within this temperature range.

To quantify the transformation of kaolinite into metakaolinite, the DOA values were determined from the reduction in the area of the 001 reflection using *Equation 1* and *Figure 6* which clearly shows that by increasing the heating temperature from 500 to 600 °C increased the DOA by more than 4 times. Further increasing the temperature only slightly increased the DOA by approximately 10%. Between 600 and 700 °C, changing the treatment time only slightly modified the DOA. Considering the results, the kaolin treated at 700 °C for 1 h (hereafter referred to as the Sk 700 °C_1h) was chosen for further tests for the purpose of comparison.

3.2. FTIR spectroscopic analysis

FTIR spectroscopy was used to characterize the changes in the molecular structures of the almost completely (~90%) and the completely (100%) mechanochemically amorphized samples presented in *Figures 7* and *8*.

In *Figure 7*, within the range of 3800-2500 cm^{-1}, the variations in the inner surface OH stretching bands (3688, 3667 and 3653 cm^{-1}) and the inner OH stretching band (3619 cm^{-1}) of the raw kaolinite can be followed [5],[6],[11]. The FTIR spectra of the ~90% amorphized samples (Sk_30 min, 75Sk_30 min, 50Sk_15 min and 25Sk_15 min) show the disappearance of the bands at 3667 and 3653 cm^{-1}, a large decrease in the intensity of the bands at 3688 and 3619 cm^{-1} as well as the appearance of a new broad band at around 3300 cm^{-1} due to the transformation of the structural OH groups to weakly bonded gel water [6],[10]. In the FTIR spectra of the 100% amorphized samples (Sk_90 min, 75Sk_75 min, 50Sk_45 min and 25Sk_30 min), all the characteristic OH stretching bands of kaolinite completely disappeared and only the new wide OH stretching band was visible, indicating the complete conversion of the OH groups. The FTIR spectra of the TA kaolin sample (not presented) exhibit a quite different pattern, where the OH

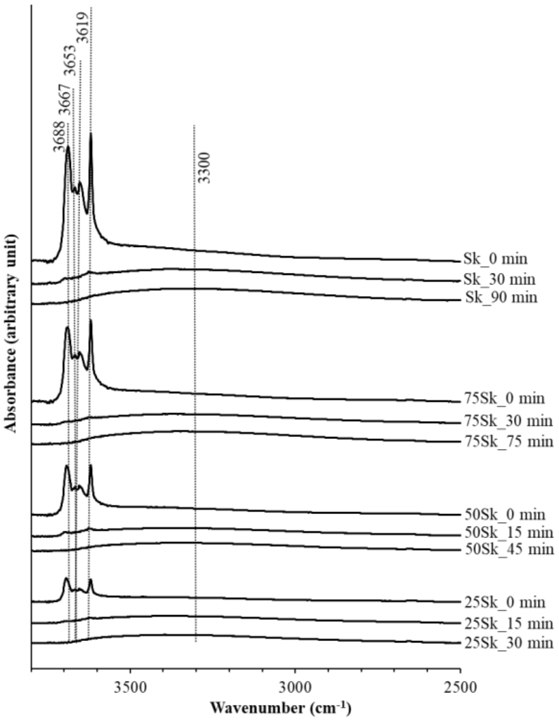

Figure 7: FTIR spectra of the unground and ground samples within the range of 3800-2500 cm^{-1}

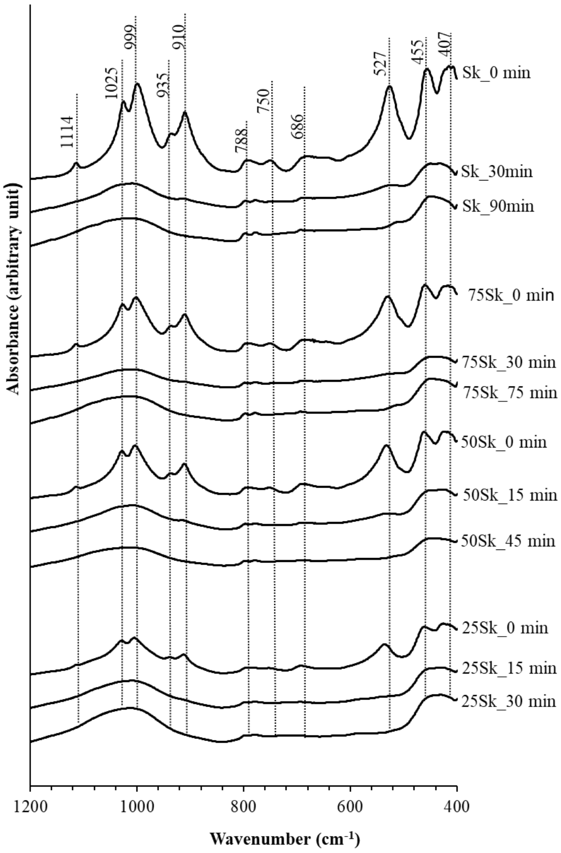

Figure 8: FTIR spectra of the unground and ground samples within the range of 1200-400 cm^{-1}

stretching bands of kaolinite have completely disappeared due to complete thermal dehydroxylation.

In *Figure 8* within the range of 1200-400 cm^{-1}, the Si-O stretching bands (1114, 1025 and 999 cm^{-1}), the OH deformation bands (935 and 910 cm^{-1}) as well as the lattice vibration bands (788, 750, 686, 527, 455 and 407 cm^{-1}) also exhibit important information about the mechanochemical structural changes of kaolinite [5],[6],[11]. In the spectra of the almost completely amorphized samples (Sk_30 min, 75Sk_30 min, 50Sk_15 min and 25Sk_15 min), a significant decrease in the intensity of the OH deformation bands of the inner surface and inner hydroxyls at 935 and 910 cm^{-1} is observed, indicating the breakage of Al-OH bonds [6],[10]. In the FTIR spectra of the fully amorphized samples (Sk_90 min, 75Sk_75 min, 50Sk_45 min and 25Sk_30 min), both OH deformation bands disappeared completely.

In addition, the ~90% and 100% amorphization of kaolinite transformed the SiO stretching bands into a single broad band at approximately 1000 cm^{-1}, while the intensity of the lattice vibration bands was strongly reduced, signifying a drastic structural alteration in tetrahedral and octahedral sheets [5],[6]. In the FTIR spectrum of the TA kaolin (not shown), only wide absorption bands were observed at approximately 1030, 800 and 450 cm^{-1}, which can be to the random structural arrangement of (Si–O) and (Al–O) stretching and bending vibrations [10],[14].

3.3. SEM analysis

SEM analysis was used to characterize the morphological changes of the almost completely and completely mechanochemically amorphized samples (*Figure 9*) [6],[10]. The SEM image of the raw kaolin (A) shows the plate-like structure typical of kaolinite with individual kaolinite plates forming stacks. The SEM image of the raw trass (B) consists of irregular and coarse particles with smooth edges over a wide range of particle sizes. The SEM images of the ~90% amorphized kaolin (C) show that the kaolinite plates were destroyed after 30 mins of grinding and compact aggregates or agglomerates of nano-sized spheroidal particles were produced [10],[14]. The SEM images of the ~90 and 100% amorphized 75Sk sample (D and E) show that particles with different sizes as well as the shapes of the raw kaolin and trass were transformed into uniform round primary particles, which form larger secondary particles due to aggregation and agglomeration. In these samples, the kaolinite and trass particles were morphologically indistinguishable. The SEM images of the 50Sk and 25Sk mixtures (not presented) show similar changes in the morphology of the raw materials after ~90 and 100% amorphization. However, as the trass content increased, it was observed that the transformation in the size and morphology of the raw particles took place over a shorter and shorter time period. The SEM image of the TA kaolin (F) was completely different to that of the MCA kaolin (C-E) as the characteristic plate-like shape was retained after the heat treatment. The morphological properties of the mechanochemically and thermally activated kaolin from Sárisáp observed here are in good agreement with previous results obtained with other kaolins [11],[14].

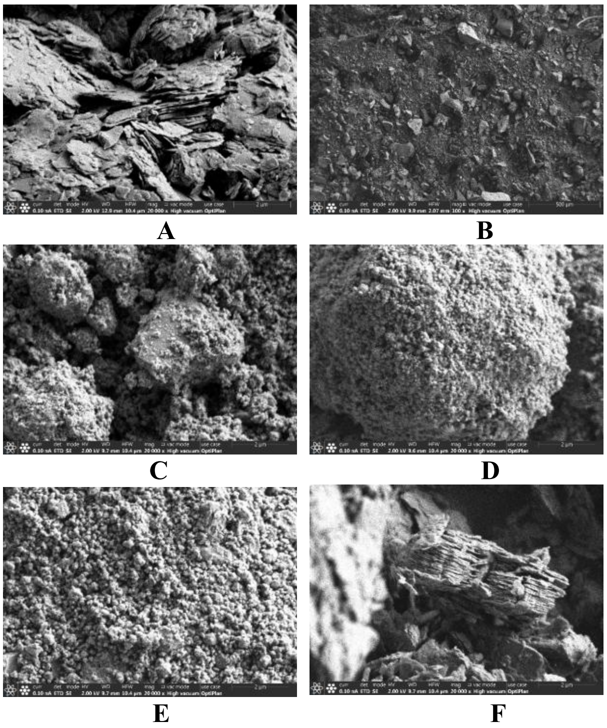

Figure 9: SEM images of the samples: Sk (A), Tr (B), Sk ground for 30 mins (C), 75Sk ground for 30 and 75 mins (D-E) and Sk heat treated at 700 °C for 1h (F)

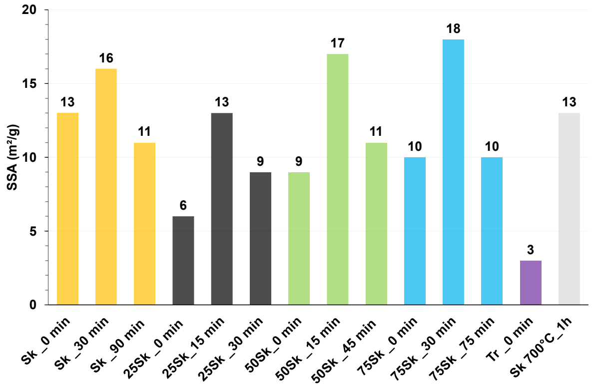

Figure 10: Changes in the SSA as a function of grinding time and heat treatment

3.4. Analysis of the specific surface area

The specific surface areas (SSA) of the activated and non-activated samples are presented in *Figure 10*. SSA ranged from 3 to 18 m^2/g based on sample composition and treatment. The SSA values of the raw kaolin and trass were 13 and 3 m^2/g, respectively. A general trend was that the grinding time (~90 and 100% amorphization of kaolinite) influenced the SSA significantly. For example, the SSA of the unground 25Sk mixture was 6 m^2/g, which

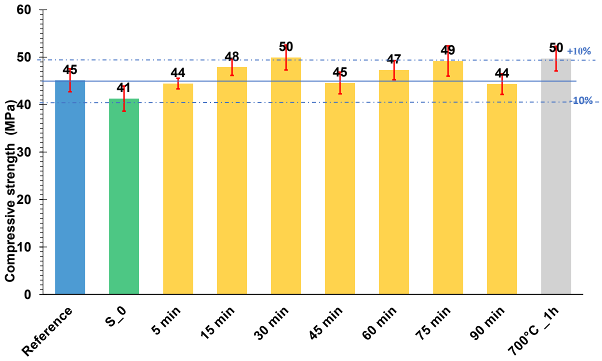

Figure 11: The 28-day-long compressive strength of the OPC (Reference) and the binder containing the quartz sand (S_0), the Sk sample ground for 5-90 mins or heated at 700°C for 1 h (the blue dashed lines indicate the ±10% range of compressive strength of the Reference)

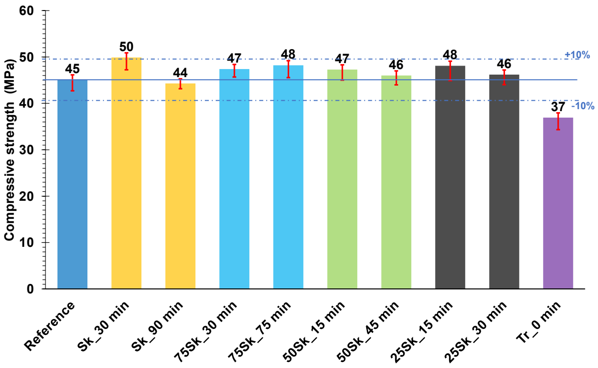

Figure 12: The 28-day-long compressive strength of the OPC (Reference) and the binder containing the Sk sample ground for 30 and 90 mins as well as the 75Sk, 50Sk and 25Sk mixtures ground for 15, 30, 45 and 75 mins (the blue dashed lines indicate the ±10% range of compressive strength of the Reference)

increased to 13 m²/g after 15 mins of grinding (~90% amorphization) and decreased to 9 m²/g after 30 mins (100% amorphization), suggesting that initially the reduction in primary particle size dominated, followed by strong agglomeration of the particles. The SSA of the 50Sk and 75Sk mixtures varied in a similar manner with the grinding time and DOA of kaolinite. The shorter grinding time for ~90% amorphization always resulted in a higher SSA than for the 100% amorphization as agglomeration was still negligible. Comparing different samples, the highest SSA of 18 m²/g was obtained for the 75Sk sample after 30 mins of grinding, that is, ~90% amorphization. The TA of kaolin (Sk 700°C_1h) resulted in an SSA of 13 m²/g, which was the same as that of the raw kaolin (Sk_0 min) but lower than that of the ~90% amorphized kaolin (Sk_30 min). This suggests that although the TA modified the crystal structure of kaolinite, it did not change its particle size nor morphology. In summary, the results show that shorter grinding times, resulting in ~90% amorphization, positively increased the SSA but longer grinding times, achieving 100% amorphization, decreased it due to significant agglomeration [9].

3.5. Analysis of compressive strength

The 28-day-long compressive strength of binders containing the mechanochemically and thermally activated kaolin are illustrated in *Figure 11*. The reference OPC had a compressive strength of 45 MPa, which was reduced by approximately 10% to 41 MPa by adding 10% w/w of inactive quartz sand (S_0), indicating that it acted as a filler. In the case of MCA, increasing the grinding time to between 5 and 30 mins increased the compressive strength due to the formation of the active amorphous phase as shown by XRD analysis. For grinding times between 45 and 90 mins, the compressive strength fluctuated but remained close to or above the reference value, indicating that these grinding times no longer increased the pozzolanic activity, probably due to increased agglomeration. After 30 mins of grinding, resulting in ~90% amorphization, the highest compressive strength of 50 MPa was obtained, indicating the optimal activation level. With this activation at ~90% amorphization, an 11% increase in compressive strength was achieved with the Sárisáp kaolin, while previously with the Zettlitz kaolin, only a 7% increase was achieved after 240 mins of activation which achieved 100% amorphization [10]. The TA of Sárisáp kaolin (Sk 700°C_1h) also resulted in a high compressive strength of 50 MPa, almost identical to that obtained after 30 mins of MCA. This suggests that the pozzolanic reactivity achieved by the TA at 700°C after 1 h is equivalent to that achieved after 30 mins of MCA.

The compressive strength of binders made with the raw trass as well as activated mixtures containing kaolin and trass are shown in *Figure 12*. In general, the activated mixtures yielded better compressive strengths than the reference OPC and the binder containing the raw trass, indicating that pozzolanic reactivity increased by grinding the two materials together. Shorter grinding times achieving ~90% amorphization (75Sk_30 min, 50Sk_15 min, 25Sk_15 min) mostly resulted in higher levels of compressive strength than those that resulted in 100% amorphization (75Sk_75 min, 50Sk_45 min, 25Sk_30 min) as no noticeable agglomeration had occurred up to that point. Regardless of the amount of trass, the compressive strength was between 46 and 48 MPa, which exceeded the reference value. The highest compressive strength of 48 MPa was obtained for the 75Sk_75 min and 25Sk_15 min samples, but both were approximately 4% lower than that of the Sk_30 min sample. This indicates that although partial replacement with trass resulted in a relatively high compressive strength, the performance of pure activated kaolin was not exceeded. However, the addition of trass significantly reduced, sometimes halved, the grinding time and energy required for ~90% amorphization by accelerating amorphization of the kaolinite phase owing to the fact that the trass particles acted as grinding bodies, which accelerated and enhanced the amorphization of kaolinite [9],[10]. Note that previously the addition of diatomaceous earth to the Zettlitz kaolin led to an increase in compressive strength of approximately 10% resulting in ~90% amorphization [10], which is close to the 7% increase obtained by adding trass to the Sárisáp kaolin.

4. Conclusions

In the present work, the mechanochemical activation of local kaolin from Sárisáp with the addition of local trass from Pálháza and the application of the activated mixtures as SCMs were investigated. The addition of trass accelerated the amorphization of the kaolinite phase, which significantly reduced the grinding time required for complete (100%) and almost complete (~90%) amorphization. The glassy trass particles, acting as grinding bodies, had almost the same effect on the mechanochemical amorphization of kaolinite as grains of crystalline quartz or amorphous diatomaceous earth [10]. A significant reduction in the grinding time from 90 to 15 mins was achieved by combining ~90% amorphization with the addition of trass. Due to this almost complete amorphization, the Si-O, Al-O-Si and Al-OH bonds of kaolinite were mostly broken, moreover, a significant proportion of the structural OH groups were transformed into gel-water, forming reactive silica and alumina groups. Meanwhile, the typical kaolinite and trass particles were transformed into nano-sized spherical ones of similar size which were yet to form strongly bound agglomerates. Regarding pozzolanic reactivity, the ~90% amorphized samples clearly demonstrated a higher increase in compressive strength than the 100% amorphized ones due to their higher pozzolanic reactivity. The highest compressive strength was achieved by the kaolin ground for 30 mins, achieving ~90% amorphization, which was almost the same compressive strength as that obtained with the thermally activated kaolin at 700 °C for 1 h. All of the trass-containing mixtures achieved higher compressive strengths than the reference, indicating that the mixtures can improve the compressive strength of the binder due to their pozzolanic reactivity. Finally, compared to the high-grade Zettlitz kaolin and abrasive diatomaceous earth, it was observed that SCMs with similarly good levels of pozzolanic reactivity can be efficiently produced from local medium-grade kaolin and trass resulting in ~90% amorphization.

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