Solid State Amorphizing Reaction of $\text{Al}_{30}\text{Ta}_{70}$ Alloy Powders by the Rod Milling Technique

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M. Sherif EL-ESKANDARANY, K. AOKI and K. SUZUKI: Solid State Amorphizing Reaction of $\text{Al}_{30}\text{Ta}_{70}$ Alloy Powders by the Rod Milling Technique.

A rod-milling technique has been investigated for producing $\text{Al}_{30}\text{Ta}_{70}$ amorphous alloy powders. To follow the amorphization reaction, the rod-milled powders were characterized by X-ray diffraction, differential thermal analysis, optical microscopy, scanning electron microscopy, electron probe microanalysis and chemical analysis. The results obtained by rod-milling were compared with those from a conventional ball-milling technique. The rod-milled $\text{Al}_{30}\text{Ta}_{70}$ amorphous alloy powders have high amorphization rate, high thermal stability and a low contamination content.

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I. Introduction

Amorphous metallic alloys have been traditionally prepared by solidification from melts, condensation from vapors and precipitation from solutions. These conventional processes are based on removing rapidly the kinetic energy of atoms in the liquid and gas phases⁽¹⁾. Currently, great interests are focused on the solid state amorphization process⁽²⁾. Mechanical alloying (MA) using ball-milling (BM) technique for processing amorphous alloy powders was first demonstrated by Koch⁽³⁾. So far BM technique has been commonly used for producing amorphous alloys such as Al-Ta⁽⁴⁾ and Cu-Ta⁽⁵⁾ binary systems that are impossible to produce by the traditional methods.

In fact, almost all ball-milled alloy powders are contaminated with iron, when stainless steel balls are used as milling media. Therefore MA techniques must be puzzled by this serious problem which makes the progress rather difficult. Since the ball-powder-ball collision in a ball mill is considered as the main source of iron contamination, we have used rod-milling (RM) technique in which there is no collision between its milling media.

The present work attempts to provide a unique MA process for producing amorphous alloy powders. The main benefits in RM process are high amorphization rate, high thermal stability and low iron contamination.

II. Experimental procedures

In this study both RM and BM processes were carried out for producing $\text{Al}_{30}\text{Ta}_{70}$ amorphous alloy powders. The rod-mill can be described as a horizontally rotating stainless steel cylindrical shell in which stainless steel rods are used as milling media. Pure elemental powders of

Al and Ta were mixed to give the desired composition in a glove box and sealed in the RM under purified argon atmosphere. The milling was carried out as described in a previous paper⁽⁶⁾. The direct observations of the rods movement inside the shell have shown that the milling occurs with the shear force induced by the line contact of rod-powder-rod extending the full length of the shell. The BM technique used in this study is essentially same to that described in previous papers^{(4),(7)}. The RM and the BM alloy powders were characterized by X-ray diffraction (XRD) with $\text{MoK}\alpha$, differential thermal analysis (DTA), optical microscopy, scanning electron microscopy (SEM), electron probe microanalysis (EPMA) and chemical analysis using ICP emission method.

III. Results and discussion

Figure 1 shows optical micrographs for polished $\text{Al}_{30}\text{Ta}_{70}$ alloy powders after 4(a), 11(b), 22(c) and 43(d) ks of RM time. As shown, the morphology of the powder particles changes with increasing the RM time. After 4 ks of milling time, the particles have a flake-like morphology with irregularly layered structure. As the RM time increases, the thickness of each layer becomes narrow and uniform. At the stage of 43 ks of milling time, the layered structure is quite homogeneous and the direction of these layers has almost the same trend of arrangements. This suggests the great effect of shear force on the rod milling process.

Figure 2 shows the effect of the RM (closed symbols) and BM (open symbols) time on the particle size of $\text{Al}_{30}\text{Ta}_{70}$ alloy powders. As shown in Fig. 2, the milling process can be classified into three stages. At the initial stage, which is called the agglomeration stage, the starting elemental powders of Al and Ta

Fig.1 Optical micrographs of $\text{Al}_{30}\text{Ta}_{70}$ powder particles as a function of RM time.

Fig.2 Particle size of $\text{Al}_{30}\text{Ta}_{70}$ alloy powders as a function of RM and BM time.

agglomerate at this early stage of milling (4 to 22 ks) to form powders of greater diameter, as large as several hundred μm . During this stage, the characteristic structure containing Al and Ta layers appears, as had been shown in Fig.1. In the middle stage corresponding to the RM time from 43 to 360 ks, the solid state amorphization reaction proceeds drastically by the atomic diffusion through the interface between the layers and the powder particles are subjected to continuous disintegration until the particle size is less than 50 μm , as shown

in Fig. 2. During this stage the layered structure disappears gradually to be hardly visible after 173 ks of RM time, as shown in Fig. 3(a). However, in BM, the layered structure can be seen easily even after 173 ks of milling time, as shown in Fig. 3(b). As illustrated in Fig. 2, the average diameter of the RM powder particles is small and subsequent disintegration into fine powders proceeds at a high rate to provide a narrow size distribution. In contrast, the milling process of BM occurs at low rate with a wide size distribution.

During the last stage of milling, which is called homogenization stage (720 to 1440 ks), the amorphous structure homogenized is and the particle size is nearly constant, as shown in Fig. 2. Moreover, the shape of both RM and BM particles is changed to be almost spherical after 1440 ks of RM and BM time, as shown in Figs. 3(c) and 3(d), respectively.

Figure 4 shows the XRD patterns of $\text{Al}_{30}\text{Ta}_{70}$ alloy powders as a function of RM time. The XRD patterns of BM $\text{Al}_{30}\text{Ta}_{70}$ alloy powders is presented in a previous paper⁽⁶⁾. As shown in Fig. 4, the intensities of the Bragg peaks for pure Al and Ta crystals decrease quickly as the RM time increases. As illustrated, after 360 ks of RM time a large decreasing of the crystalline peaks for Al and Ta are observed. A single amorphous phase was

Fig.3 Scanning electron micrographs of $\text{Al}_{30}\text{Ta}_{70}$ alloy powders as a function of RM and BM time.

Fig.4 XRD patterns of rod-milled $\text{Al}_{30}\text{Ta}_{70}$ alloy powders as a function of milling time.

Fig.5 Iron contamination content in $\text{Al}_{30}\text{Ta}_{70}$ alloy powders as a function of milling technique, milling time and repetition of milling process.

formed, characterized by broad and smooth peak after 1440 ks of RM time, as shown in Fig. 4.

Figure 5 illustrates the iron contamination content in mechanically alloyed $\text{Al}_{30}\text{Ta}_{70}$ powders as a function of RM (a) and BM (b) time, and the repetition of milling process. At the starting of the first milling run, the milling media were used in the absence of the powders coating. The milling media which had been coated after the end of the first milling run, the milling were used again in the second and the third runs. A drastic decreasing of the iron contamination content from the first to the third runs are observed. It is also shown that the amount of iron contamination in the RM alloy powders is lower than that in the BM one.

Figure 6 shows (a) the crystallization enthalpy change; ΔH_x and (b) the crystallization temperature; T_x as a function of RM (closed symbols) and BM (open symbols) time. As shown in Fig. 6(a), the value of ΔH_x decreases sharply during the amorphization stage of

milling, suggesting a drastic change to the amorphous alloy state. At the homogenization stage, the value of ΔH_x approaches a saturation state.

As shown in Fig. 6(b), the T_x increases drastically in the initial and the middle stages of milling suggesting that the alloying and amorphization occur simultaneously and inhomogeneously. Through the last stage of milling, the T_x approaches a saturation state, as shown in Fig. 6(b). It is also shown that the T_x of RM amorphous alloy powders is higher than that of BM one. This is due to the low contamination of iron and homogeneity in the former.

IV. Conclusions

In this study we conclude that the *rod-milling* is a successful method for producing $\text{Al}_{30}\text{Ta}_{70}$ amorphous alloy powders with a *high amorphization rate* and *high thermal stability*. The *low iron contamination content* in the rod-milled alloy powders is the main benefits of this process.

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Fig.6 Crystallization enthalpy change, ΔH_x , and crystallization temperature, T_x , of $\text{Al}_{30}\text{Ta}_{70}$ alloy powders as a function of RM and BM time.