The Effect of Type and Amount of Acid and Base Catalysts on the Crystallization and Morphology of Mullite Particles Synthesized by the Sol-Gel Method

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1. Introduction

High temperature (synthesizing) routes are the most common methods for synthesizing the mullite phase containing ceramics. However, it is possible to synthesize mullite containing compounds at lower temperature via the sol-gel method. Synthesizing the mullite ceramics via the sol-gel route is dependent on some factors such as homogeneity, pH of the precursors, type and amount of the catalysts, hydrolysis and condensation rate, time of aging and drying, and heat treatment cycles. Among the mentioned factors, changing the pH during the sol-gel process could change the chemical and physical properties of the final product. Utilizing different amount of acid and base catalysts is the easiest way to investigate the change of the pH variations during the sol-gel process. The effect of the change of the pH during the synthesis process on the chemical and physical properties of the final ceramics has not been studied yet. Therefore, the current study aims to investigate the effect of the type and amount of acid and base catalysts on the microstructural properties of mullite phase particles prepared by the sol-gel method.

2. Materials and Methods

The mullite particles were synthesized via the sol-gel route. Nona-hydrated aluminum nitrate (ANN), Tetraethyl orthosilicate (TEOS), ethanol, and water were used as starting materials. After the precise preparation method, obtained gels were dried in an inert atmosphere and heated with specific heat treatment cycles at 900, 1100, 1250, 1350, and 1550 ℃. Differential Scanning Calorimetry and thermogravimetric analyses (DSC-TG) were used to identify the proper crystallization temperature and weight loss of the particles. To investigate the phase crystallization and the morphology of the created phases, X-ray diffraction pattern (XRD) and scanning electron microscopy (SEM) analyses were utilized, respectively. All sample codes were introduced in Table2.

3. Results and Discussion

The XRD analysis was performed to study the effect of the selected acid or base catalyst during the sol-gel process of the particles and heat treatment temperatures on the crystallization trend of the synthesized samples.

Table 2. Sample Codes

Sample code	Synthesis Condition
NA	Synthesized sample in acidic condition
NB	Synthesized sample in the base condition
W0	Synthesized sample at pH=0
W13	Synthesized sample at pH=13

The XRD results of the W0 samples heat-treated at 900, 1100, 1250, 1350, and 1550 ℃ were depicted in Fig 1. According to Fig 1, the samples heat-treated at 900 and 1100℃ had amorphous nature. Thermal treatment of the samples at 1250, 1350, and 1550 ℃ led to the crystallization of the samples. Therefore, the crystallization of the mullite phase created some sharp peaks in the XRD graphs of the samples.

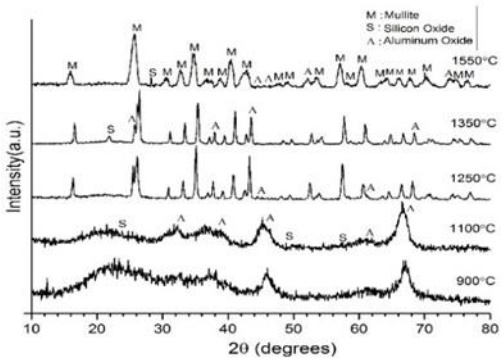

Fig 1. XRD results of the synthesized samples at pH=0 (w0) heat-treated at 900, 1100, 1250, 1350, and 1550 ℃

Fig 2 shows the thermogravimetric and Differential Scanning Calorimetry (DTA/TG) results of the W0 as-synthesized particles. The whole weight loss of the w0 sample during the test was approximately 61%. The first endothermic peak at the range of 150-220 ℃ could be attributed to some endothermic events such as water and ethanol evaporation, dehydration, and removal of catalysts and nitrate groups. The mentioned event caused approximately 58% drop in weight. The second weight loss which is 2% was attributed to the hydrolysis process and oxidation of the remained organic OR groups.

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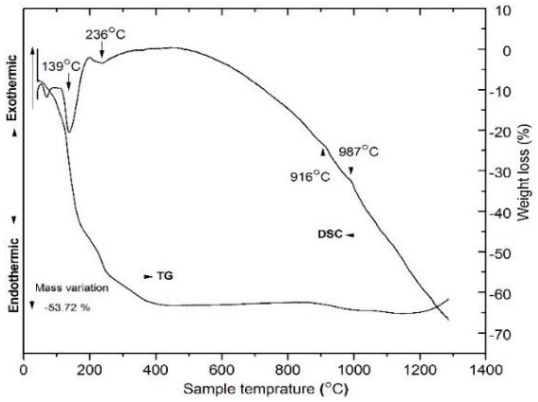

Fig 2. DSC-TG results of the synthesized samples at pH=0 (w0).

The SEM analysis was performed to characterize the microstructure of the samples synthesized in the presence of different acid or base catalysts. Based on the SEM results, the samples synthesized in acid conditions with constant pH have plate shape morphology while synthesizing the particles in base pH resulted in the formation of spherical morphology. SEM images of the w0 particles were depicted in Fig 3 and 4.

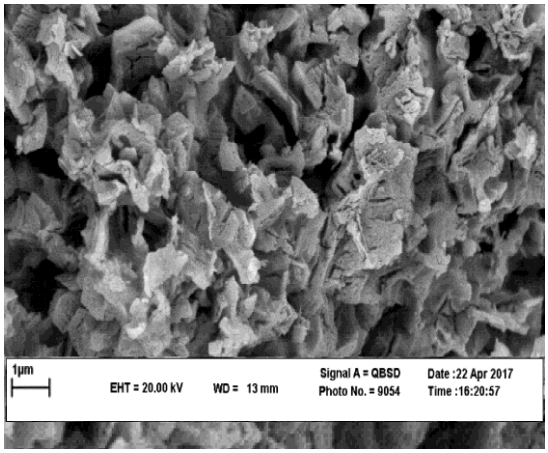

Fig 3. SEM images of the synthesized samples at pH=0 (w0) heat-treated at 1250℃.

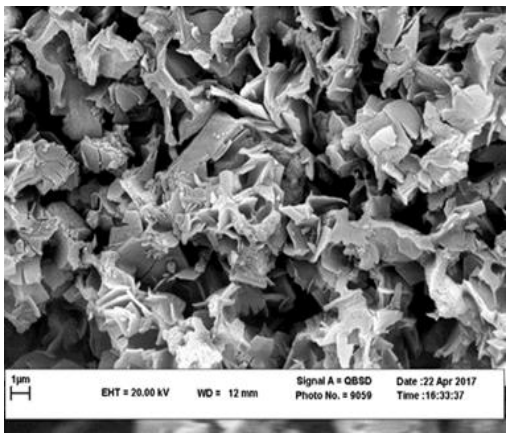

Fig 4. SEM images of the synthesized samples at pH=0 (w0) heat-treated at 1550℃.

4. Conclusion

XRD analysis showed that mullite is crystallized in all samples with acidic or basic catalyst. Furthermore, the mullite phase created at 900 and 1100℃ were grown up with increasing heat treatment temperature to 1250 and 1350℃. The DSC-TG results also showed that the temperature of the formation of the transformation phases was completely dependent on the homogeneity of the Al-Si compositions during the sol-gel synthesizing process. Based on the SEM results, the samples synthesized in acid conditions with constant pH have plate shape morphology while synthesizing the particles in base pH resulted in the formation of spherical morphology. In addition, some small alumina phases were found in the samples synthesized in acid pH, which were created during heterogeneous nucleation. In conclusion, comparing the results showed that the samples synthesized in constant acid conditions have better chemical and physical properties.