

STRUCTURE CHANGES OF Al/Pb AND Ag/Pb POWDER MIXTURES DURING MECHANICAL ALLOYING

Teruo OHASHI and Hiroshi HOSOE

Dep. Mater. Sci., Nagoya Institute of Technology
Gokiso-cho, Showa-ku, Nagoya, Japan 466

ABSTRACTS Mechanical alloying(MA) effectiveness of Pb in aluminum and silver was examined focusing our attention on the structure changes of their powder mixture during milling. The sufficiently deformed and pulverized powders were obtained after a milling time of about 10 hrs and composed substantially from the crystallites the diameters of which was about 40 nm for the Al/Pb and 20 nm for the Ag/Pb mixtures. Dissolving of Pb into the base metals could be recognized in the Ag/Pb mixture but not in the Al/Pb mixture. The formation of the stacking fault was confirmed in Ag by XRD. The results of DSC and XRD showed that the stored energy in the Ag/Pb mixture by milling increased drastically with milling time up to about 10 hrs; however, an exception was made for the case of the Al/Pb mixture.

Keywords: powder, Al-Pb alloy, Ag-Pb alloy, crystallite size, stacking fault

1. INTRODUCTION

Al-Pb alloy has a monotectic reaction at 658°C where one of the liquids solidify as an Al rich phase and the remaining liquid enriched in Pb solidifies as a nearly pure Pb phase at the eutectic reaction temperature of 327°C. Al-Pb phase diagram is very similar to that of Cu-Pb alloy in which Pb performs in the lubrication ability. If Pb phase is more finely distributed in the matrix, the load capacity of this bearing alloy can be increased [1]. In order to realize such a refinement of the Pb phase, the mechanical grinding (MG) has been applied on the Cu-Pb powder alloy and brought about the sufficiently pulverized structure composed from the crystallites of about 50nm in diameter [2], [3]. The resultant redistribution of Pb in the MG Cu-Pb powder alloy could be so accepted that the sintered compacts of this powder alloy improved wear resistance [4].

Looking around the above experimental results, it is further of interest to reveal the structure changes by the mechanical milling of the other metal systems which have an effective solid solubility in the matrix and/or are composed of all of the soft elements. Thus, in the present study, the Ag/Pb powder mixture for the former and the Al/Pb powder mixture for the latter have been taken up and their structure changes by the mechanical milling have been examined; hereafter, the mechanical milling of the powder mixture composed of elemental metals is called MA (mechanical alloying).

2. EXPERIMENTAL PROCEDURE

Materials used for MA processes were the metal powders of atomized pure Ag(99mass% up), pure Pb(99.9mass%), and pure Al(99.9mass%) powders the average diameters of which were about 30 to 50 μ m. The compositions of these metal powder mixtures are 30 mass%Pb for Al/Pb system and 10, 20, 30 mass%Pb for Ag/Pb system. The mechanical milling of these powder mixtures was carried out by using a planetary micro-mill (Fritsch P7) under the steel balls' acceleration of 5G for a maximum period of 50 h in nitrogen gas atmosphere. These milled (MA) powders were subjected to X-ray diffraction (CuK- α and K- β without any filter to intensify the radiation) and differential scanning calorimetry (DSC) analyses, and scanning electron (SEM) and transmission electron (TEM) microscopy observations.

3. RESULTS AND DISCUSSION

Deformation and synthesis of the metal powders should proceed during MA and result in the change in X-ray diffraction profiles of these powders as shown in Fig. 1. It can be seen that both of the X-ray diffraction peaks obtained from the Al/Pb and Ag/Pb powder mixtures become broader and lower with milling time. However, some peak-shift of these reflections can be recognized, especially in the profiles of the Ag/Pb mixtures. Fig.2 shows SEM microstructures and the characteristic X-ray images of the Al/Pb and Ag/Pb powder mixtures ball-milled for 50h. It can be recognized that both of the MA powder mixtures are microscopically homogeneous. Such a structure has been attained in a MA powder ball-milled for more than 10h. From each of the X-ray diffraction profiles, the crystallite sizes, the lattice constants, and the lattice strains have been calculated and the results are shown in Fig. 3 to Fig.5. The crystallite size of the each constituent of the powder mixtures has been calculated by using the Hall and Williamson's relation [5]. It decreases fast in the initial stage of MA processing and approaches a definite value over a milling time of 20h; i.e., about 40nm for Al and Pb in their mixture and 20nm for Ag and Pb in their mixture (Fig.3). Change in the lattice constant of the constituents with the milling time can be recognized only for Ag (Fig.4). Its lattice constant increases with the milling time abruptly in the initial stage of MA. If this lattice constant vs. Pb content obeys the Vegard's law, the amount of the dissolved Pb into Ag can be estimated to be at least about 1 at% that is 25% excess of the equilibrium maximum solid solubility. Such a dissolving of Pb into Ag at the solid state has been caused really by MA.

On the other hand, such a dissolving phenomenon can hardly be seen in the Al/Pb mixture ball-milled for a long time. Thus, as would be expected, it is said that MA of the elemental metal mixtures the phase diagram of which has a solid solubility in the either element facilitates the dissolving phenomenon. The lattice strain has been also estimated by using the Hall and Williamson's relation [5] and its result is shown in Fig.5. An obvious increase in the lattice strain can be seen in the Ag/Pb mixtures but not in the Al/Pb mixtures. The low stacking fault energy allows to extend the dislocation and Ag is this case [6]. The cross-slip by its extended dislocations needs a high energy and thus, mobile dislocations pile up at their crossing points and result in the increase of their density, i.e., the lattice strain.

Another consideration could be given to the above mentioned MA effects on the two kinds of the metal powder mixtures by measuring the changes of the difference in diffraction angle $\Delta 2\theta$ between (200) and (111) diffraction peaks with the milling time. It is well known that the stacking fault is formed by the deformation if the stacking fault energy of the metal is low. This energy is 0.04 J/m^2 for Ag and the lowest value among the presently concerned metals [6]. The stacking fault probability α in fcc metal by deformation can be estimated from the values of $\Delta 2\theta$ [7] which are shown as a function of the milling time in Fig.6. $\Delta 2\theta$ obtained from the diffraction peaks of Ag decreases with the milling time but that from those of Al and Pb does not so and keeps a nearly constant value through all of the milling time. From $\Delta 2\theta$ at the milling time of 50h for the Ag/Pb mixture, α is about 0.04 and comparable to that of the filed brass powder [8]. When the Ag/Pb powder milled for 50h is annealed at several temperatures, an apparent increase in $\Delta 2\theta$ can be seen at the annealing temperature above about 50 to 100°C and approaches to the value of a strain-free Ag, as shown in Fig.7. On the contrary, the half-value width of (111) peak of Ag decreases with annealing temperature because of the release of the lattice strains including dislocations introduced by MA and the crystal grain growth.

An excess energy introduced to the MA powders can be also estimated by means of DSC measurement. Fig.8 shows some thermograms of Al/Pb and Ag/Pb MA powders, measured in a N_2 gas atmosphere, as a parameter of the milling times. The lowest heat release is in the as-atomized powder mixtures and a sharp endothermic peak due to the fusion (eutectic reaction in an exact expression [9]) of Pb at 326°C for the Al/Pb and 304°C for the Ag/Pb powder mixture can be seen there. Increasing the ball-milling times, a broad exothermic peak can be recognized at around 170 and 150°C for the Al/Pb and Ag/Pb powder mixtures, respectively. These released energies might have been kept in the forms of the lattice defects or strains, and/or the increased grain boundaries as well.

powder. These energies have been estimated by the difference between the each integrated broad peak and the respective base line drawn on the foots of the both sides of the peak . Fig. 9 shows these released energies as a function of the milling time. A steep increase of the released energy can be seen within about 10h and is about 40 to 50J/mol. The large angle grain boundary energy of Ag is 0.4J/m^2 [10] and, if one mole of Ag is constructed of the dodecahedron grains of 200nm in diameter, the grain boundary energy/mole becomes 53J/mol. This value is nearly the same as the released energies obtained in the above DSC measurements. On the contrary, the fact that the released energy of Al/Pb powder mixture is very small may reflect the large crystallite sizes (Fig.2) and the small lattice strains (Fig.4). Fig.10 shows the TEM structure of the Al/Pb powder mixture which was ball-milled for 50h and heated to 500°C at a rate of 5°C/min. It is interesting that there are many polyhedral Pb particles the diameter of which is more or less than 100nm (indicated by an arrow in the figure). These Pb particles are not only concentrated in the Al grain boundaries but also distributed in the Al grains. Pb particle like these can be seen in a rapidly solidified Al-Pb alloy where the Pb particles crystallize at the eutectic reaction temperature of 327°C [9] keeping a crystallographic coherency with the Al matrix [11]. Comparing these results of the rapidly solidified Al-Pb alloy with the present one, it is suggested that the solid Pb powders are enclosed with the pulverized solid Al grains by repeated pulverizing and welding events during MA processing. .

4. CONCLUSIONS

The results obtained are as follows;

(1) The sufficiently deformed and pulverized powders were obtained after a milling time of about 10 hrs and composed substantially from the crystallites the diameters of which was about 40 nm for the Al/Pb and 20 nm for the Ag/Pb mixtures.

(2) Dissolving of Pb into the base metals could be recognized in the Ag/Pb mixture the alloy of which has intrinscally a finite solubility of Pb, but does not that of the Al/Pb mixture.

(3) The formation of the stacking fault was confirmed in Ag by peak shifts of (111)Ag and (200)Ag reflections of XRD.

(4) The results of XRD and DSC measurements showed that the lattice strain and the stored energy in the Ag/Pb mixture by MA increased drastically with milling time up to about 10 hrs; however, an exception was made for the case of the Al/Pb mixture.

(5) TEM structure of the MA Al/Pb powder suggests that the solid Pb powders are enclosed with the pulverized solid Al grains during MA processing.

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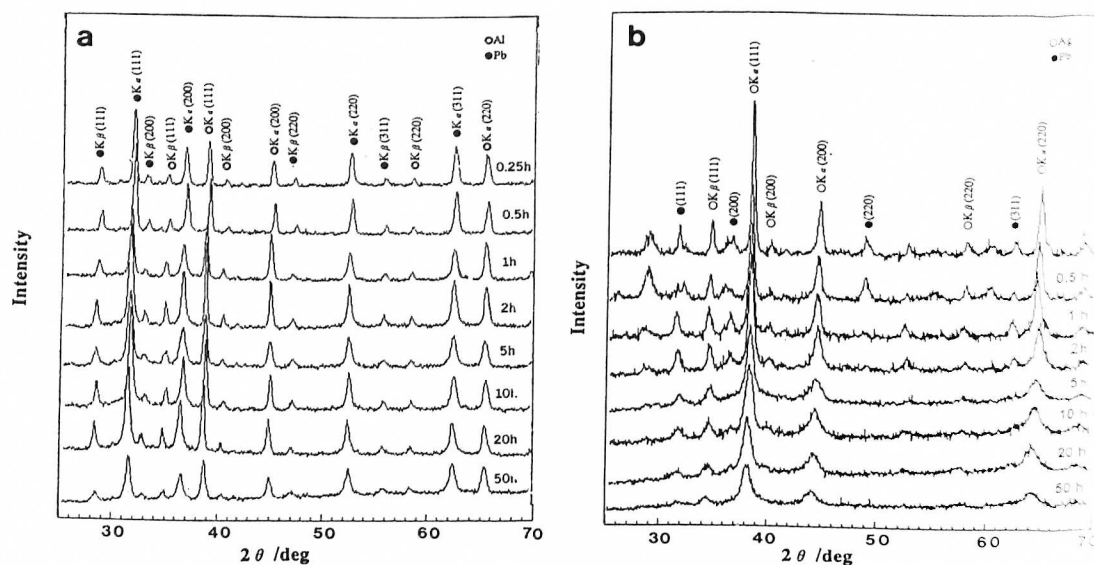


Fig.1 X-ray diffraction profiles of the Al/Pb (a) and Ag/Pb (b) powder mixtures ball-milled for several hours (Cu-K α radiation).

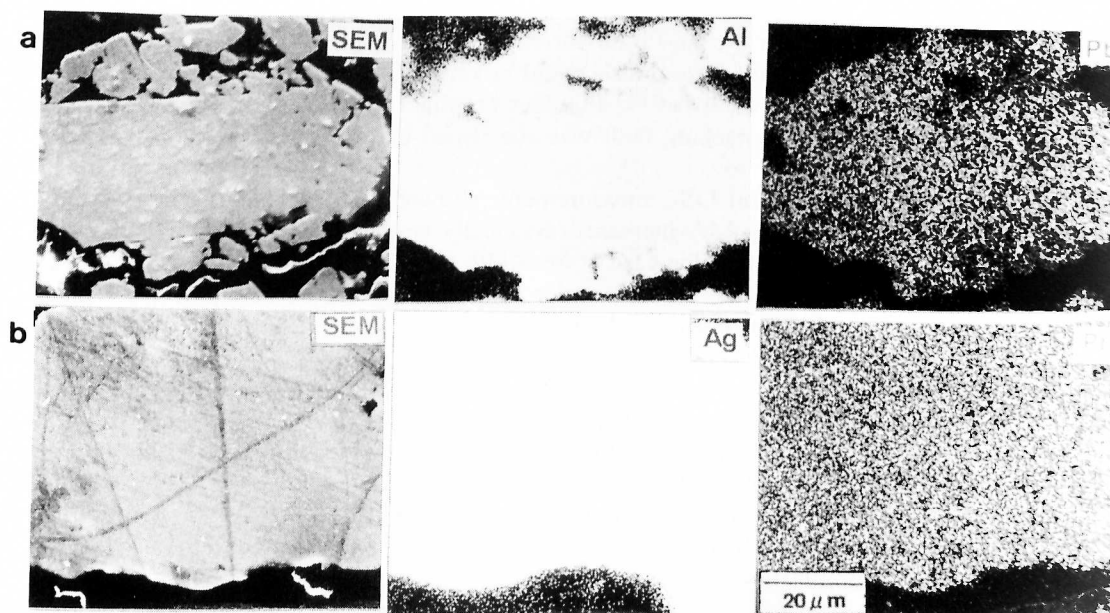


Fig.2 SEM structures and X-ray characteristic images of the Al/Pb (a) and Ag/Pb (b) powder mixtures ball-milled for 50h.

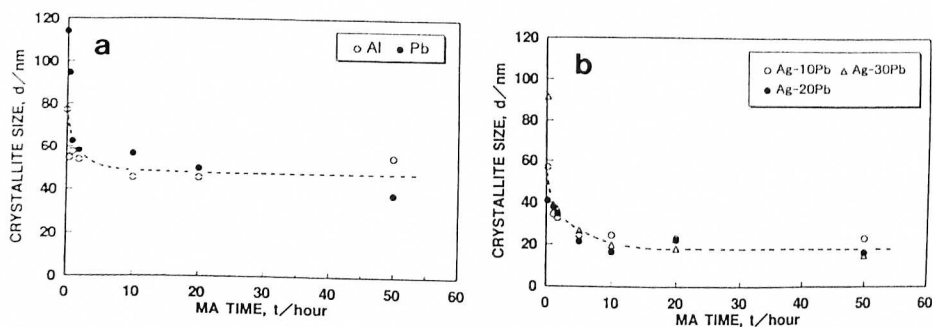


Fig.3 Changes in the crystallite sizes with ball-milling (MA) time; (a) for Al/Pb and (b) for Ag/Pb powder mixtures.

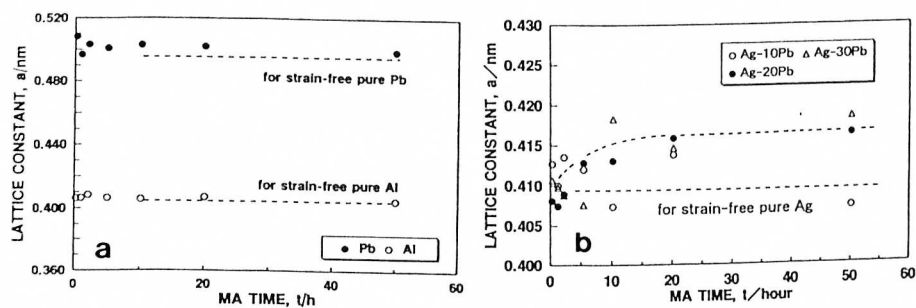


Fig.4 Change in the lattice constant of the Al/Pb (a) and Ag/Pb (b) powder mixtures with ball-milling (MA) time

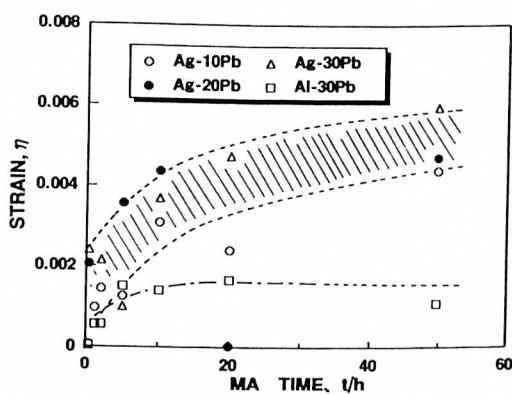


Fig5 Change in the lattice strain of the Al/Pb and Ag/Pb powder mixtures with MA time.

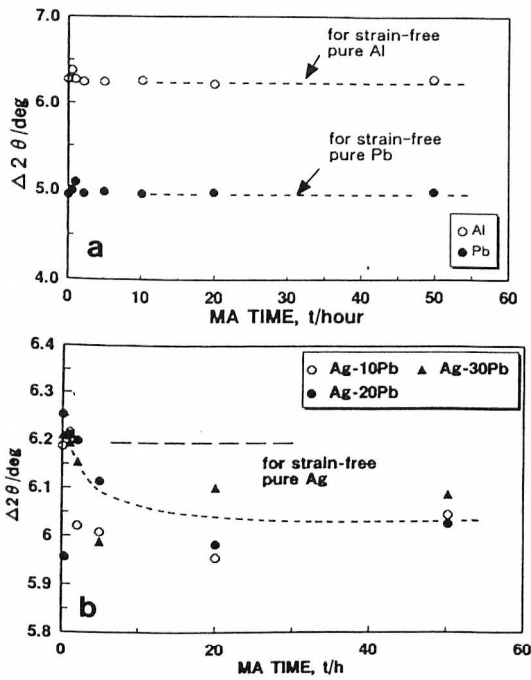


Fig.6 Change in $\Delta 2\theta$ of the Al/Pb (a) and Ag/Pb (b) powder mixtures with MA time.

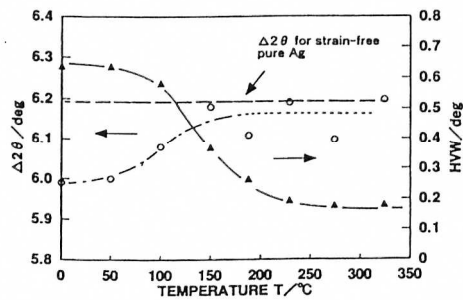


Fig.7 Change in $\Delta 2\theta$ of the Ag/Pb powder mixtures with annealing temperature; ball-milled for 50h.

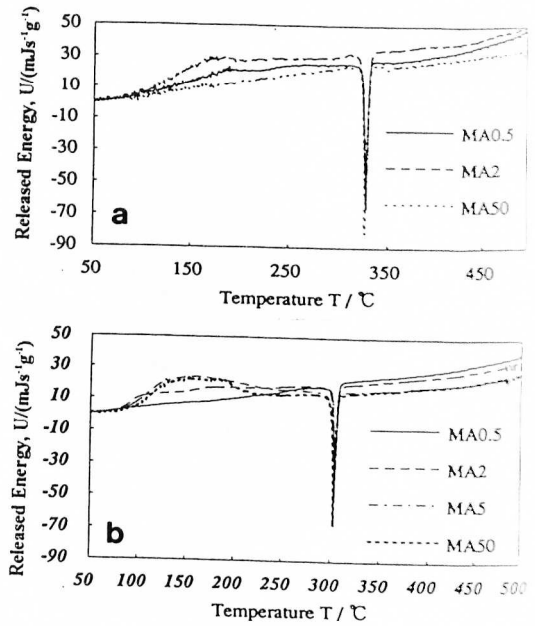


Fig.8 DSC thermograms of the Al/Pb (a) and Ag/Pb (b) powder mixtures ball-milled (MA) for several hours.

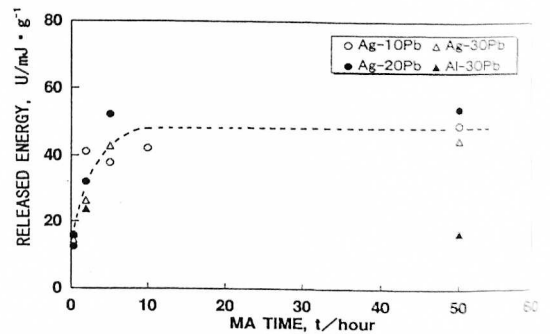


Fig.9 Change in the released energy estimated from the DSC thermograms the MA powders; ball-milled for 50h.

Fig. 10 TEM structure of the Al/Pb powder mixture ball-milled for 50h and subsequently heated to 500°C at a rate of 5°C/min.

