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The Effect pH on Purification of Fe in the Preparation of High Purity Al2O3 by Crystallization

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Abstract—minium; There are many advantages of high purity alumina. hydroxide as raw material, through making ammonium aluminum sulfate as precursor, prepared high purity alumina after high temperature in this study. By adding ammonia to adjust the pH of the precursor in this experiment. make range of pH from 1 to 5. Sintering in the muffle furnace and heat for 2 hours under 1200℃, alumina is produced. Using atomic absorption spectrophotometer to measure the content of Fe, the relationship between content of Fe and pH, the differential thermal analyzer was used to study the pyrolysis process of precursor, the reaction process of precursor were analyzed, and use XRD to analyze phase and content of Fe of alumina, use SEM observe crystal morphology of high purity alumina. Results showed that Fe content of alumina increases decreases first and increase after with pH, under pH=3, impurities of Fe content in the minimum is 7.56 mg/kg, alumina crystals for granular and dispersivity is good, stable after 1200 degrees of calcined, XRD results showed that only diffraction peak of Al203.

Keywords-Precursor; high purity alumina; Fe; pH; Crystallization

I. INTRODUCTION

There are many advantages of high purity alumina, such as high melting point, high hardness, etc. In recent years it is widely used in fluorescent body with carrier, single crystal materials, advanced ceramics, etc [1-5]. As emphasis on problems of growing energy in the country, by possesses the advantages of high energy saving, wide attention is paid on LED[6]. Therefore, the development of high purity alumina which can be applied to the LED become urgently needs to be solved.

There are a lot of methods in the prepare of high purity alumina at present, the method of sol gel is adopted in the production of high purity alumina, but process is complex and the cost is higher, and produce make pollution to the environment; method of alcohol aluminum hydrolyzing, while reducing the environmental pollution, but still has the problem of high cost; Mercuric chloride activation hydrolysis can make the new impurities in to high purity alumina[8-15]. Using $Al(OH)_3$ to prepare high purity alumina, has many advantage, such as, cheap raw material, production process is simple and suitable for industrial production. This method is widely used in manufacturer of high purity alumina. But impurity content of Fe is higher in this method. Impurities of Fe and aluminum elements forming phase, infect the performance of high purity alumina[10]. By adding ammonia to adjust the pH of the precursor in this experiment. Using atomic absorption spectrophotometer to measure the content of Fe, the relationship between content of Fe and pH, the differential thermal analyzer was used to study the pyrolysis process of precursor, the reaction process of precursor were analyzed, and use XRD to analyze phase and content of Fe of alumina, use SEM observe crystal morphology of high purity alumina

II. EXPERIMENT

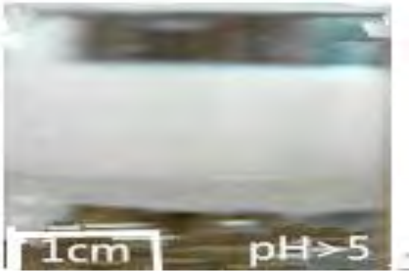
Add a certain amount of $Al(OH)_3$ and H_2SO_4 of 98% in the container. By controlling the addition of ammonia, change the pH of the precursor, make range of pH from 1 to 5. Sintering in the muffle furnace and heat for 2 hours under 1200℃, alumina is produced.

Using thermogravimetric analyze and evaluate the pyrolysis process of precursor, nitrogen as the protection atmosphere, heating speed of 20 k/min, heating range for 50℃ to 1300℃. Using the Netherlands PANalytical x ray polycrystalline diffraction instrument, observe the XRD diffraction peak under different pH, accelerating voltage 40 kv, X-ray tube for Cu target, scan rate of 0.557°/s, working current is 40 mA. German KYKY2800, resolution is 2 nm, analyze surface morphology of the powder. And through the EDS to test contained elements of powder qualitatively.

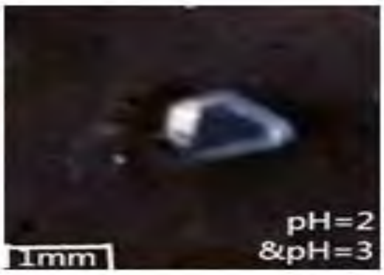
III. RESULTS AND DISCUSSION

A Optical image analyze crystallization of ammonium aluminum sulfate under different pH

(a)



(b)



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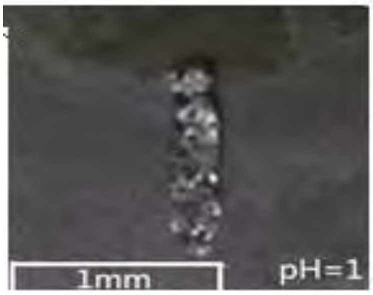
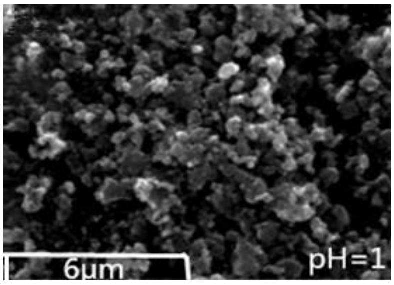


(d)

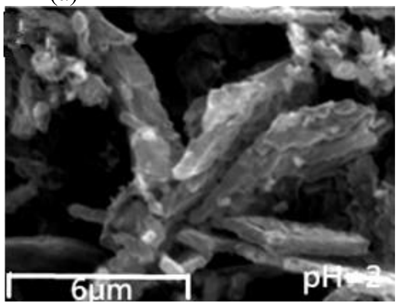
Figure 1. (a)shape of crystal particle under pH = 1, (b) shape of crystal particle under pH=2 and pH=3, (c) sedimentation of grain under pH<5, (d) flocculent of crystallization under pH>5

As shown in Fig .1, it can be seen from (a) under pH<5, precipitation of ammonium aluminum sulfate crystal in the form is grain.aUnder pH>5, began to appear flocculent crystallization in the solution. It may be that when pH>5, a solution will be aluminum hydroxide precipitation in the form of floc. With the increase of pH, flocculent crystallization increase gradually, so that under pH>5, flocculent crystals suspended inside the container, as shown in Fig .(b). From Fig .(c) and (d) showed that under pH from 1 to 3, with the increase of pH, particles of crystal decreases gradually, from rod-shaped particles into triangle granules.

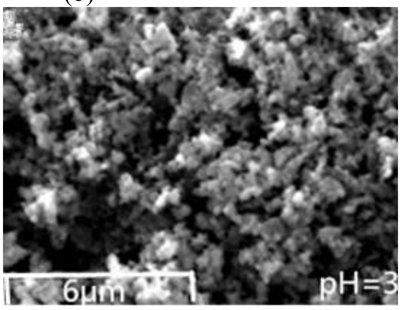
B SEM analyze alumina morphology under different pH



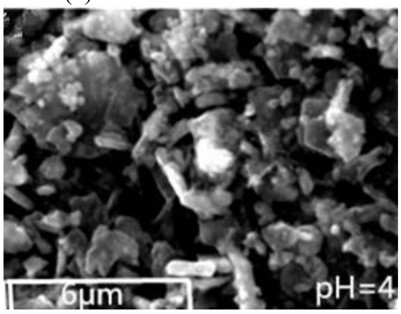
(a)



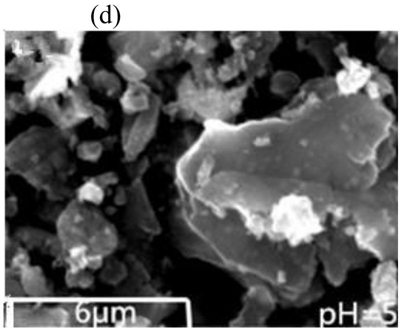
(b)



(c)



(d)



(e)

Figure 2. Fig .(a) SEM of alumina under pH=1, (b) SEM of alumina under pH=2, (c) SEM of alumina under pH=3, (d) SEM of alumina under pH=4, (e) SEM of alumina under pH=5

As shown in Fig .2the diameter of alumina crystal grain increase the pH and dispersion of particle is weakened gradually. Under pH=1,alumina particles is small and granular(figure(a)). Under pH=2, alumina particles present plate as shown in figure (b), crystals of alumina is granular as shown in figure (c) under pH=3, particles of alumina appear together, dispersion decreased as shown in figure (d), in the pH=5 alumina appear obvious reunion phenomenon as shown in figure (e). This is mainly due to the formation of alumina, caused by the different nature of ammonium aluminum sulfate crystal under pH<5, particle precipitation in the form of tiny grain as shown in Fig .1(a). Under pH>5, crystal precipitation is in the form of floc as shown in Fig .1 (b).

C EDS analysis of the elements of alumina under different pH

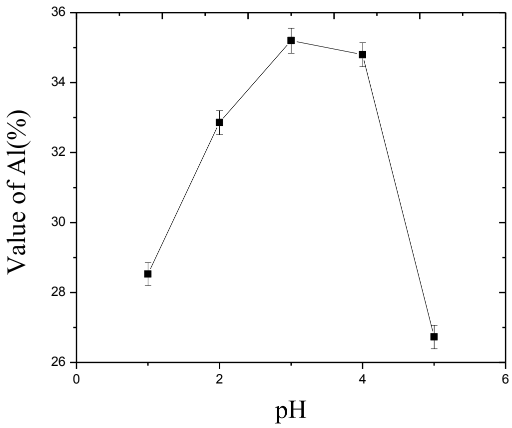


Figure 3. weight percentage of Al in alumina change with pH

Fig .3 is weight percentage of Al change with pH in EDS of alumina. As can be seen from the Fig .3, with the increase of pH, weight percentage of Al in alumina increased first and decreased again, under pH=3, Al reached the maximum weight percentage.

TABLE I. WEIGHT PERCENTAGE OF AL N ALUMINA UNDER DIFFERENT PH

pH	weight percentage of Al %
1	28.53
2	32.86
3	35.2
4	34.8
5	26.73

Table 1 is weight percentage of Al n alumina under different pH obtained from EDS. Weight percentage of Al is 35.2% reached the maximum under pH = 3.

D AAS analyze content of Fe in alumina under different pH

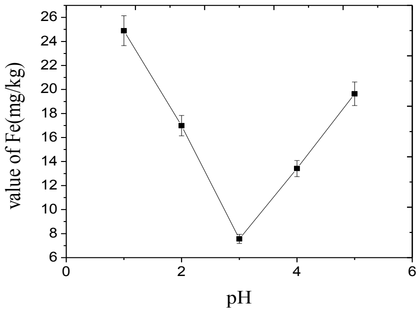


Figure 4. content of Fe in Al2O3 under different pH

AAS analyze content of Fe in alumina under different pH. Content of Fe changes with pH is shown in Fig .4. With the increase of pH, the impurities of Fe content decreased first and increased after, content of impurity have been the least in alumina under pH=3 after high temperature sintering

TABLE II. VALUE OF FE IN ALUMINA UNDER DIFFERENT PH

pH	value of Fe (mg/kg)
1	24.89
2	16.99
3	7.56
4	13.42
5	19.64

From table 2 can conclude that the impurity under pH=3, value of Fe is at least 7.56 mg/kg, this is mainly because under pH<3, ammonium aluminum sulfate crystal precipitation in the form of particles, shape of grain. With the increase of pH, precipitation of the grain size decreases as shown in Fig .1 (c) and (d), the Fe is not easy to be wrapped in the grain size of precipitation. When the pH>3 began to have a small amount of flocculent crystals in a small amount of precipitation, and with the increase of pH, flocculent crystal increase until when pH>5, all take the form of flocculent crystals as shown in Fig .1(b), because of flocculent crystal has good adsorption, so then Fe which wrapped in flocculent crystals also will increase.

E XRD characterization of alumina under different pH

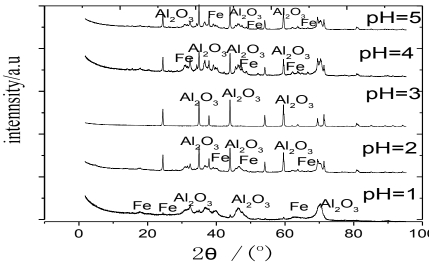


Figure 5. XRD of Al2O3 under different pH

Fig .4 is XRD diagrams of the alumina under different pH. The precursor is calcined at 1200℃, and preserved 2h, the 1 type of crystal is Al_20_3. Through the contrast can be seen that, in the pH=1, diffraction peak of the XRD is ferrosilicon aluminum, so impurity is more. In the pH=4 and pH=5, diffraction peak of Fe appear, so content of the impurity is more. In the pH=3, the intensity of the diffraction peak is the largest, only diffraction peak of Al_20_3 appear, so content of impurity is minimum.

F TG/DSC analysis of precursor under different pH

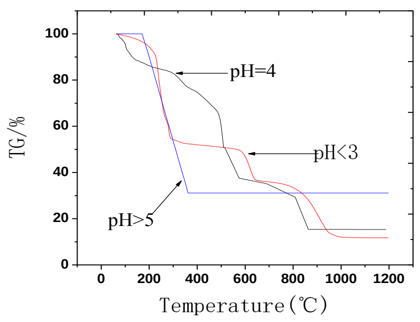


Figure 6. TG curves of precursor under different pH

Use thermo gravimetric analyzer analyze the thermal decomposition of ammonium aluminum sulfate crystal, the result is shown in Fig .6, under pH<3, ammonium aluminum sulfate crystal had obvious three weightlessness, when pH = 4, ammonium aluminum sulfate crystal weightlessness process for three times, but relative to the pH<3 is not particularly evident, under pH>5, only one process of weight loss.

This is because the pH<3 when thermal decomposition of ammonium aluminum sulfate crystals without water of crystallization of three weightlessness of ammonia and desulfurization process, as shown in equation (1) :

$$NH_4Al(SO_4)_2 \cdot 12H_2O \longrightarrow Al_2O_3 + NH_3 + SO_2 + H_2O \quad (1)$$

Under pH>5, precursor only experience one process of weightlessness, may be decomposition of aluminum hydroxide, weightlessness reaction at about 200℃. Under pH=4, the TG/DSC curve of precursor, peak shape is chaos, but basically accord with ammonium aluminum sulfate pyrolysis process. May be due to the differential thermal analysis results ammonium aluminum sulfate crystal precipitation in the form of grain as shown in Fig .1 (a) under pH<5, flocculent crystals as shown in Fig .1(b) under pH>5, two different crystal forms make crystals appear different conditions of pyrolysis.

IV. CONCLUSION

Fe content of alumina increases decreases first and increase after with pH, under pH=3, impurities of Fe content in the minimum is 7.56 mg/kg, alumina crystals for granular and dispersivity is good, stable after 1200 degrees of calcined, XRD results showed that only diffraction peak of Al203.

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