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Effect of duration and type of grinding on the particle size distribution and microstructure of natural pumice with low pozzolanic reactivity

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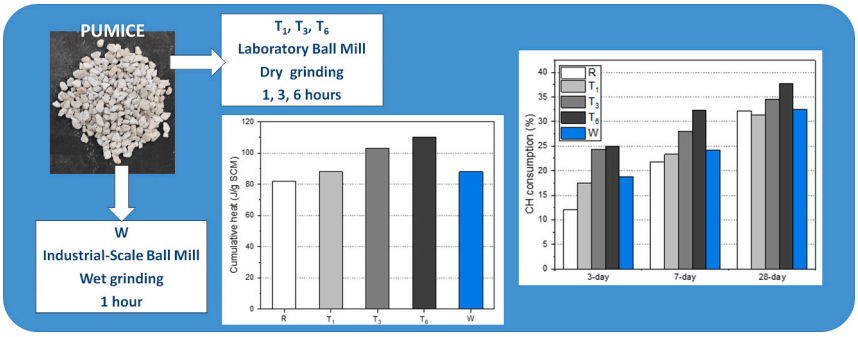
HIGHLIGHTS

- The pumice was ground in dry and wet conditions for comparison.
- XRD results showed the change in crystal structure during grinding.
- Dry-grinding led to higher lime consumption and heat evolution.
- Non-pozzolanic pumice could be activated via right grinding method and time.

GRAPHICAL ABSTRACT



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ABSTRACT

Pumice, with low pozzolanic reactivity, was ground for 1, 3, and 6 h with a laboratory ball mill in dry conditions and it was ground for 1 h in wet conditions via an industrial-scale ball mill. Based on derivative particle distribution, grinding for longer periods led to the disappearance of bimodal distribution and the development of unimodal distribution. Furthermore, the phase characterization, assessed through XRD, demonstrated appreciable changes in intensities of the peaks of quartz and dachiardite. The extension of grinding time resulted in a significant uptake at the early-age lime consumption and evolution of hydration heat. According to SEM images, the number of particles between 10 and 20 μm was less in the powder ground for 3 and 6 h. Moreover, it was found that the prismatic shapes of raw pumice tended to transform to spheroid shapes after prolonged grinding, and the smooth surfaces of pumice particles became more rugged.

1. Introduction

Pumice, a volcanic tuff, is a pozzolanic material that was ejected from volcanic eruptions and cooled rapidly which caused its amorphous phases and low density [1]. Due to its high volume of voids, the pumice can be utilized in concrete as a lightweight aggregate [2]. In nature, the appearance of pumice ranges from white to black, with gold, grey, and brown [3]. The coloring of pumice depends on its chemical composition;

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generally, it consists of high amounts of SiO_2, medium amounts of Al_2O_3 and Na_2O, and low amounts of CaO, Fe_2O_3, TiO_2, and SO_3 [4–7]. It is similar, in terms of mineralogical composition, to volcanic ashes; whereas, the former possesses much coarser particles compared to the latter [8]. The popular usage of volcanic ashes in concrete production is its incorporation to concrete mix as a mineral admixture, differently the pumice. Since they have similar chemical composition, the only constraint restricting pumice from being used as a natural supplementary cementitious material (SCM) is its lower pozzolanic reactivity [9–11].

Nowadays, natural pozzolans are integrated into cement as replacements for ordinary Portland cement (OPC) to reduce the carbon emissions due to the grinding, calcining [12], and transportation of OPC [13]. Some pozzolanic materials are found in powder form in nature, such as volcanic ash, which doesn't require extensive grinding or calcining to make them usable in concrete [14]. In case of their local presence in high volumes, extra energy consumption is not involved in their transportation [15]. According to the Mineral Commodity Summaries 2023 published by U.S. Geological Survey, in 2022, approximately 15 million tons of pumice was produced globally. It is reported that the reserve of pumice in the US is >1 billion tons. It is also abundant in European countries such as Turkiye, Italy, Greece, France, and Spain; South America; Asia; and African countries such as Uganda. However, utilization of pumice as a SCM in concrete is limited by its low pozzolanic reactivity. Improving the pozzolanicity of pumice, without any thermal process, may result in its increased usage in cement; thereby lowering the CO_2 emission and energy expenditure due to the production and transportation of OPC, and abating the depletion of limestone reserves used for cement production.

Pumice was studied as a cement additive where hardened-state, fresh-state, and durability properties were analyzed [16,17]. Although the pumice-containing mix enhanced the fresh-state properties and lowered the permeability with respect to the reference mix, the compressive strength reduced as the pumice content increased. Since the pumice has favorable attributes (namely; low density [18], thermal resistance [19,20] and high impermeability [21]), its usage in concrete as a lightweight aggregate is quite attractive and ample research has been conducted on its effects. The total substitution of coarse aggregate with pumice led to a decrease of 27% in slump value [17]. In the same study, the replacement of fine aggregate with pumice resulted in a 17% decrease in slump value. Kashyap and Sasikala [22] reported that a 40% replacement of natural coarse aggregate with pumice led to decrease in the 28th day compressive strength from approximately 37 MPa to 30 MPa. Similarly, Degirmenci and Yilmaz [23] reported that in mortars a 100 % replacement of natural fine aggregate with pumice drastically lowered the mechanical strength in all ages. In another study [24], coarse aggregate and fine aggregate, independently and together, were replaced with lightweight aggregate (pumice) and 28th day compressive strength decreased by 20% and 45%, respectively. Sancak et al. [25] investigated the fluctuation of compressive strength under 20, 100, 400, 800, and 1000 °C for normal-weight concrete and lightweight concrete including pumice instead of fine and coarse aggregate. Up to 400 °C, the degradation of strength in both lightweight and normal concrete was similar. However, at 800 °C, the reduction in strength for the lightweight concrete is significantly lower compared to that for normal-weight concrete.

The pozzolanic activity is defined as the reaction between lime and pozzolana, which produces calcium silicate hydrate gel [26]. There are three main categories of methods that are used in determining pozzolanic activity: mechanical methods (e.g., strength activity index (SAI)), physiochemical methods (e.g., XRD and XRF analyses), and chemical methods (e.g., lime-consumption) [27]. The pozzolans such as fly ash (FA) [28], silica fume (SF) [29], metakaolin (MK) [30], rice husk ash (RHA) [26], ground granulated blast furnace slag (GGBFS) [31], demonstrate considerable pozzolanic activity. In the study conducted by Karatas et al. [32], 30% replacement of OPC with ground pumice powder presented SAI of 94% and 97% on the 28th and 90th day, respectively. Even after extensive curing, the mixes containing pumice at a considerable amount didn't exceed the control mix in terms of compressive strength. Moreover, in the study of Kabay et al. [11], the pozzolanic effect of fly ash and pumice powder was investigated. It was found that a 10% substitution of ordinary cement with pumice powder and FA led to a 5% decrease and a 5% increase in the 28th day-compressive strength, respectively. Kasaniya et al. [33], utilized a strength activity index, modified lime-reactivity test, and electrical resistivity in determining the pozzolanicity of FA, SF, MK, perlite, lassenite, and a few types of pumice, and categorized the examined materials according to their pozzolanicity. While FA, SF, and MK were grouped as very high reactivity and high reactivity pozzolans, pumice was classified as very low reactivity and low reactivity pozzolan, Therefore, based on inspected literature, natural pumice possesses subpar pozzolanic activity and doesn't promote the short-term or long-term strength of concrete on an appreciable scale.

Grinding kinetics of materials were studied by many researchers to explain their comminution [34–38]. The increase in the fineness of powdered materials including waste materials, such as waste glass, incorporated into concrete usually enhances the compressive strength and durability [39–41]. Moreover, the longer the grinding duration is, the finer the waste material becomes. Nonetheless, the energy consumption and cost increase exponentially after grinding for a certain period. This threshold can vary based on the type of material. Moreover, if a fine powder is subjected to further grinding, the phenomenon called "particle agglomeration", which can lead to increase the particle size and affect the performance of concrete negatively, occurs [42]. Therefore, it is essential to reveal the impact of grinding time on particle size distribution, fineness, equivalent particle size as well as the activity index of powder material [34].

Natural pumice is a pyroclastic rock that doesn't demonstrate considerable pozzolanic activity, therefore it is not preferred to use as a mineral admixture in concrete. In this investigation, to fill the gap in literature, the improvement of the pozzolanicity of pumice by decreasing its size and correspondingly changing its morphology was aimed. Therefore, the parameters representing grinding type (wet and dry grinding) and grinding durations affecting the pozzolanic activity were considered. The particle size distribution (PSD) and grinding kinetics, which is used as a gauge to compare the increase in the pozzolanicity with the decrease in particle size was experimentally measured and calculated, later fitted with a mathematical model. Additionally, the mineralogical composition was determined with XRD and the pozzolanic activity was measured via lime-consumption. The hydration heat evolution curves were obtained as well. The microstructure and morphology by SEM analyses of ground pumice, which have not been systematically established before, were also investigated.

2. Materials and methods

2.1. Material characterizations

Pumice used in this study was obtained from Ankara, Turkiye. The image of the pumice is given in Fig. 1. Its particle size distribution before being subjected to any grinding, but after crushing with a jaw crusher was measured by a particle size analyzer. The oxide composition of pumice and lime used in this study was determined via X-ray fluorescence (XRF) analysis is given in Table 1. Additionally, to gain insight into the morphological features and crystalline phases of pumice, scanning electron microscopy (SEM) and X-ray diffractometry were employed.

2.2. Methods

One of the major challenges in determining pozzolanic activity of a siliceous material is establishing a correlation between the results of

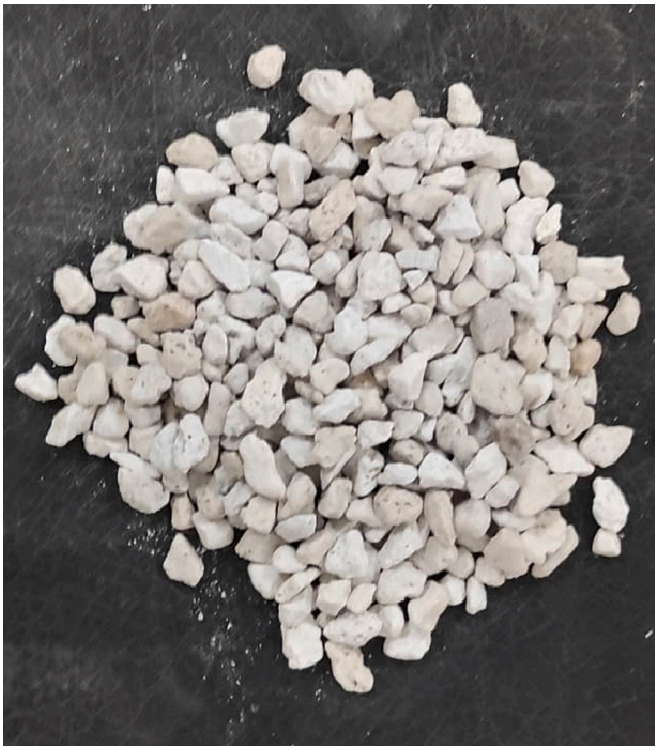


Fig. 1. Natural pumice before being fed to jaw crusher.

Table 1
Chemical composition.

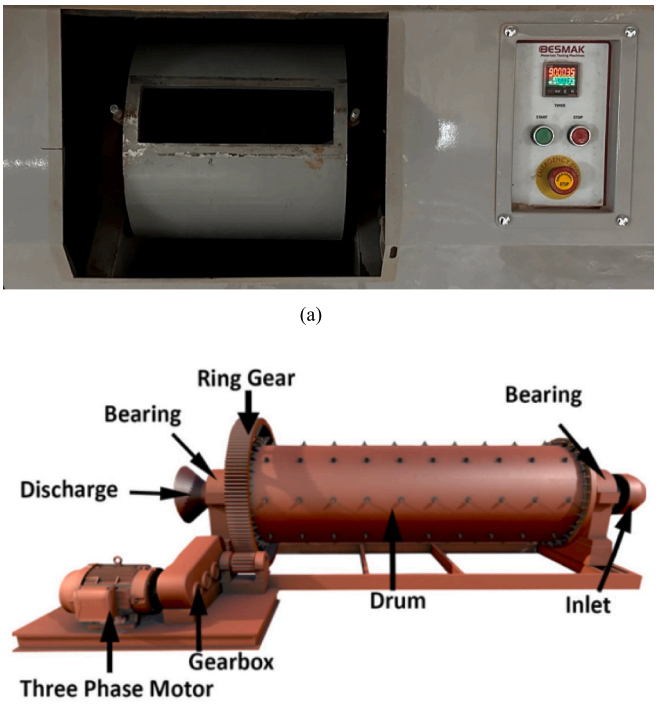
Compounds (%, by mass)	Pumice	Lime
SiO_2	70.90	0.11
Al_2O_3	11.39	0.17
Fe_2O_3	2.55	0.17
CaO	2.16	70.49
MgO	0.84	1.01
SO_3	0.08	3.98
Na_2O	2.49	0.26
K_2O	4.16	0.35
Loss on ignition	2.16	23.34

varying methods [43]. There is a wealth of research in the literature that reports the affinity of the results of mechanical methods with those of chemical methods [44]. However, a strong tie between chemical and physio-chemical methods is yet to be found. The initial step in assessing the change in pozzolanicity of a material is determining its particle size distribution (PSD). Therefore, the PSD of pumice powder before and after grinding was measured for the range of 0.01 to 1000 μm by a particle size analyzer that operates based on laser light scattering technology. Similarly, the XRD analysis was performed via Rigaku Ultima-IV to physio-chemically compare pozzolanic activity. The data were gathered at angles ranging from 10 to 70 2-theta. Furthermore, to observe the effect of grinding on the morphology of the pumice particles, SEM analysis was conducted with a Tescan GAIA3 + Oxford XMax 150 EDS. The same labels were used for both raw ground powders and pastes; such that R represents unground powder or the paste produced with it and W represents wet ground sample. T1, T3, and T6 are used to label the samples that were dry ground for 1, 3, and 6 h, respectively. For the chemical approach of measuring the pozzolanic reactivity of pumice, calcium hydroxide (lime) consumption in lime-pumice pastes was found. The pastes consisted of 50% lime, 50% pumice powder, by weight, and had 0.55 water-to-solid ratio. These ratios are based on a previous study conducted by Uzal et al. [45], where the pozzolanic reactivity of relatively unreactive lime-stone powder was investigated. Their heat of hydration evolution was obtained on their pastes with an isothermal calorimeter at a constant temperature of 50 °C for 7 days according to ASTM C1679 [46].

The pumice was ground for 1, 3, and 6 h in a laboratory ball mill after being crushed with a conventional jaw crusher. The dry mill had a chamber with internal dimensions (D x H) of 30.5 cm × 30.5 cm that amounts to a volume of around 22 l (Fig. 2a.). The rotational speed of the ball mill was fixed at 70 rpm. A continuous ball charge was used, to simulate the most effective grinding per time [47]. The charge of balls contained 50, 25, and 540 steel spherical balls with a diameter of 38, 30, and 20 mm, respectively. In total 615 balls equaled a mass of approximately 27 kg which corresponded to a ball-filling-ratio (BFR) of 28.6% and 3005 g of pumice was introduced to the chamber that corresponded to a material-filling-ratio (MFR) of 47%. To examine the impact of wet grinding, an industrial-size mill was used for a duration of 1 h at 20 rpm (Fig. 2b.). The mill had a drum with a diameter of 2 m and a length of 6 m. Balls of various sizes were placed in the mill, the largest of which had a diameter of 15 cm. The wet grinding of pumice is as follows; the raw material is transported via a conveyor belt from the bunker located outside the production building into the mill. The belt weighs the pumice and feeds the material and water simultaneously into the ball mill. When the sludge is ground for an assigned duration, sludge is sent to the slurry tank. Since pumice is not a cementitious material, there is no risk of hydration during the wet grinding process.

The primary metric used to evaluate the performance of a grinding process is the particle size distribution (PSD) [48]. The PSD models such as Rosin-Rammler-Bennette (RRB) [49] and Swrebec [50] are typically employed to predict and analyze the size distribution of particles in a granular material. Since there is scant of knowledge on whether pumice conforms to the above-mentioned models; in this study, the compatibility of the RRB model in predicting the PSD of natural pumice was evaluated. To this end, a brief overview of the parameters and evolution of the RRB model is explained below.

Sieve residue (R) is a measure of the size distribution of a particulate material and is defined as the percentage of the material that remains on a sieve after it is subjected to sieving. The value of R drops continuously



(a)

(b)

Fig. 2. Dry grinding mill (a) vs. industrial scale dry grinding mill (b).

as the grinding time increases. Divas-Aliaviden equation shows the relationship of the reduction rate of R with a fixed particle size (−dR/dt) with the sieve residue (R).

$$\frac{dR}{dt} = -K_t R \tag{1}$$

The grinding rate constant, denoted as K_t, is a key component in the equation that describes the grinding process. The equation, which has been modified to better explain the process, is presented below,

$$R = R_0 e^{-(K_t)^M} \tag{2}$$

The kinetic equation for the grinding process requires two parameters: R_0, the initial sieve residue, and M, the time index. The RRB distribution model, which has been utilized by many researchers [42,51,52] to determine the optimal grinding process, is also expressed in the following equation,

$$fR = exp\left[-\left(\frac{D}{D_e}\right)^n\right] \tag{3}$$

The equation for the RRB distribution model includes R, the cumulative sieve residue of powder; D, the particle size; and D_e, the characteristic particle size (which represents the particle size at which R is equal to 36.79 %). The model also includes the distribution index, n.

3. Results and discussions

3.1. Particle size distribution and grinding kinetics of pumice

Cumulative particle distribution and differential particle size distribution plots are presented in Fig. 3. And Fig. 4., respectively. These illustrate that grinding for longer periods produced much finer particles as a whole. The most apparent result from the cumulative PSD plot is that wet grinding resulted in the coarsest particles after the unground powder. Additionally, as the grinding time was increased from 1 h to 6 h, a continuous improvement in fineness (maximum particle sizes in T1 and T6 are 24 and 34 μm, respectively) was observed. In Fig. 4., two peaks in the curves representing different grinding times/methods can be seen, which is mainly due to the friction comminution [53]. As the duration of grinding increased, the peak at 2 μm grew, while the peak at 12 μm shrunk. Furthermore, the equivalent particle sizes (D_{10}, D_{25}, D_{50}, D_{75}, and D_{90}) of the pumice powder are reported in Table 2. As the grinding time was increased from 1 to 6 h, the decline in all of the equivalent particle sizes was observed. The decrease in D_{10} after grinding is not as significant as it is in the other equivalent particle sizes, which supports the claim that approximately 10% of particles remain around the size of 0.5–1.5 μm even after extensive grinding. After grinding for 6 h, D_{90} dropped to 13.87 μm, which is even significantly finer than that of cement [54].

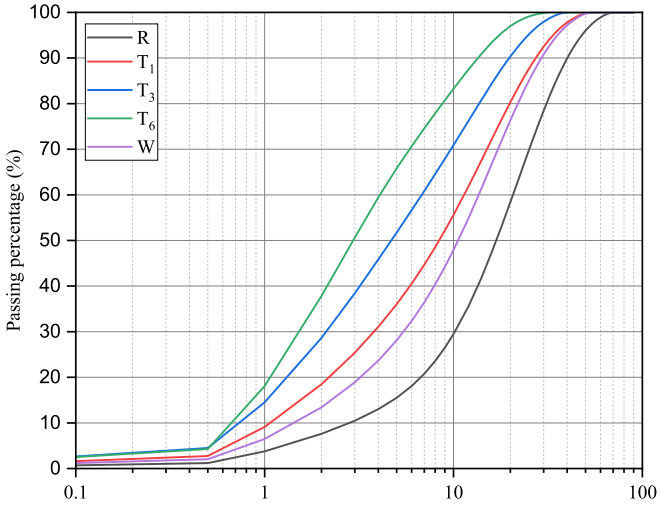


Fig. 3. Cumulative particle size distribution.

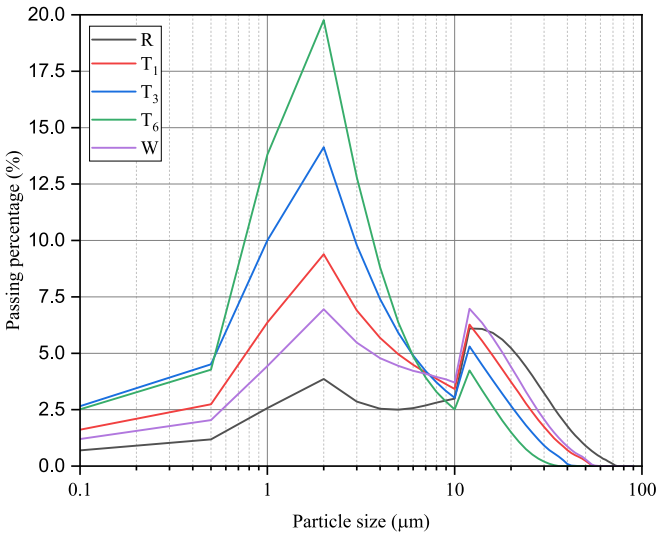


Fig. 4. Derivative particle size distribution.

Table 2
Equivalent size of pumice powder.

Series	D_{10} (μm)	D_{25} (μm)	D_{50} (μm)	D_{75} (μm)	D_{90} (μm)
R	2.86	8.49	16.52	27.91	40.02
T_1	1.10	2.95	8.24	17.63	27.76
T_3	0.69	1.74	4.83	11.81	19.92
T_6	0.55	1.34	2.96	7.04	13.87
W	1.49	4.22	10.44	19.64	29.74

Furthermore, to interpret the particle distribution of pumice, Eq. (1) and Eq. (2) of the RRB model were employed to correspond to measured sizes from the cumulative PSD. As can be seen in Table 2., the distribution index, n, had a value of around 1, which elucidated a tight particle size distribution. In other words, n value displayed the compactness of the PSD of any particle system. The characteristic particle size of pumice put forth by the RBB model was comparable to the experimental characteristic particle size, which is a gauge for measuring compliance

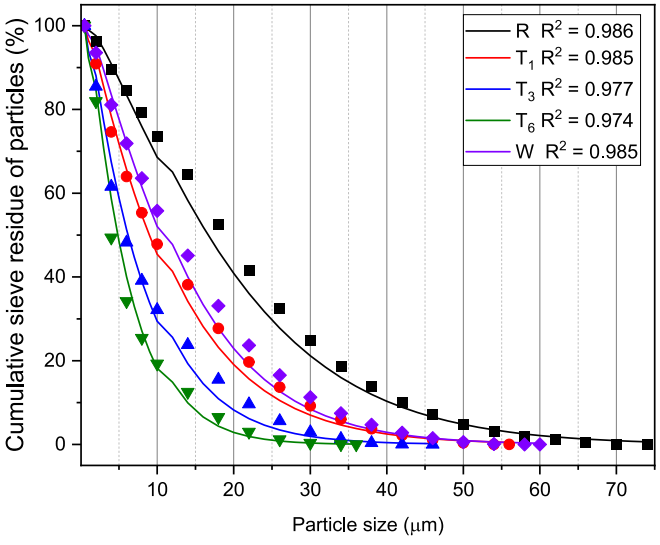


Fig. 5. Comparison of fitting (curves) and experimental (symbols) cumulative particle size distribution of pumice powder.

of the RRB model with cumulative PSD. In Fig. 5, the PSD of the RBB distribution model (shown by solid lines) was matched with that of experimental data (shown by symbols), where the coefficient of determination (R^2) was also demonstrated. As can be seen, the PSD curves of the RBB model correlated well with the experimentally measured values. The RRB model distribution corresponded well to the experimentally gathered data, which showed that the cumulative PSD confirms to RRB model. Based on Table 3, it is obvious that the characteristic particle size of pumice estimated via the RRB distribution model was close to the experimentally measured characteristic particle size.

3.2. X-ray diffraction

XRD spectra of powder pumice are illustrated in Fig. 6. The peaks of quartz, calcite, and anorthite were observed in all of the samples. The peaks at 21.1, 26.6, 27.2, 37, 39.6, 50.1, 60, and 68 2-theta are representative of quartz [55]. The major peaks at 27.9 2-theta represent dachiardite which is zeolite [6]. The peaks at 30.5, 31.5, and 23.1, 29.5, 35.7, 48.6 2-theta correspond to anorthite and calcite, respectively [56,57]. Grinding, however, seems to have lowered some of the peaks. The dachiardite peak at 27.9 2-theta declined as the grinding time was increased, while the highest peaks were observed in R and *W. major* and minor peaks representing quartz diminished as the duration of grinding was extended. In other studies, a similar deduction was made about the relationship of intensity of crystalline quartz peaks and grinding time [58] All these changes in the XRD spectra point to the loss of crystallinity [58–60], hence increasing reactivity [61]. It shows an increase in degree of amorphization. The fused silica, which is a reactive phase, is formed because of disintegration of quartz. It is suspected that considerable amounts of this reactive silica were formed as a result of grinding. The literature confirms that an increase in degree of amorphization, the amount of reactive phase in a material, is affected by the duration of grinding [58]. The most palpable change was recorded in the peaks related to dachiardite, which is a crystalline mineral [62]. It is hypothesized that grinding, especially dry grinding, broke down the well-ordered structure of this mineral in pumice. The variation in the intensity of peak belonging to dachiardite can be correlated in accordance with the results of physical and physio-chemical tests (e.g., isothermal calorimetry and lime-consumption). Therefore, wet and dry grinding result in the disintegration of the crystalline structure of pumice and formation of the amorphous silica phase. However, according to the XRD spectra, W and T_1 displayed limited weakening in the structure of crystalline minerals, whereas T_3 and T_6 showed a tremendous change.

3.3. Lime-consumption

The pozzolanic reactivity of the pumice powder was estimated by calcium hydroxide (lime) depletion in the lime-pumice powder pastes (Fig. 7.). The lime consumption increased gradually as the age of pastes increased. Even for the 3rd day, T_6 showed 25% consumption and on the 28th day, this percentage increased to over 37%. Equivalently, the lime consumption increased from the 3rd to the 28th day in all the mixes. In parallel with PSD, extended grinding time was beneficial to the pozzolanic activity. On the contrary, the W paste bore extremely resembling

Table 3
Distribution index (n) and fitted (D_e) and measured (D) characteristic particle sizes.

Series	n	D_e (μm)	D (μm)
R	1.2466	19.68043	22
T_1	1.0699	11.22949	12.49
T_3	1.031	7.40412	7.64
T_6	1.0577	5.43738	4.59
W	1.1733	12.9408	14.35

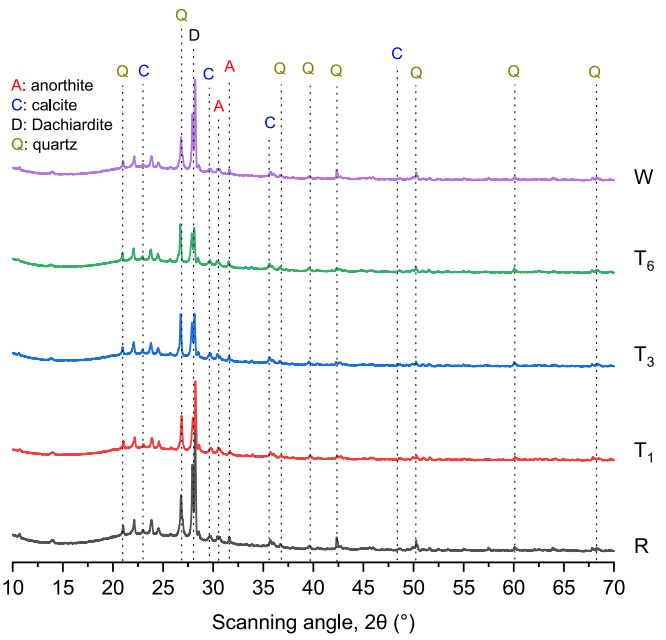


Fig. 6. X-ray diffraction spectra of ground and unground pumice.

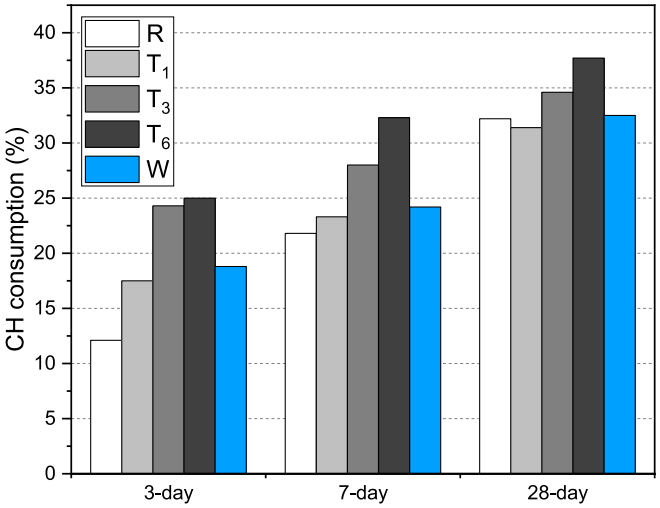


Fig. 7. Lime-consumption of the pumice on 3rd, 7th, and 28th day.

results to the R paste in terms of lime consumption on the 28th day. From the data, it seems that in the early ages, extended grinding boosted lime consumption drastically (T_6 bound the lime (25%) two times higher than T_1 (12.1%) did on the 3rd day). According to Uzal et al. [58], a high specific surface area (SSA) and a highly amorphous structure enhance pozzolanic reactivity. For the early age, a strong connection between the amorphous phase (i.e., the lack of crystalline phase) discerned with XRD and the percentage of lime-consumption is found. Thus, dry grinding of pumice, especially for up to 3 h, improved the pozzolanic reactivity significantly; however, grinding beyond this yielded negligible gains. On the other hand, in later ages, the range between lime-consumption of all the pastes decreased (31.4–37.7% on the 28th day). Based on the results of the lime-consumption test, it is apparent that in the long run, i.e., 28th day, the effect of grinding significantly depreciated, and the aggressive lime uptake detected on the 3rd and 7th days cannot be repeated. Other methods estimating lime-consumption, such as Frattini, which matches well with the results of mechanical tests, however, these tests require up to 15 days [44]. In conclusion, it is safe to say that the utilization of this

rapid method in estimating the pozzolanicity of pumice, probably even other types of siliceous materials, is conducive and correlates well with the findings of other pozzolanic activity indicators.

3.4. Isothermal calorimetry

In order to determine the pozzolanic reactivity of pumice powder with another method, total heat discharge at the end of 7 days of age was measured and shown in Fig. 8. The magnitude of heat is presented in joules per gram of paste. Isothermal calorimetry is used to successfully evaluate the pozzolanic reactivity of mineral admixtures [45,63]. According to this method, pozzolanicity and potential of reaction are commensurate with the cumulative heat evolution. In a study done by Feng et al. [64], the effects of ground copper slag on blended cement were investigated. Sun et al. [65] measured the heat flow rate and total heat release of binary systems of FA; ground or unground, and cement, where ground FA-containing mix discharged more heat compared to the unground mixture and pure cement specimens. According to their report, it was elucidated that finer particles produced as a consequence of grinding facilitated more nucleation sites for the cement grains to hydrate, thus increasing the heat of hydration. However, due to the absence of cement in the specimens of this study, hence nullifying the influence of nucleation, it is probable that other factors are effective to increase heat discharge. The inherent inclination of finely ground pumice in breaking CH bonds and forming C-S-H gel could have raised the heat of hydration. As can be seen from Fig. 8., the T_6 paste demonstrated the highest heat release of 110 J/g at the end of the 7th day. According to Kumar and Kumar [66], the reactivity of FA increases significantly when the median particle size is <5–7 μm. Similarly, based on the PSD data, both T_3 and T_6 had a lower D_{50} than 5 μm. Therefore, the threshold of a worthwhile increase in the reactivity of pumice with respect to its particle size is not far off that of FA. Akin to previous analyses, wet grinding resulted in a trivial raise in pozzolanicity.

3.5. Scanning electron microscopy

SEM images of pumice powder ground with different techniques and for different durations are given in Fig. 9. and Fig. 10. Similar to PSD data, Fig. 9. clearly shows the size difference of pumice. In Fig. 9a., particles mostly sized approximately 100 μm are observed, whereas grinding even for 1 h of dry grinding has reduced the amount of these larger particles significantly (Fig. 9b.). Wet grinding, on the other hand, seems to have less effect on the fineness of pumice than dry grinding. Amongst the ground samples, large particles sized >100 μm are the most frequent in W feed, which are significantly rarer in the dry ground samples (Fig. 9e.). Grinding for 3 h eliminated almost all particles sized >100 μm (Fig. 9d.). Additionally, based on Fig. 9c. and Fig. 9d., the discernible difference in particle sizes of T_3 and T_6 couldn't be noticed, however, tiny roundish spots were observed, uncovering the nature of which required closer inspection.

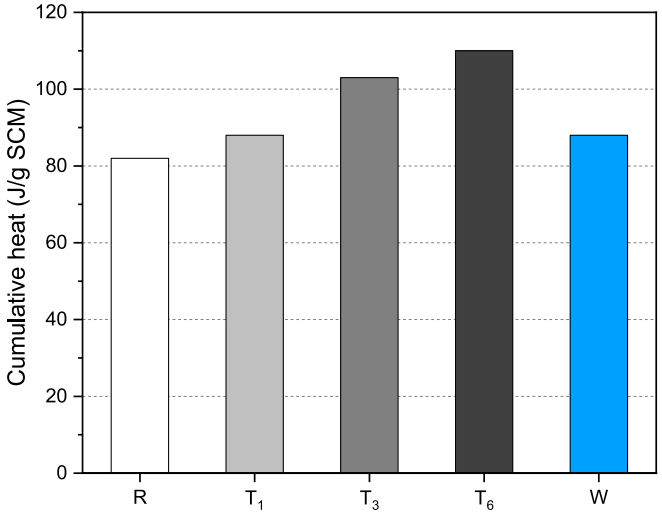


Fig. 8. 7-day cumulative heat of hydration.

To make a detailed inspection of the morphology of the particles, images of powders are from closer were also analyzed (Fig. 10.). Although a discontinuous surface and porous structure is pervasive in pumice in nature [67], in Fig. 10a., R shows a smooth and regular-shaped pumice particle which is an indicator of well-ordered crystalline particle [68]. In Fig. 10b. and Fig. 10e., smaller, irregularly shaped, and uneven-surfaced particles are the outcome of grinding for 1 h, similarly. A ragged surface caused by grinding results in increases surface area, thus increasing the reactivity of pozzolan [69]. Additionally, the particles sized between 10 and 30 μm are prevalent. The fact that the surfaces of the particles tended to become more abrasive as the grinding duration was increased, is in accordance with the literature [67,70]. SEM images of T_3 and T_6 show that dry grinding for >1 h led to a significant increase in the non-uniformity of the surface of pumice particles. According to Yao et al. [69], abrasive edges and sides of particles increase the reactivity of cementitious materials like in this case and this is in parallel with the results of lime-consumption and heat of hydration. It is worth mentioning that SEM images of T_3 and much more so of T_6 exhibit spheroid particles (Fig. 10c and Fig. 10d.) that are much smaller than cubic particles detected in R. It is postulated that the spheroid exterior is the ultimate shape of siliceous materials, especially pumice, acquires after a prolonged grinding. Therefore, most of the pumice particles have approached their final particle shape in T_6. Moreover, in both samples ground for 3 and 6 h, larger particles than 10 μm can be seen, elucidating that grinding pumice for longer than 3 h did not reduce the size of some percentage of larger particles. Furthermore, in T_6, the particles smaller than 2 μm exhibit some agglomeration (Fig. 10d.) which hinders further splintering of the particles, as a matter of fact, it causes the formation of larger particles [68]. According to previous studies, this agglomeration effect is attributable to high particle surface energy [68,70]. All in all, dry grinding with the assistance of a ball mill for 3 h seems more promising in enhancing morphology or fineness than that for 6 h.

4. Conclusion & recommendations

To briefly summarize, this paper investigated the effects of various methods and durations of grinding on pozzolanic reactivity of pumice utilizing the chemical and physio-chemical methods. The PSD plots of pumice shed insight on the effect of grinding duration, as well as the applicability of RRB distribution in estimating the cumulative PSD curve and characteristic particle size. Grinding up to 6 h reduced the size of particles significantly in the D_{25}-D_{90} range. XRD peaks revealed that peaks of quartz and dachiardite diminished as the grinding time was increased, which indicates the decomposition of their crystalline structure. Lime consumption and the amount of dissipated heat increased as the grinding time was extended, thus correlating the pozzolanic activity of pumice determined via chemical and physio-chemical methods. The increased pozzolanicity resulting via grinding is justified by not only the linear decrease in particle size but the divergence of morphology, also. SEM images reveal that from a morphological and microstructural point of view, the grinding changed the initially flat surface of pumice particles as well reduced the size of pumice particles drastically. Wet grinding, in general, was not as successful as dry grinding for 1 h, according to the results of the tests and analyses performed. It is believed that the slower rotational speed, unbalanced ball charge, and the industrial size of the wet mill inhibited the effective comminution. In future investigations, it is strongly encouraged that a relationship between chemical and mechanical method (e.g., compressive strength) is established, and the effect of more grinding characteristics such as ball-

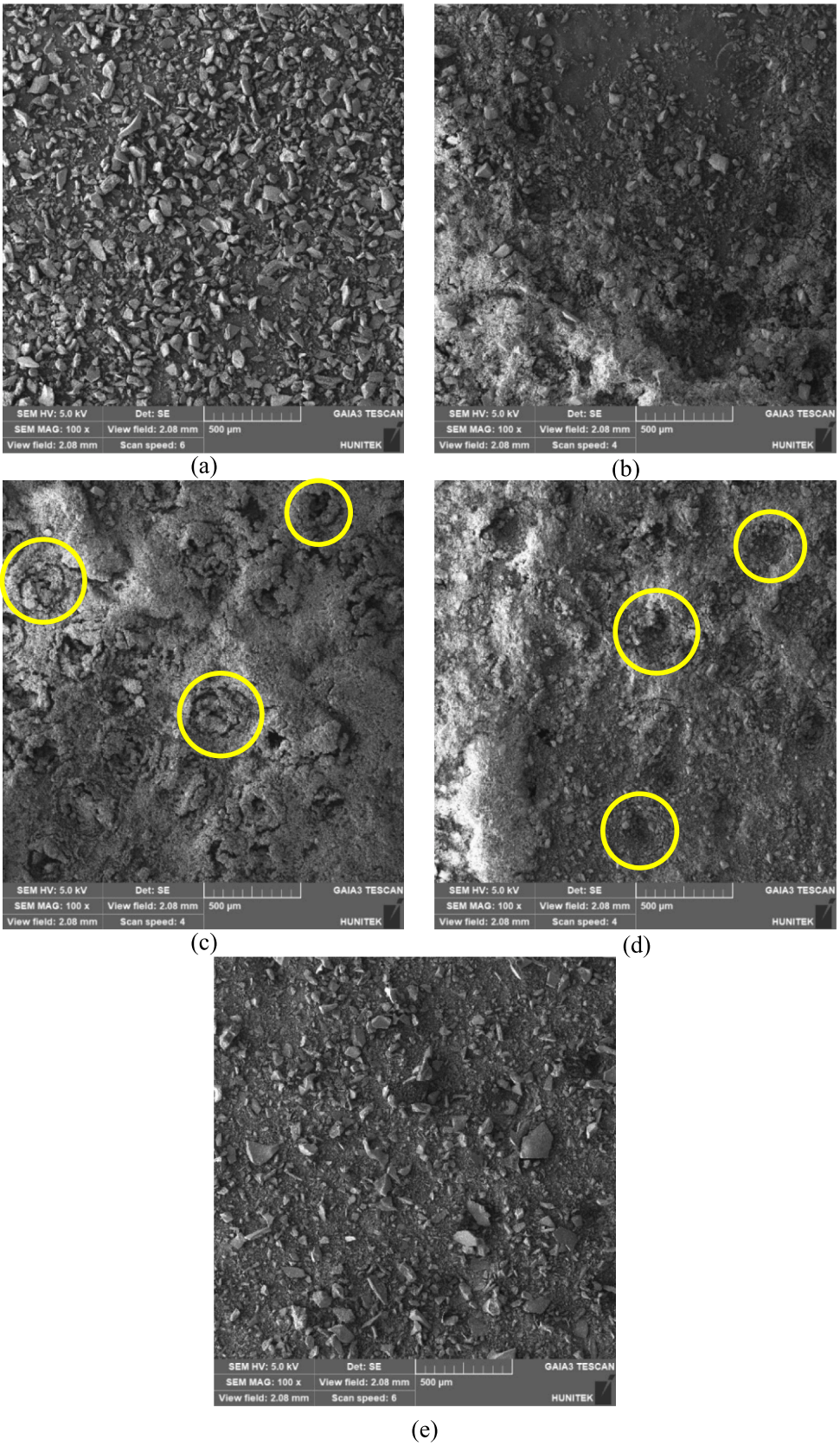


Fig. 9. 100× scanning electron microscopic images depicting the general particle size of R (a), T_1 (b), T_3 (c), T_6 (d), and W (e) (yellow rings mark some of the observed round spots). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

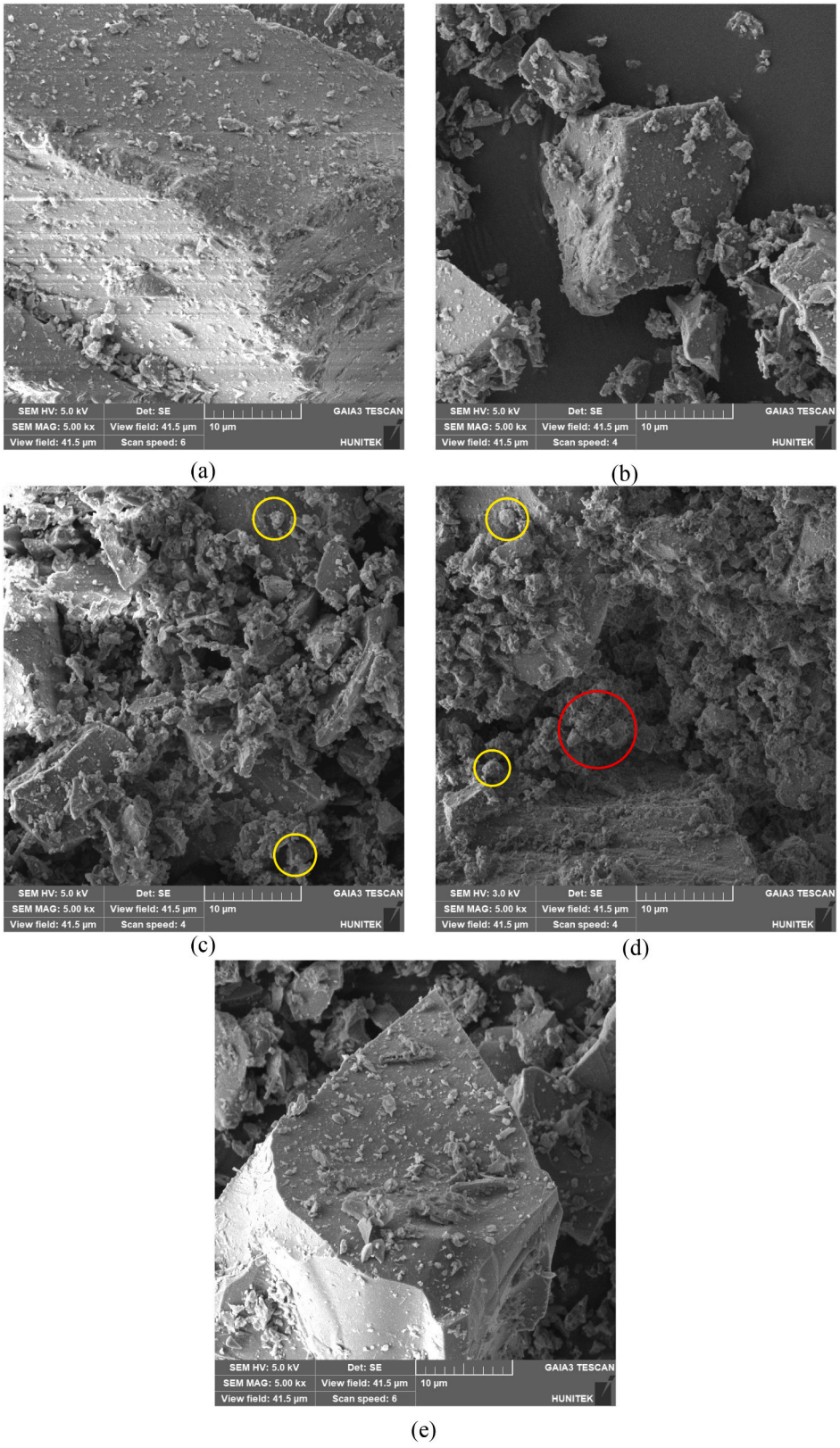


Fig. 10. 5000× scanning electron microscopic images of R (a), T_1 (b), T_3 (c), T_6 (d), and W (e) (yellow and red rings indicates spheroid particles and agglomeration, respectively). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

filling ratio, material-filling ratio, and different ball charges on PSD and pozzolanic activity of pumice is examined.

CRediT authorship contribution statement

Khalilullah Taj: Writing – original draft, Formal analysis, Investigation, Visualization. **Hüseyin İlcan:** Methodology, Investigation. **Eray Teksin:** Methodology. **Gizem Argın:** Methodology. **Mehmet Kemal Ardoğa:** Writing – review & editing. **Burak Uzal:** Conceptualization, Project administration. **Mustafa Şahmaran:** Conceptualization, Methodology, Resources, Supervision, Funding acquisition, Project administration.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Huseyin Ilcan reports financial support was provided by The Scientific and Technological Research Council of Türkiye. Mustafa Sahmaran reports financial support was provided by The Scientific and Technological Research Council of Türkiye. Mustafa Sahmaran reports a relationship with The Scientific and Technological Research Council of Türkiye that includes: consulting or advisory.

Data availability

Data will be made available on request.

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