SYNTHESIS, CHARACTERIZATION AND RHEOLOGICAL PROPERTIES OF ALUMINA-ZEOLITE MIXTURES

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ABSTRACT

In this research study, Alumina-based zeolite ceramic mixtures were prepared using mechanical activation method. An elaborate examination of structure and microstructure have been carried out using X-ray diffraction (XRD) and scanning electron microscopy (SEM). The XRD investigation of the natural zeolite from Tokaj region has shown many phases with different contents of minerals like montmorillonite, quartz, cristobalite, clinoptilolite, and calcite. Moreover, the compacting and rheological characteristics of the complex mixtures were studied based on forming pressures and times. When the applied pressure is gradually increased, a dramatic change in the rheological behaviour of the complex ceramic mixtures has been observed.

1. INTRODUCTION

The research development in the field of ceramic industries is becoming a more popular and fast-growing field of material science [1-16]. Aluminium oxide (alumina; Al2O3) has superior thermal, mechanical, physical and chemical properties when compared with many other ceramics materials. These characteristics make alumina the
most popular candidate for several applications [17] such as refractories, an electronic substrate, bio-ceramic material, wear resistance materials, catalyst, cutting tools and armour [18], and it is the same to the ceramic industry as steel is to the metallurgical industry [19]. But generally, ceramic materials suffer from brittle behaviour which leads to inferior mechanical characteristics that restrict their applications. Intensive work has been devoted lately to specify methods for synthesizing ceramics reinforced ceramic-matrix composites (CMCs) with better properties. Since the pioneering work of Niihara in the 1990s [20]. The introduction of reinforcement as a secondary phase is considered to be among the most promising methods of enhancing the properties of polycrystalline ceramic-based materials. A flurry of researches on the topic demonstrated that reinforcing polycrystalline alumina with ceramic materials lead to a significant improves in the mechanical strength [21, 22, 23], fracture toughness [24, 25] wear resistance [26, 27] and creep resistance [28], compared to monolithic polycrystalline alumina. However, even though large numbers of researches have been done in the field of “ceramic-composites” but it’s still a topic of scientific research. Due to the complex structure of the ceramic reinforced ceramic composite and the varieties on the physical and mechanical characteristics, different forming methods could be used to prepare ceramic-based composite materials such as, uniaxial pressing, hot pressing, injection molding, extrusion, slip casting, spark plasma sintering, hydrothermal, sol-gel and several other technical methods [29]. Selecting a suitable forming technique, demand knowledge of specific process parameters, for instance, applied pressure, force, and power consumption. Such variables are strongly attached to the rheological characteristics and, therefore, the material parameters of the compounds [30].

This research aims to prepare Alumina-zeolite ceramic mixtures materials through mechanical activation technique. Analysis of the complex structure and microstructure of the mixture has been presented using several characterization techniques including X-ray diffraction (XRD) and scanning electron microscopy
(SEM), In addition, the change in rheological properties corresponding to the applied force during the compaction has been described.

2. MATERIALS AND METHODS

Alumina-based zeolite mixtures were produced using mechanical activation techniques. Al$_2$O$_3$ powder (98 %; MOTIM) and Zeolite powders from Tokaj region (Hungary) were used as starting materials. Stoichiometric amounts of these powders were weighted using sensitive balance according to the compositions as demonstrated in table 1, the prepared mixtures were ground using a planetary miller with silica ball for 15 minutes in 150 rpm. The ground powders were used for the rheological investigation.

Table. 1 The percentage of the raw materials used to prepare the mixtures

<table>
<thead>
<tr>
<th>Alumina %</th>
<th>Zeolite %</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>0</td>
</tr>
<tr>
<td>90</td>
<td>10</td>
</tr>
<tr>
<td>80</td>
<td>20</td>
</tr>
<tr>
<td>70</td>
<td>30</td>
</tr>
<tr>
<td>60</td>
<td>40</td>
</tr>
<tr>
<td>50</td>
<td>50</td>
</tr>
</tbody>
</table>

2.2. Characterization

The raw materials (zeolite and alumina) ceramic powders were characterized by X-ray diffractometer (XRD) operated in the Bragg-Brentano geometry and the samples were scanned between 2θ of 10-70° with a scanning rate of 1°/min and a step size of 0.01016° using CuKα radiation ($\lambda= 1.54184$ Å). For the computer-based investigation, DIFFRACT measurement software was used. The topographical feature
and the surface morphology of both raw materials and the prepared ceramic samples were examined using scanning electron microscopy (SEM).

3. RESULTS AND DISCUSSION

3.1. Structures

XRD investigation of the natural zeolite reveals the existence of several minerals including, montmorillonite, quartz, cristobalite, clinoptilolite, and calcite as shown in figure 1 and table 2. The amount of Cristobalite was found to be the largest amount represent about 50% of the total percentage then comes Montmorillonite (20%) then the other minerals. According to the chemical composition analysis the total amount of SiO$_2$ in natural zeolite account to 82.92 w% which confirm to be found as pure silica or in different minerals such as Montmorillonite and Clinoptilolite.

Figure. 1 XRD diffractogram of the natural Zeolite
Table 2. Mineralogical constituents, Chemical composition and loss of ignition (LOI) of the natural zeolite in wt%, obtained from XRD analysis

<table>
<thead>
<tr>
<th></th>
<th>Quartz</th>
<th>Cristobalite</th>
<th>Montmorillonite</th>
<th>Calcite</th>
<th>Clinoptilolite</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>wt %</td>
<td>8.00</td>
<td>50.00</td>
<td>30.00</td>
<td>2.00</td>
<td>10.00</td>
<td>100.00</td>
</tr>
<tr>
<td>CaO</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1.12</td>
<td>1.12</td>
</tr>
<tr>
<td>SiO₂</td>
<td>8.00</td>
<td>50.00</td>
<td>19.13</td>
<td></td>
<td>5.79</td>
<td>82.92</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>4.06</td>
<td></td>
<td></td>
<td></td>
<td>1.89</td>
<td>5.95</td>
</tr>
<tr>
<td>MgO</td>
<td>3.21</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3.21</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.74</td>
<td></td>
<td></td>
<td></td>
<td>0.57</td>
<td>1.31</td>
</tr>
<tr>
<td>CO₂</td>
<td>0.88</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.88</td>
</tr>
<tr>
<td>H₂O</td>
<td>2.87</td>
<td></td>
<td></td>
<td></td>
<td>1.60</td>
<td>4.47</td>
</tr>
<tr>
<td>LOI</td>
<td>0.00</td>
<td>0.00</td>
<td>2.87</td>
<td>0.88</td>
<td>1.75</td>
<td>5.50</td>
</tr>
</tbody>
</table>

3.2. SEM investigation of the raw materials

Figure 2 shows secondary electron images of the natural zeolite and alumina powders. The typical grains size of zeolite and alumina were found to be in the range of 0.5 - 25 µm and 0.5 - 50 µm, respectively. The microstructure of the alumina-zeolite mixture prepared by milling in the planetary ball mill is shown in figure 3. It is clear that the milling process leads to a dramatic reduction in the particles size.
Figure 2  SEM images of a) natural zeolite and b) alumina powders

Figure 3  SEM images of the alumina-zeolite mixture powders a) 60% alumina-40% zeolite b) 50% alumina-50% zeolite

3.3 Rheological behaviour of the ceramic samples
Figure 4. The connection a) between the compaction time and the applied pressure, b) between the compaction time and the deformation, c) the compaction time and the pressure d) the applied force and the deformation

In the first place, the compaction experiment was carried out at relatively low applied pressure, Fig. 4 demonstrates the relationship between the compaction time and the applied pressure, the compaction time and the deformation, the compaction time and the pressure and the applied force and the deformation of the experiment, at the very beginning and when the ceramic mixtures were subjected to relatively low applied pressure, a dramatic growth in the deformation has been observed possibly due to the fact that the materials behave like a typical viscous material. Further stress generates deformation which leads to rearrangement in the microstructure through sliding of the different grain at the grain boundary filling the gaps (pores) of the materials, in this stage, the materials can be considered as a viscous-elastic body. In the next stage, the materials are further densified through continues filling of the gaps, while the applied pressure was fast grown, the compaction is nonlinear, due to the frictions occur between the mixture particles and the die margins. In this stage, the materials can be modelled as the parallel-linked viscous-elastic body. In the final stage, very small compaction was obtained in correspondence to high applied pressure, as the materials
are highly densified and the particles of the mixture are very close to each other, in this stage, the materials can be represented as “quasi-plastic” material.

4. CONCLUSION

From this research work we can conclude that, the alumina-zeolite mixtures which produced by ball milling have complex structure due to the existence of many minerals, for instance, alumina, montmorillonite, quartz, cristobalite, clinoptilolite, and calcite, this complex structure reveals different rheological characteristic at different applied pressure which can be described by gömze model [31]. The mechanical stress relaxation takes place during the uniaxial pressing of ceramic mixtures and continue for a longer time but never reach zero showing the residual strain.
REFERENCES


