Fabrication of Non-firing Ceramic Utilizing Paper Sludge Ash Treated by Planetary Ball-milling

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Abstract: Non-firing ceramic made of paper sludge ash was successfully fabricated by chemically activated particles with planetary ball-milling. This mechanical treatment was found to have a significant affect on the fabrication of non-fired ceramics. This process is an attractive route for the shaping of waste material into a valuable ceramics without using the conventional sintering step.

Keywords: Non-firing ceramics, Planetary ball-milling, Mechanical treatment

1. INTRODUCTION
Industrial wastes are one of the most serious environmental problems. For example, the paper sludge dumped on land has increased, which demand attention on its environmental effects. The paper sludge is usually landfilled or burned to produce paper sludge ash. The typical paper sludge ash is composed of more minerals such as silica, and aluminum silicate, calcia, etc [1]. Such waste including minerals seems to be able to be utilized as raw material.

On the other hand, it is well known that ceramic powders mixed with potassium silicate glass solutions proceeds in a relatively high rate of polycondensation of silica complexes, resulting into metal-siloxane bridges having $\hat{\text{O}}\hat{\text{O}}\hat{\text{Si}}\hat{\text{O}}\hat{\text{O}}\hat{\text{M}}\hat{\text{O}}\hat{\text{O}}\hat{\text{Si}}\hat{\text{O}}\hat{\text{O}}\hat{\text{O}}\hat{\text{O}}$ as inorganic polymer, where $\hat{\text{M}}$ is a foreign metallic ion. This polymers act as binder between ceramic particles, which responsible for solidification into strong green body at room temperature [2,3]. These ceramic particles are usually compounded with kaolin and minerals including the Al and Si, which are used for activating ceramic particles in many cases. However, activation of particles depends on the amount of eluted alkali in their crystal phases [4]. It has been reported that chemical treatment with planetary ball-milling process activates the ceramics particles. Through this process, it is possible to convert waste material into valuable ceramics by using chemically activated ceramics particles.

The objective of the present investigation is to prepare an activated particle from paper sludge with planetary ball-milling and to fabricate ceramic without firing using the treated waste particles.

2. EXPERIMENTAL PROCEDURE
The paper sludge ash powder was used as a raw material. Planetary ball mill was used for mechanical treatment of solids. Each milling was carried out with a 100g sample in a 500ml capacity zirconia pots containing zirconia balls of 10mm diameter. The mill was rotated at constant revolution speed at 300rpm. The raw material was ground for about 10 to 60 minutes.

The products obtained by planetary ball milling were added with 2N: KOH solution. Afterwards, they were mixed with little amount of potassium silicate glass ($\text{SiO}_2/\text{K}_2\text{O}=3.0$) solution as reaction accelerator. The resulting pre-mix slurry was coercively agglutinated by centrifugal force. Finally, they were demolded and dried at room temperature for 7 days in air.

Phases present and crystallite size of the products treated by planetary ball-milling were identified by XRD and their morphologies were observed by SEM. The specific surface area and particle size were measured by BET and laser diffractometry, respectively.

Elution analysis was conducted, using inductively-coupled plasma technique (ICP), to determine the amount of eluted $\text{Al}^{3+}$ ions in liquors collected after teaching. In 100ml plastic beaker, 0.1N: KOH solution was prepared and about 50ml was used for each trial. The solution was kept warmed beforehand by a magnetic stirrer and kept stirred at room temperature (25 °C) for 1hr. Finally, the tested liquor was collected by a centrifugal separation technique.

3. RESULTS AND DISCUSSION

Fig.1 shows the XRD pattern of the products treated for about 0 to 60 minutes. (0min means not treated raw material). The raw material is mainly composed of the gehlenite (Ca$_2$Al$_2$SiO$_7$). It is well known that paper sludge ash is also composed of gehlenite, soda-lime glass and aluminum silicate etc. These minerals, however, such as aluminum silicate, were not detected by XRD signifying amorphous structure.

Even in the samples treated for long time, the peak intensity of crystallite was decreased. Therefore, crystallite size shows an abrupt decrease at 20min, above which it decreases gradually. It was suggested that the particles were involving in miniaturization and amorphilization with influence of high energy action of ball milling. This behavior of crystallite size such mineral is consistent with that of result reported in a previous study [5].

Fig. 2 shows the average particle size and specific surface area of the products treated for 0 to 60minutes. The average particle size was 29.1-2.1(μm), which was appreciable changed as compared with the raw powder. When the milling time is longer than 20min, the average

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particle size was not almost change despite long milling time. On the other hand, the specific surface area of the products decreases abruptly for 20 minutes milling. Above this milling time, the surface area decreases gradually.

Increasing milling time, but, in the samples milled for 20 to 40 minutes, these particles are agglomerating. This fact agrees with those obtained by average particle size and specific surface area as depicted in Fig. 3.

Fig. 4 shows the elution analysis of the treated particles. It is known that Al³⁺ ion mainly leached out from ceramic particles [6], when ceramics particles are mixed with potassium silicate glass solutions. The paper sludge ash showed a high elution of Al³⁺ ion leaching in all samples. Al³⁺ ion content was increased with an increasing in the milling time. The eluted amount shows a maximum at 20 minutes. We found that the eluted amount depends on the specific surface area, that is, the content of paper sludge ash. Finally, Fig. 6 shows a picture of solidified ceramic was fabricated using chemically activated ceramic particles treated for 20 minutes.

![Fig.1. XRD patterns of the products treated by planetary ball-milling at 0-60min.](image1)

![Fig.2. Specific surface area and mean particle size of the products treated by planetary ball-milling at 0-60min.](image2)

![Fig.3. SEM photographs of the products treated for 10 to 60minutes. All of the treated samples consist of fine particles having decreasing size with an increasing milling time, but, in the samples milled for 20 to 40 minutes, these particles are agglomerating. This fact agrees with those obtained by average particle size and specific surface area as depicted in Fig. 3.](image3)

![Fig.4. Specific surface area and mean particle size of the products treated by planetary ball-milling at 0-60min.](image4)

![Fig.5. SEM photographs of the products treated for 10 to 60minutes. All of the treated samples consist of fine particles having decreasing size with an increasing milling time, but, in the samples milled for 20 to 40 minutes, these particles are agglomerating. This fact agrees with those obtained by average particle size and specific surface area as depicted in Fig. 3.](image5)

![Fig.6. SEM photographs of the products treated for 10 to 60minutes. All of the treated samples consist of fine particles having decreasing size with an increasing milling time, but, in the samples milled for 20 to 40 minutes, these particles are agglomerating. This fact agrees with those obtained by average particle size and specific surface area as depicted in Fig. 3.](image6)
Despite of the non-firing step, high bulky density was achieved at 1.68 g/cm³ when it was compared with the use of untreated particles. Therefore, it can be concluded that chemically treated paper sludge ash with planetary ball-milling is suitable for making useful ceramics without firing.

4. CONCLUSION
Non-firing ceramic was successfully fabricated from paper sludge ash. This was done by combination of planetary ball-milling and chemical activation of ceramic particles. This mechanical treatment was found to have a significant affect on the fabrication of non-fired ceramics. Through this process, the waste material could be shaped into valuable ceramics.

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