Journal of Ceramic Processing Research. Vol. 14, No. 1, pp. 17~21 (2013)

J O U R N A L O F
Ceramic Processing Research

Synthesis of alumina nano powder by a gel combustion method

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In this study a gel combustion method was used to prepare alumina nano powder. Aluminum nitrate, citric acid and glycine and urea fuels were used. An analysis of the effect of the type of fuels and their synthesis was done and followed by calcination at temperatures of 400-700-1000 °C. The results showed that using urea as the fuel to raw material ratio of 1 to 1 and in the fuels synthesis, citric acid to urea (50% + 50%) ratio of 1 to 1 and a calcination temperature of 700 °C, the powerful optimum conditions for the synthesis of aluminum oxide nano powder Al_2O_3 at dimensions of 20 nm was obtained.

Key words: Nano powder, Alumina, Gel combustion, Urea, Glycine, Citric acid.

Introduction

The importance of nano alumina is due to the high pressure solidity, high strength, high resistance to erosion, high heat conduction, and high electrical resistance, transparency in front of frequencies or microwave blaze.

There are different methods for the production of nano powders. Such as Sol gel, combustion gel, co precipitation, and hydrothermal [1, 2]. The importance of combustion gel is its higher purity, lower energy and the minimum amount of agglomeration. In the combustion gel method the raw materials are usually in the form of nitrate complexes and a fuel which is dissolved in water. The complex solution is heated until it is converted from a sol to a gel with a high viscosity.

The temperature increase causes an exothermic combustion process. The organic material as the reducing agent and nitrates as the oxidizing agent convert the gel to very granular and highly porous materials and then with the help of calcination the ultimate product will be obtained [3, 4]. Pathak and others [5] also by using the combustion gel method and urea fuel synthesized alumina nano powder and succeeded to synthesize alumina nano particles in the dimensions of 50 to 200 nm.

Khorami and others [6] using the combustion gel method and glycine fuel synthesized alumina nano particles and by the calcination of the combustion ash in the temperature of 1100 °C obtained alpha alumina.

Shuravlev and others [7] using the gel method synthesized aluminum oxide nano particles and analyzed different phases of alumina. These researchers used a calcination temperature of 1000 °C for the calcination of combustion ash and they succeeded to obtain nano particles of 20 - 70 nm.

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Experimental procedures

For all the experiments different materials were used such as aluminum nitrate, urea, glycine and citric acid.

Table 1 shows the characteristics of the raw materials. Materials were weighed, dissolved in distilled water and homogenized on a magnetic stirrer based on table 2. After mixing and homogenizing the metallic nitrate with fuel, while heating, the gel combustion processed was carried out and Al_2O_3 nano powders were synthesized. The powders obtained were calcined at 400, 700 and 1000 °C and then XRD and SEM were used and the results were analyzed.

Results and discussion

At first raw material and fuel were selected by a ratio of 1 to 1 and to obtain an appropriate temperature for calcination 3 temperatures of 400, 700 and 1000 °C were tested. The XRD patterns of the samples are shown in 1-3 figures and all the crystallites were measured by the Scherrer technique. Scanning electron microscopy (SEM) pictures of the samples are shown in figures 4 to 6.

The size of the synthesized crystallite nano powders with different fuels at different temperatures are shown in table 3.

As is shown in table 3, the effect of a high calcination temperature is 700 °C for urea fuel and 1000 °C for citric acid and glycine fuels and it is obvious by increasing the temperature from 400 to 1000 °C, the size of the crystallites is decreased for citric acid and glycine fuels, but for urea this trend is

Table 1. Raw material characteristics.

Raw materials	Formulation	Molecular weight (g/mol)	purification	Physical state	manufacturer
Aluminum nitrate	$Al(NO_3)_3.9H_2O$	375	99%	solid	MERCK
Urea	NH_2CoNH_2	60	99%	solid	MERCK
Glycine	$NH_2CH_2.COOH$	75.05	99%	solid	MERCK
Citric acid	$C_6H_8O_7.H_2O$	210.14	99%	solid	MERCK

Table 2. Materials weighed in this research.

Sample code	Fuel to nitrate ratio (f/m)	Aluminum nitrate (g)	fuels (g)	temperature (°C)
U1	1 to 1	15	6	Before calcinations
U2	1 to 1	15	6	400
U3	1 to 1	15	6	700
U4	1 to 1	15	6	1000
G1	1 to 1	15	3	Before calcinations
G2	1 to 1	15	3	400
G3	1 to 1	15	3	700
G4	1 to 1	15	3	1000
C1	1 to 1	15	8.4	Before calcinations
C2	1 to 1	15	8.4	400
C3	1 to 1	15	8.4	700
C4	1 to 1	15	8.4	1000

Table 3. Crystalite sizes of Al_2O_3 nano powder by the Scherer technique.

sample	nm
U1	29
U2	13
U3	9
U4	18
G1	32
G2	19
G3	16
G4	12
C1	34
C2	28
C3	21
C4	15

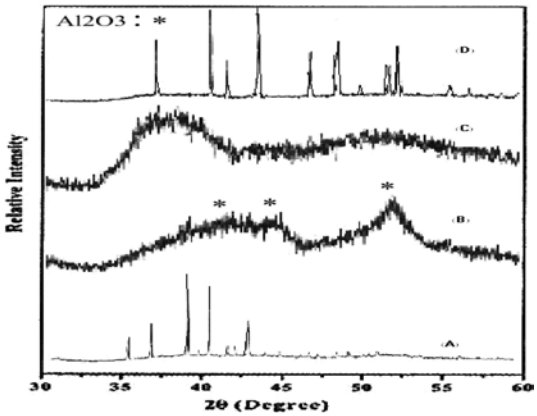

Fig. 1. XRD patterns of synthesized nano powders with a stoichiometric amount of urea (A) before calcination and calcined at the temperatures of (B) 400 °C, (C) 700 °C and (D) 1000 °C.

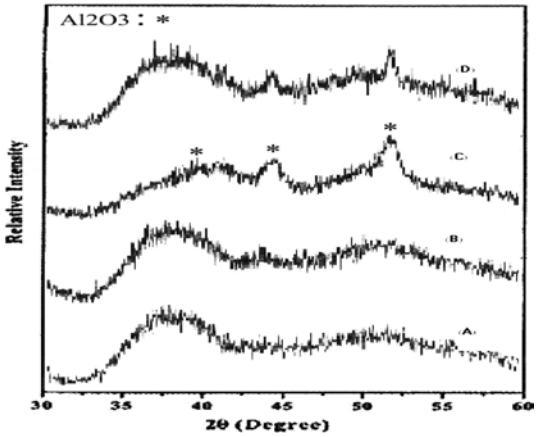

Fig. 2. XRD patterns of synthesized nano powders with a stoichiometric amount of glycine (A) before calcination and calcined at the temperatures of (B) 400 °C, (C) 700 °C and (D) 1000 °C.

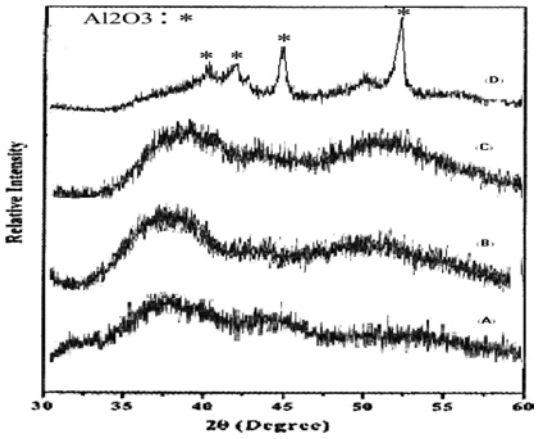

Fig. 3. XRD patterns of synthesized nano powders with a stoichiometric amount of citric acid (A) before calcination and calcined at the temperatures of (B) 400 °C,(C) 700 °C and (D) 1000 °C.

not systematic. With increasing the calcination temperature, the size of the particles is increased, this process can be attributed to the preparation of the necessary activation energy for the growing of the crystallites at higher temperature. The reason for this is generally with increasing temperature, the size of the particles is decreased is probably because of the

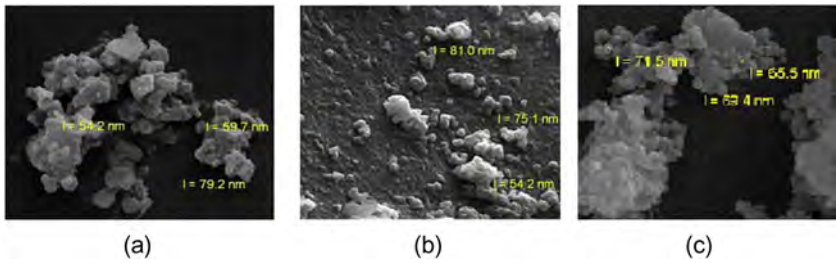

Fig. 4. SEM pictures of synthesized powders with a stoichiometric ratio of urea calcined at the temperatures of (a) 400 °C, (b) 700 °C and (c) 1000 °C.

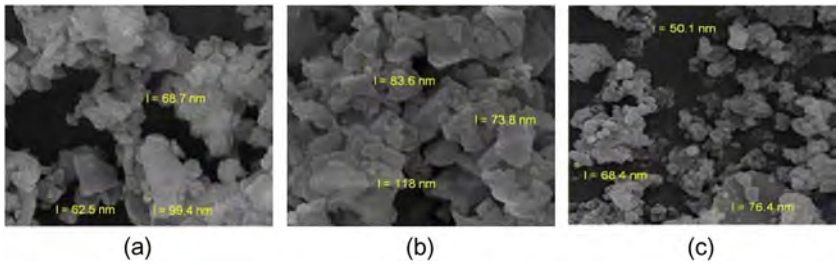

Fig. 5. SEM pictures of synthesized powders with a stoichiometric ratio of glycine calcined at the temperatures of (a) 400 °C, (b) 700 °C and (c) 1000 °C.

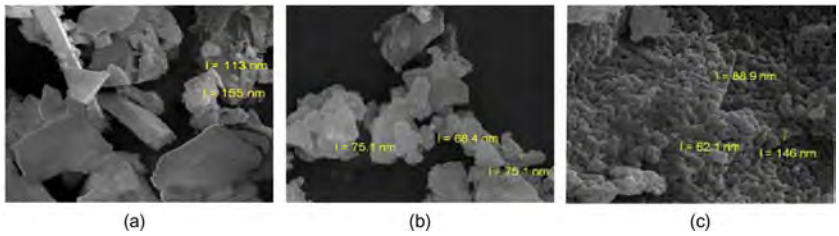

Fig. 6. SEM pictures of synthesized powders with a stoichiometric ratio of citric acid calcined at the temperatures of (a) 400 °C, (b) 700 °C and (c) 1000 °C.

Table 4. The weighed materials for the complexation of the fuels.

Sample code	Fuel to Nitrate (f/m)	Al $(NO_3)_3$. $9H_2O$ (g)	Urea Fuel (g)	Glycine Fuel (g)	Acid citric Fuel	Fuel mixtures percent	Temperature (°C)
A	1 to 1	4	0.75		1.05	%50U + %50C	1000
B	1 to 1	4	0.9		0.3	%60U + %40C	1000
C	1 to 1	4	1.12		0.18	%75U + %25C	1000
D	1 to 1	4		0.37	1.05	%50G + %50C	1000
E	1 to 1	4		0.45	0.84	%60G + %40C	1000
F	1 to 1	4		0.56	0.52	%75G + %25C	1000
G	1 to 1	4	0.7	0.37		%50U + %50G	1000
H	1 to 1	4	0.9	0.3		%60U + %40G	1000
K	1 to 1	4	1.12	0.18		%75U + %25G	1000

formation of another phase of alumina. Since the alumina phase changes from gamma to alpha at about 1150 °C, it seems that at the temperature of 1000 °C, alpha alumina nuclei started to nucleate and since the temperature had not been high, they could not grow. In the case of using urea fuel, it seems that at a temperature of 700 °C, because combustion is generated a higher temperature and this matter did not occur, the act of calcination and combustion caused the actual temperature to be increased to more than 700 °C. Therefore alpha phase has been formed at a temperature of 700 °C. So with an increase of the temperature to 1000 °C, this phase causes the alumina particles to grow and therefore it can be concluded that a suitable temperature for calcination in the case of applying urea fuel is 700 °C and in the case of applying

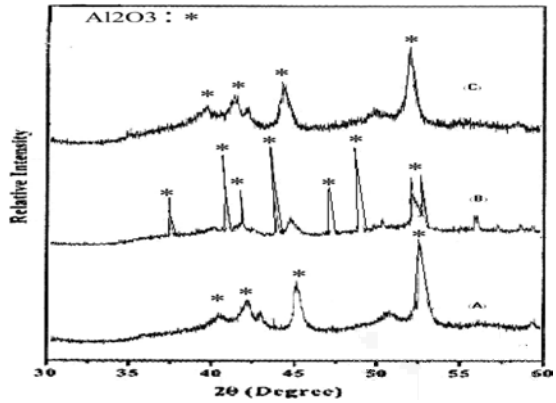

Fig. 7. XRD pattern of the synthesized nano powders with the fuel mixtures of (A) (%75 urea + %25 citric acid) (B) (%60 urea + %40 citric acid) (C) (%75 urea + %25 citric acid) at a temperature of 1000 °C.

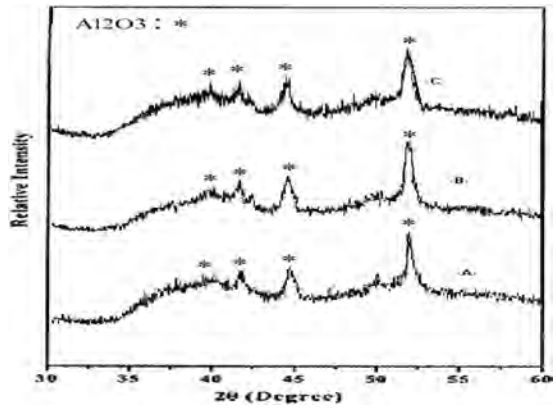

Fig. 8. XRD pattern of the synthesized nano powders with the fuel mixtures of (A) (%75 glycine + %25 citric acid) (B) (%60 glycine + %40 citric acid)(C) (%75 glycine + %25 citric acid) at a temperature of 1000 °C.

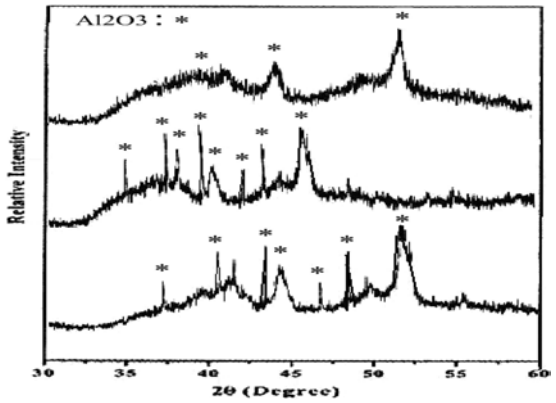

Fig. 9. XRD pattern of the synthesized nano powders with the fuel mixtures of (A) (%75 urea + %25 glycine) (B) (%60 urea + %40 glycine) (C) (%50 urea + %50glycine) at a temperature of 1000 °C.

the citric acid and glycine fuels it is 1000 °C and the best size of crystallites is for the urea fuel at 700 °C. Another analyzed parameter in the synthesis of alumina nano powder is using mixtures of the fuels. In table 4 the weighed materials for the mixtures of the fuel are shown.

The XRD patterns of the fuel mixtures are shown in figures 7 to 9. The sizes of the crystallites has been obtained by Scherrer technique and also scanning electron microscopic pictures of sample using fuel mixtures are shown in figures 10 to 12.

The size of the synthesized crystallite nano powders with fuel mixtures are shown in table 5.

With regard to the results, we understand that when we use the ratios (%40 + %60) in the fuel mixtures of citric acid and urea and (%50 + %50) in the fuel mixtures of urea and glycine, the maximum size of the

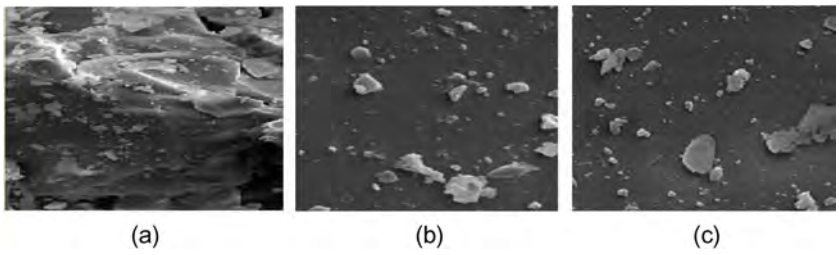

Fig. 10. SEM pictures of the synthesized nano powders with the fuel mixtures of (a) (%75 urea + %25 citric acid) (b) (%60 urea + %40 citric acid) (c) (%75 urea + %25 citric acid) at a temperature of 1000 °C.

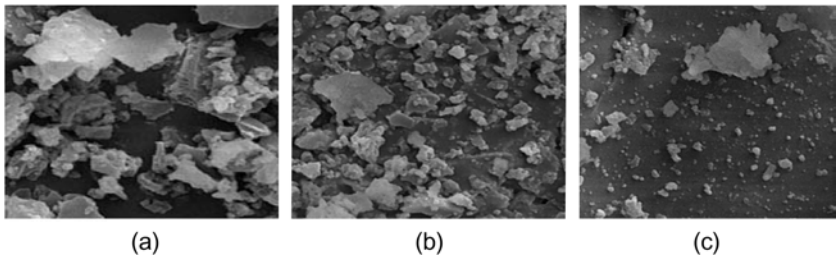

Fig. 11. SEM pictures of the synthesized nano powders with the fuel mixtures of (a) (%75 glycine + %25 citric acid) (b) (%60 glycine + %40 citric acid) (c) (%75 glycine + %25 citric acid) at a temperature of 1000 °C.

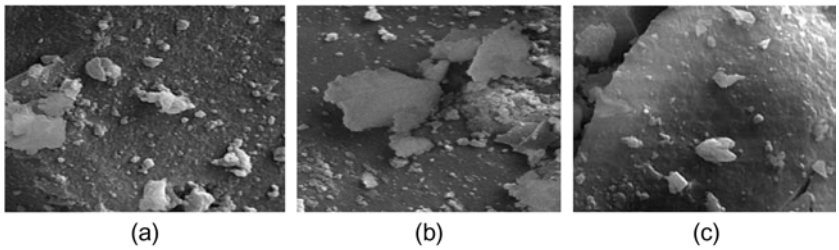

Fig. 12. SEM pictures of the synthesized nano powders with the fuel mixtures of (a) (%75 urea + %25 glycine) (b) (%60 urea + %40 glycine) (c)(%50 urea + %50 glycine) at a temperature of 1000 °C.

Table 5. The size of the synthesized crystallite nano powders with fuels complexation.

Sample code	nm	Sample code	nm
A	10	F	12
B	31	G	32
C	15	H	29
D	11	K	30
E	18		

crystallites is obtained. When we use the ratios (%50 + %50) in the fuel mixtures of citric acid and urea and citric acid and glycine, this leads to the production of smaller particles that show a decrease in the reaction heat and powders with smaller crystallite sizes are produced.

Therefore with the use of fuel mixtures it is possible to change the temperature and the time of the combustion so that the alumina crystallites produced after reaching a critical size have no chance of growing more and their increase in size at a higher temperature is prevented.

Conclusions

- It is possible to prepare nano powders using a gel combustion method.
- It is possible to obtain a nano alumina with the size of 20 nm using aluminum nitrate as the agent and urea, citric acid and glycine as the fuel in the gel combustion method.
- In examining the calcination temperatures of 400 °C, 700 °C and 1000 °C using urea, citric acid and glycine fuels, the most suitable temperature for urea is 700 °C and for citric acid and glycine is 1000 °C.
- In examining the fuel mixtures of urea with citric acid, urea with glycine and glycine with citric acid the best result for the crystallite sizes is for the fuel mixtures of urea with citric acid (%50 + %50).

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