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Repeated thermal shock processes for fragmenting zeolite particles and the possible existence of a critical size

Devi Tiyan Cahyaningrum [a], Handika Dany Rahmayanti [a], Edy Wibowo [b], Khairurrijal [a], Mikrajuddin Abdullah [a,c,*]

[a] Department of Physics, Bandung Institute of Technology, Jalan Ganesa 10, Bandung 40132, Indonesia
[b] Department of Engineering Physics, Telkom University, Jalan Telekomunikasi, Bandung 40257, Indonesia
[c] MIBE S&T Institute, Jalan Sembrani 19, Bandung 40295, Indonesia

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ABSTRACT

In our previous work, we observed that a thermal shock process could fragment zeolite particles to approximately one-fourth of the initial size. Here, we further investigate this finding by applying repeated thermal shocks to the zeolite particles and observed the effect on the resulting particle size. We determined that repeated thermal shocks progressively reduce the zeolite particle size, though the resultant size was not always one-fourth of the previous size. We claim that the thermal shock generated a high-temperature gradient, and the zeolite structure converted into a quartz structure at the point at which the temperature gradient reached a maximum, resulting in a silicon-rich surface of the produced particles (higher Si/Al ratio). We also developed a model to predict the possible existence of a critical radius, below which the size will not be reduced by thermal shock. The critical radius decreases when the shock temperature increases. The prediction is qualitatively consistent with the observed final particle size after applying different shock temperatures.

1. Introduction

Thermal shock is a quick change in temperature by several hundred degrees Celsius. The thermal shock process is usually used for testing the resistance of a material [1–4]. However, thermal shocks may create unexpected results [5]. For example, in the production of ceramics, thermal shock failure leads to the rejection of more than one-third of ceramic components [6].

When examining zeolite activation, we accidentally observed that the thermal shock process was able to fragment zeolite particles into smaller ones, the largest fraction of which has a diameter of approximately one-fourth of the initial diameter [7,8]. A thermal shock step can be added to the zeolite activation process to increase the adsorption and sorbent capacity. In previous work, we only performed one thermal shock process. We also proposed a model to explain why the size of the produced particles were nearly one-fourth of the initial size [7,8]. The challenging question is what happens when the thermal shock process is repeated? When the smaller particles produced by a previous thermal shock are subjected to further thermal shock, does the size further decrease? Does the size continue to decrease when the thermal shock is repeated, or is there any critical size below which the fragmentation will cease?

The objective of this work is to investigate the effect of repeated thermal shocks on the zeolite particle size and to develop a model for predicting the possible existence of a critical size below which the fragmentation will cease.

Recently, Shao et al. investigated the effect of thermal shock on the failure of ceramics [5]. They used ceramic balls with sizes up to 2 mm made of alumina powder with sizes of approximately 0.5 μm. The balls were initially heated at preset temperatures and then dropped by free fall into a water bath. They investigated the fault of cracks on the ceramic surface and the effects of the ceramic size and shock temperature (difference between the preset temperature and bath temperature). Our work is different from the work of Shao et al. [5]. Instead of creating cracks, we used thermal shock to fragment zeolite particles or powder and explored the effects of the initial particle/powder size and the temperature difference on the resulting particles. Therefore, this work complements the results reported by Shao et al. [5].

2. Experiment

We used commercially available natural zeolites with sizes of tens of micrometers. Some particles were used as received and others were ground using a mechanical mortar to produce smaller initial sizes. For first treatment, the mass of sample inside the alumina crucible was 26.000 g. Having completed one step of heat treatment, 1 g of sample was withdrawn for characterization, and the rest was used for next heat treatment. The initial particles were thermally treated using

* Corresponding author at: Department of Physics, Bandung Institute of Technology, Jalan Ganesa 10, Bandung 40132, Indonesia.
E-mail address: din@fi.itb.ac.id (M. Abdullah).

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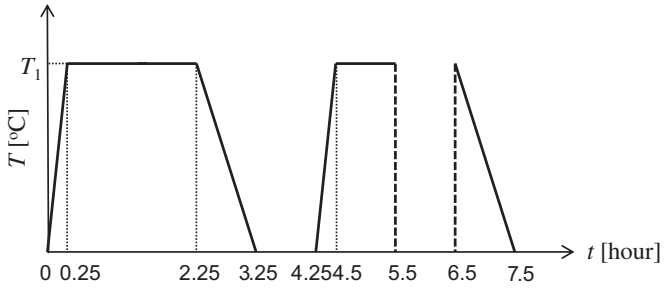

Fig. 1. Heating profiles applied to the samples. The standard heating program is shown by the profile from t = 0 to t = 3.25 h. The additional thermal shock process is shown by the profile from t = 4.25 h to t = 7.5 h [7,8].

different routes: standard heating and pre-standard heating followed by thermal shock. The temperature profile is displayed in Fig. 1 [7,8]. The standard heating profile is shown in the left part of Fig. 1, from t = 0 to t = 3.25 h. The program consisting of pre-standard heating followed by thermal shock is shown by the entire profile in Fig. 1.

We examined different temperature steps using the same standard pre-thermal heating procedure. By temperature step, we mean the difference between the hot air temperature and the initial temperature of the zeolite. Three temperatures steps were investigated: 250 °C, 370 °C, and 410 °C. For the temperature step of 250 °C, we used a standard oven for both small and large initial particle sizes, but for the temperature steps of 370 °C and 410 °C, we fired the samples using a carbide welding fire for a few seconds.

Scanning electron microscope (SEM) equipped with an electron dispersive X-ray (EDX) spectrometer (SEM JEOL JSM-6510LA) was used to image the sample morphology and to analyze the elemental composition of the sample. XRD patterns of the sample were characterized using a Phillips PW1710.

3. Results and discussion

Fig. 2 shows the SEM images of (a) the initial zeolite particles and the zeolite particles after applying thermal shock processes without pre-standard heating: (b) one thermal shock, (c) two thermal shocks, and (d) three thermal shocks. No pretreatment was conducted prior to SEM observations. The temperature step was set at 250 °C. The corresponding log-normal size distributions are shown in Fig. 3. The distribution was determined by measuring the diameters of approximately one hundred particles from the SEM images and then fitting the counting frequency using a log-normal function [7,8]. The diameter of the initial particles was approximately 7.57 ± 0.26 μm. As clearly shown in Fig. 3, the thermal shock process without pre-standard heating only slightly affected the particle size. Based on the size distribution, we estimated the average particle size after one, two and three thermal shock processes to be 3.83 ± 0.13 μm (≈1/1.9), 3.32 ± 0.12 μm (≈1/1.1), and 6.51 ± 0.68 μm (≈1/0.5), respectively. The figures in parentheses represent the ratio of the average resulting particle diameter to the average initial particle diameter. Surprisingly, instead of reducing the particle size, three thermal shock processes produced a larger particle size than that produced after two thermal shock processes. It is likely that agglomeration or some kind unification mechanism occurred during the three thermal shocks.

Clear fragmentation was observed when applying pre-standard heating prior to the thermal shock process. Fig. 4 shows the particle size after applying several thermal shock processes, where all samples have been initially treated with pre-standard heating. The temperature step was slightly different, e.g., 250 °C. We also determined the average particle size by fitting the measured size distributions with a log-normal function. The average initial particle diameter of 5.9 ± 0.12 μm became 2.72 ± 0.06 μm (≈1/2.2), 1.24 ± 0.04 μm (≈1/2.2), and 0.89 ± 0.02 μm (≈1/1.4) after one, two and three thermal shock processes, respectively. It is clear that the repeated thermal shock process was able to progressively fragment zeolite particles when pre-standard heating was applied.

We then examined larger sizes of the initial zeolite particles. We applied pre-standard heating to all particles before applying the thermal shock process. Fig. 5 shows images of the particles (a) prior to thermal shock and after (b) one, (c) two and (d) three thermal shock processes. The average initial particle diameter of 36.87 ± 0.71 μm became 8.39 ± 0.35 μm (≈1/4.4), 2.95 ± 0.05 μm (≈1/2.8), and 1.99 ± 0.03 μm (≈1/1.5) after one, two and three thermal shock

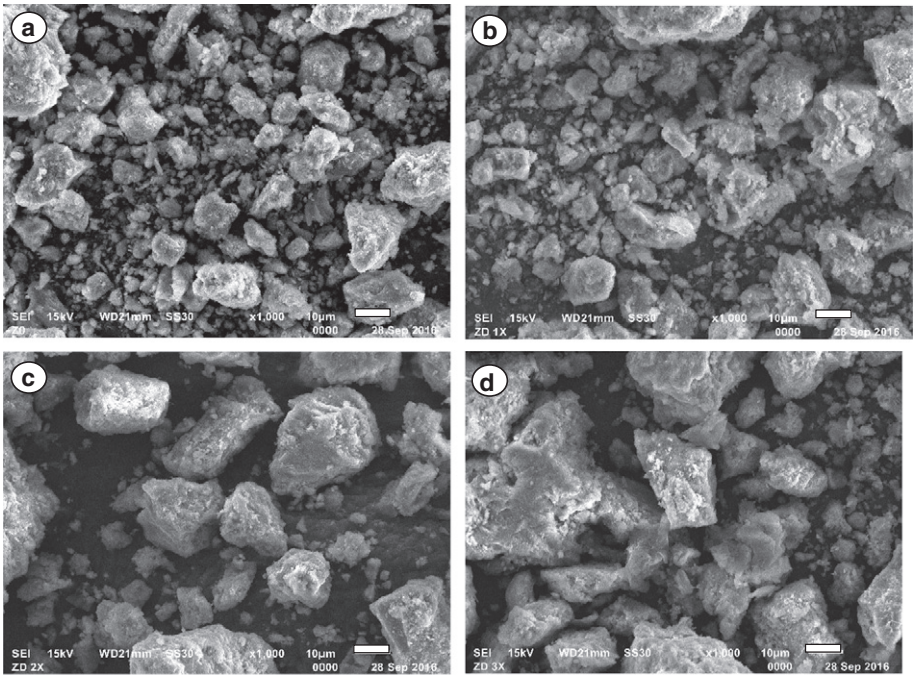

Fig. 2. SEM images of (a) the initial zeolite particles and the zeolite particles after being subjected to thermal shock processes without pre-standard heating: (b) one thermal shock, (c) two thermal shocks, and (d) three thermal shocks. The shock temperature was 250 °C. The bar scale length is 10 μm.