PREPARATION OF ULTRAFINE Sb_2O_3-Sb_2O_5 COMPOSITE POWDER [1]

Duan, Xuechen Zhao, Tiancong
Department of Nonferrous Metallurgy,
Central South University of Technology, Changsha 410083

ABSTRACT

Ultrafine Sb_2O_3-Sb_2O_5 composite powder was prepared by the ammonolysis method of metal alcoxides(0.008μm). The technique of preparing powder and properties of the product were studied. This is a method with simple equipment, low lost and fine powder of uniform size.

Key words: metal alcoholate ultrafine powder composite powder antimony oxide

1 INTRODUCTION

Ultrafine Sb_2O_3-Sb_2O_5 composite powder can be used as increasing effect agent of retard burn of artificial fiber, resin and plastic as well as catalyst, and used in pigment and fine engineering material and so on. Particle size is an important index of antimony oxide products. When they are used in artificial fiber to retard burn, the finer the particle size, the less the used amount, and the lower the cost. If 0.0$x$um grade of Sb_2O_3 is used, the used amount is 1/3 less than that of ordinary grain grade, but effect is same and grains can not clog jetting fiber holes[1].

Used in engineering material, ultrafine antimony oxide can produce particular results because the particle size is fine so that it has large surface and volume effects. Ultrafine Sb_2O_3-Sb_2O_5 composite powder not only has fine particle size, but simultaneously contains Sb(Ⅲ) and Sb(Ⅴ) so that it has binary properties. It is worth developing a deep-product of antimony.

Liquid phase method has the advantage of mixture uniform. Now it has been mainly used to prepare composite powder of various metal oxides in China and other countries. Generally, ultrafine powders of single antimony oxide were prepared. The ultrafine powder of Sb_2O_3 was prepared mainly using plasma body method and gel method[2,3] The colloidal sol of Sb_2O_5 was prepared by organic reagent scatter method[5,6]. This paper tried to develop an ammonolysis method of metal alcoxides to prepare ultrafine Sb_2O_3-Sb_2O_5 composite powder.

2 EXPERIMENTAL

The starting reagents were analytical pure-grade $SbCl_3$, $SbCl_5$, isopropanol, benzene and ammonia water. Distilled water was obtained by two-time distilling.

Fig. 1 shows the flowsheet of preparation of ultrafine Sb_2O_3-Sb_2O_5 composite powder. In experiment, the starting agent $SbCl_3$ and $SbCl_5$ were to be reacted respectively with isopropanol containing benzene. In order to react throughout the solution was refluxed at boiling temperature to obtained Sb(Ⅲ) alcoholate and Sb(Ⅴ) alcoholate solution respectively. Then the two kinds of solution obtained were mixed on different proportions and reacted further. At certain temperature the mixed solution reacted and $NH_3 \cdot H_2O$ solution containing cation surface-active agent reacted to obtain precipitate. The reaction solution was filtered and pricipitate was washed. The precipitate was dried to obtain ul-

① Received Jun. 29, 1993

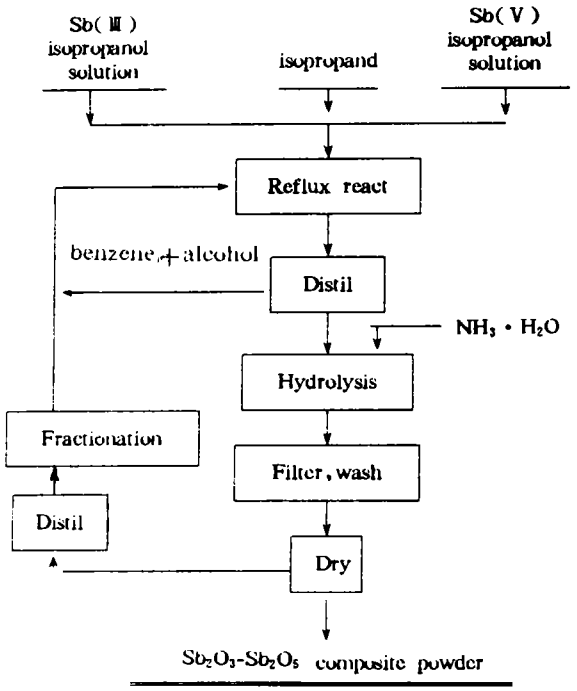

Fig. 1 Flowsheet of preparation of Sb_2O_3-Sb_2O_5 composite powder

trafine Sb_2O_3-Sb_2O_5 composite powder.

The analytical procedures included analyses of H-800 electron microscope for size measurement of particle, PW-1404 X-ray diffractometer for crystal form, PEB ultrared spectrometer for observing the compositional change of the composite powder, BET mehtod for ratio-surface area and difference-thermal method for identifying thermal absorption property.

3 RESULTS AND DISCUSSION

3.1 *Preparation of Ultrafine* Sb_2O_3-Sb_2O_5 *Composite Powder*

The experimental results are listed in Table 1. The experiments show that alcholation effect of Sb (Ⅲ) and Sb (Ⅴ) is relative with alcoholization reagent and experimental conditions. The higher the reaction temperature and the longer the reaction time, the better the alcoholization effect.

From Table 1, experimental results show that when ultrafine compostie powder is prepared in different proportions of $SbCl_3$ and $SbCl_5$, average particle size diminishs with increasing proportion of $SbCl_5$. When the proportion of $SbCl_3$: $SbCl_5$ is 1 : 3, the average size is the smallest and about 0.008 μm. Fig. 2 is its photograph gained by electron microscope. From Fig. 2, the image of particle also can be observed.

The formation of Sb_2O_3-Sb_2O_5 composite powder is relative with hydrolysis conditions. Experiments show that appropriate hydrolysis temperature is 60～70 ℃, starting acidity is pH2 and ending point acidity is about pH8. In order to control the acidity of hydrolysis, appropriate amount $NH_3 \cdot H_2O$ was added. When cation surface actived agent was added into hydrolysis solution, it can change ratio surface energy of the powder surface and be used as protectant and washing agent of the powder so that not only average size is small, but also this can ensure less aggregation of the powder in long-time store. On the other hand, absorbed impurity of the product is little and purity is high.

3.2 *Ultrared Spectrum Experiments*

Table 1 Experimental results of preparation of Sb_2O_3-Sb_2O_5 composite powder

$SbCl_3$: $SbCl_5$ (wieght ratio)	Hydrolysis temperature / C	Acidity of hydrolysis (pH)	Ending point acidity (pH)	Average particle size /μm	Remarks
1 : 0	60—70	2—4	7—8	0.015	
3 : 1	60—70	2—4	7—8	0.033	
2 : 1	60—70	2—4	7—8	0.017	
1 : 1	60—70	2—4	7—8	0.015	
1 : 2	60—70	2—4	7—8	0.010	dense
1 : 3	60—70	2—4	7—8	0.008	dense
0 : 1	60—70	2—4	7—8	0.013	dense

For understanding ultrared spectrum property and composition of ultrafine Sb_2O_3- Sb_2O_5 composite powder, ultrared spectrum were taken. In order to understand compositional change the ultrared spectra of Sb_2O_3 and Sb_2O_5 also were taken. Figs. 3, 4 are the ultrared spectra of Sb_2O_3 and Sb_2O_5 respectively.

Comparing the ultrared spectrum of the composite powder with that of Sb_2O_3 and Sb_2O_5, the form of ultrared spectrum of Sb_2O_3 Sb_2O_5 powder is similar to that of Sb_2O_3. But both are not same. Fig. 5 is ultrared spectrum of ultrafine Sb_2O_3-Sb_2O_5 composite powder. This explains that Sb_2O_3-Sb_2O_5 composite powder is a mixture of Sb_2O_3 and Sb_2O_5. Perhaps its composition has some changes.

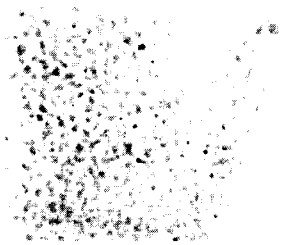

Fig. 2 Photograph of electron microscope of Sb_2O_3-Sb_2O_5 composite powder

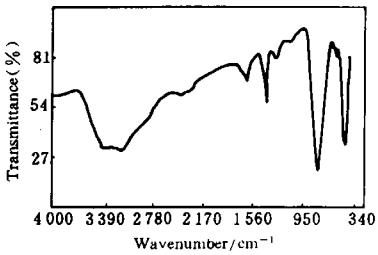

Fig. 3 Ultrared spectrum of Sb_2O_3 powder

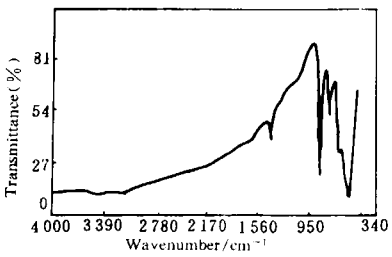

Fig. 4 Ultrared spectrum of Sb_2O_5 powder

3.3 *Properties of Composite Powder*

In order to understand thermal stability, crystal form and specific surface area of ultrafine Sb_2O_3-Sb_2O_5 composite powder, these properties were taken. Fig. 6 is its high temperature differential thermal analysis pattern. Fig. 7 is its X-ray diffraction pattern. The specific surface area of the composite powder was taken by BET method.

Fig. 6 shows that Sb_2O_3-Sb_2O_5 composite powder has two strong absorption peaks at 442.53 ℃ and 486.4 ℃. Because drying temperature is low (at about 120 ℃), there is still thermal absorption

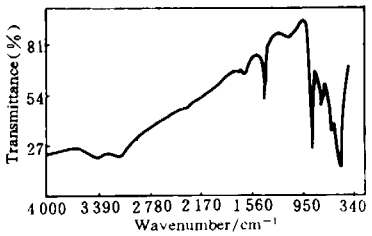

Fig. 5 Ultrared spectrum of Sb_2O_3-Sb_2O_5 composite powder

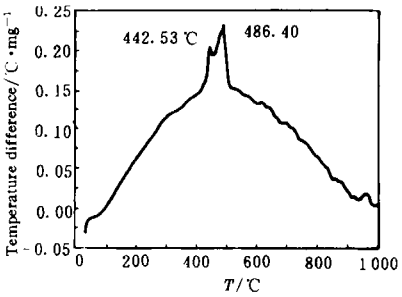

Fig. 6 High temperature differential thermal analysis pattern of Sb_2O_3-Sb_2O_5 composite powder

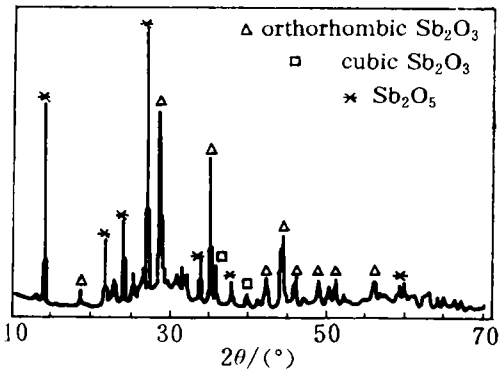

Fig. 7 X-ray diffraction pattern of Sb_2O_3-Sb_2O_5 composite powder

below 440 ℃. Fig. 7 shows that Sb_2O_3 in the com posite powder is mainly orthorhombic form and there is a few cubic form. Sb_2O_5 also is orthorhombic form. Therefore, we think that the composite powder is mainly made of orthorhombic form. From this, we think further that the composite powder is a mixture of Sb_2O_3 and Sb_2O_5 powder. specific surface area of the composite powder is about 7.42 m^2/g. Its specific surface area is very large. This shows that the product is very fine. On the other hand, the composite powder can be used as catalyst etc.

4 CONCLUSION

The technique for preparing ultrafine composite powder was studied. Under appropriate conditions Sb(Ⅲ) and Sb(Ⅴ) chloride are alcoholized, then mixed and ammonolised. Cation surface reactive agent was added into $NH_3 \cdot H_2O$ solution. The starting acidity of hydrolysis is pH2～3 and ending acidity is pH8 to obtain ultrafine composite powder. The powder is fine with uniform particle size and has good transparence. This technology has many advantage: simple in preparing equiment, low cost and good properties of the product.

Some properties of the composite powder were taken. When the proportion of $SbCl_3$: $SbCl_5$ is 1 : 3, particle size of the powder is 0.008 um. The composite powder is orthorhombic form. Its specific surface area is 7.42 m^2/g. The composite powder has two strong absorption peaks at 442.5 ℃ and 486.4 ℃. It is a mixture of Sb_2O_3 and Sb_2O_5.

REFERENCES

1 Li, Reilin. The particle size of antimony system retard burn reagents. Hunan Non-ferrous Metal Information net. 1987. 4.

2 Chang, Pengbei. In: Symposium of Ultrafine Grain and Surface Science in China, Changsha, 1988. 6.

3 Zhu, Zhaosen. Metallurgy of Heavy Metals, 1987, 3: 22.

4 Duan, Xuechen; Zhang, Duomo; Zhao, Tiancong. Journal of Central South Institute of Mining and Metallurgy, 1990, 5: 485.

5 Richardson, John G. US4341655. 1982.

6 渡部淑胤, 铃木启太郎, 寺西雅幸. JP 6041536. 1985.