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Electrorecrystallization of Metal Alloy

Ying-Ta Chiu [a,*], Kwang-Lung Lin [a], Albert T. Wu [b], Wei-Luen Jang [c], Chung-Li Dong [c], Yi-Shao Lai [d]

[a] Department of Materials Science and Engineering, National Cheng Kung University, No.1, University Road, Tainan, Taiwan
[b] Department of Chemical and Materials Engineering, National Central University, No. 300, Jhongda Road, Jhongli, Taiwan
[c] National Synchrotron Radiation Research Center, No. 101, Hsin-ann Road, Hsinchu Science Park, Hsinchu, Taiwan
[d] Central Labs, Advanced Semiconductor Engineering, Inc., No. 26, Chin 3rd Road, Nantze Export Processing Zone, Kaohsiung, Taiwan

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ABSTRACT

This work first reports the behavior of electrorecrystallization of metal alloy, solder in this study. The electrorecrystallization is the recrystallization of metal alloy induced by electrical current stressing without the need of prior treatment. The synchrotron radiation X-ray diffraction (XRD) *in situ* analysis shows that the crystal facets of the Pb matrix of 95Pb5Sn solder alloy gradually diminish during electrical current stressing. The XRD analysis incorporating with *in situ* synchrotron extended X-ray absorption for fine structure (EXAFS) reveals complete dissolution, giving rise to supersaturation, of the Sn-rich secondary phase upon current stressing. The supersaturation behavior was explained through the crystal relaxation of the Pb matrix induced by current stressing. The recrystallization gives rise to Sn nanorod. High resolution TEM analysis of the nanorod reveals the semi-coherent/incoherent interfaces and provides explanation for the formation of the Sn nanorod.

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1. Introduction

The diffusion of metal atoms in response to current stressing may generate stress and damage to metal conductor [1–3]. The electrical current passing through a metal conductor will result in the momentum transfer from electron to the metal atoms [4,5]. The momentum transfer may give rise to excess atomic flux, J_{em}, and thus the electromigration of metal atoms [6].

$$J_{em} = C\frac{D}{kT}Z^{*}ej\rho \tag{1}$$

where C is concentration, D is the diffusivity, kT has the usual meaning, Z^{*} is the effective charge number, e is the electron charge, j is the current density and ρ is the resistivity of object. The constituent elements may respond differently to current stressing in a metal alloy. The electron flow drives Sn to diffuse toward the cathode while Pb to the anode [7,8] due to the difference in diffusivity of the elements. Electromigration has been known to cause dissolution of the metallization of a solder joint [9,10]. Fast dissolution of metallization was ascribed to both electromigration and thermomigration [11] induced by current stressing. A temperature gradient of 1000 °C cm^{-1} gives rise to thermomigration in a eutectic SnPb solder joint [12]. In addition to dissolution of the metallization layer, the current stressing also causes reorientation of Sn grain and rotation in the pure Sn conductor [13] caused by the unbalanced torque [14,15]. Grain reorientation has also been observed through EBSD study for the Pb matrix of 95Pb5Sn solder during current stressing [16].

In spite of the various effects mentioned above, there is no report on the electrorecrystalliztion, the recrystallization of metal alloy under current stressing without the need of any prior condition. Very few study investigated the dissolution behavior of the second phase in the matrix of an alloy. The current stressing will cause supersaturation of Sn in Pb matrix of the 95Pb5Sn solder [17]. Nevertheless, the dissolution behavior of the alloy element, for example Sn in 95Pb5Sn solder alloy, is hard to investigate as the elements dissolved will soon precipitate and reach equilibrium status after current stressing. The present study applied synchrotron X-ray diffraction (XRD) and extended X-ray absorption for fine structure (EXAFS) to conduct *in situ* investigation for 95Pb5Sn solder of the electrorecrystallization of Pb matrix and secondary Sn-rich phase. The electrorecrystallization behavior results in crystal reorientation of the Pb matrix and nanorefining of the secondary Sn-rich phase.

2. Experimentals

Commercial 95Pb5(wt.%)Sn and 97Pb3Sn (for bench mark) solder strips, 10 × 2 × 0.025 mm, were put on the Kapton tape and fixed on the Si substrate as shown in Fig. 1. The two ends of solder strip were connected to electrodes with 95Pb5Sn solder alloy. The electron flows from right to left as indicated by the arrow. The mark "X" was the area of X-ray analysis, located at the longitudinal central line and 1.1 mm from the cathode side of the solder strip. The average Pb grain size of the as received strip was 20±5 μm. The solder strip was subjected to 6×10^{3} A cm^{-2} on the stage of synchrotron (NSRRC, National Synchrotron Radiation Research

* Corresponding author.
E-mail address: yingta@mail.mse.ncku.edu.tw (Y.-T. Chiu).

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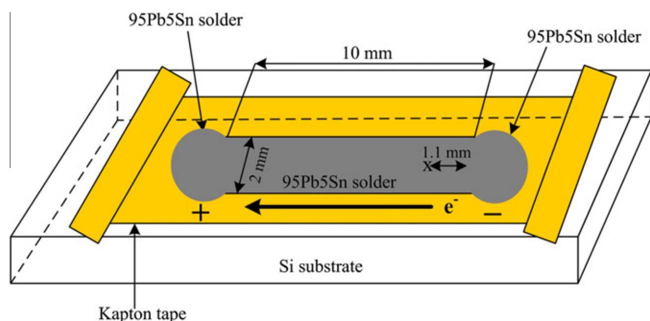

Fig. 1. A schematic of 95Pb5Sn solder strip which is fixed with Kapton tape. The "X" is the area investigated *in situ* with synchrotron radiation X-ray diffraction (XRD) and extended X-ray absorption for fine structure (EXAFS).

Center, Hsinchu, Taiwan) X-ray working station under ambient condition. The XRD and XAS (X-ray absorption spectrum) were conducted at wiggler beamlines BL17B1 and BL01C1 at NSRRC. The XAS spectra were measured using the fluorescence mode in a conventional ionization chamber.

The crystal orientation and the local atomic structure surround the absorbing atom of Sn and Pb were investigated, respectively, with X-ray diffraction (XRD, spot size 500×700 μm) and extended X-ray absorption for fine structure (EXAFS, spot size 1×1 mm). The XRD investigation was conducted *in situ* up to 36 h, while up to 120 min after current stops. The EXAFS investigation was performed *in situ* for current stressing or thermal aging up to 48 h. The as-prepared specimens of 95Pb5Sn and 97Pb3Sn solders were also investigated for EXAFS. The Sn K-edge spectra of as-prepared 95Pb5Sn and 97Pb3Sn solders were also done for comparing the local environment of Sn atoms. The recrystallization behavior of the Sn-rich phase was further investigated to reproduce and confirm the behavior with a flip chip 95Pb5Sn solder joint 24 days after current stops. The solder joint was stressed with electrical current at 4.2×10^4 A cm^{-2} for 68 h under ambient condition. The microstructure of the recrystallized Sn grain, 24 days after current stops, was investigated with high resolution transmission electron microscope (HRTEM) at 200 kV.

3. Results and discussion

Fig. 2(a) shows the *in situ* synchrotron XRD patterns of 95Pb5Sn solder strip with respect to the current stressing sequence. Each scan took 40 min to complete. As shown in the 1st scan, five peaks correspond to Pb and two peaks correspond to Sn were found before current stressing. Only three peaks can be found in the second scan as the sample at 35 h of current stressing. The peaks of Sn(2 1 1), Pb(3 1 1), Sn (3 2 1), and Pb(2 2 2) disappeared coincide with an obvious shift to a lower two theta angle of the remaining peaks. A prominent increase in the intensity of Pb(1 1 1) peak can also be found at the second scan of 35 h current stressing. With the time further increased to 36 h, the intensity of all the peaks decreased. Pb(1 1 1) dominates at the third scan of 36 h but it also disappeared at the fourth scan of one minute after current stops. A comparison between the third and the fourth scans seems to tell that all diffraction peaks disappear at some moments after 36 h of current stressing. For the following scans without current stressing, the recrystallization starts from the Pb(2 0 0) plane after current stops. Sn(2 1 1) appears in the sixth scan as 60 min after current stops. A strong Pb(2 2 0) peak appears at the seventh scan as 120 min after current stops. The peak intensity of Pb(2 0 0) continues to grow and becomes the strongest one at the seventh scan. The Pb(3 1 1), Sn(3 2 1), and Pb(2 2 2) only appears in the as-prepared specimen but becomes invisible in the following scans.

The evolutions of the facets of the Pb matrix and Sn secondary phase delineate that the current stress induces an electrorecrystallization of the 95Pb5Sn solder. The evolution occurs through the diminishing of the Sn crystal, the relaxation of crystallinity of the Pb matrix, and the recrystallization of Pb and Sn. The disappearance of Sn is believed to be a result of the dissolution of Sn in the Pb lattice as the electrical current stressing induces supersaturation of Sn in Pb matrix [17]. As revealed in the expanded XRD (Fig. 2(b)), the Pb peaks remaining in the second scan shift to a lower two theta angle. The shift means the relaxation of the crystal lattice, mainly induced by electrical current stress. An eminent relaxation occurs to the strongest Pb(2 2 2) of the as-prepared strip. The Pb(2 2 2) converts to Pb(1 1 1) at 35 h of current stressing. The complete relaxation of all the crystalline lattices, including Sn, seems to occur after 36 h. The phenomenon of Sn dissolution, and thus the supersaturation is believed to be a result of the crystal relaxation of the Pb matrix. Relaxation of crystal structure proceeds through the enlargement of lattice parameter as seen in Fig. 2(b), the expanded spectrum of Fig. 2(a) between 30° and 38°. The peaks of Pb(1 1 1) and (2 0 0) shift to the left at 35 and 36 h of current stressing as indicated by the vertical dotted line. It indicates that the lattice parameter of Pb matrix increases 0.6% during current stressing. The enlargement of the lattice parameter of the Pb matrix during current stressing gives rooms to the excess dissolution of Sn in the matrix and thus the disappearance of the Sn crystals.

It is seen that the Pb(200) orientation disappears upon current stressing as reflected by the third scan in Fig. 2(b). The disappearance of the crystalline orientation is believed to be a result of crystal collapsing by the impingement of electron force. The matrix microstructure variation under current stressing is of interest and is worthy of further investigation in the future. The phenomenon of the relaxation of entire crystal lattices indicates that the Pb matrix reaches an amorphous or amorphous-like status after the long period of current stressing in this study. This is an interesting observation for metal alloy may be amorphized by means of electrical current stressing. It deserves further investigation for later study. The high energy meta-stable status will induce the subsequent recrystallization of Pb matrix after current stops. The recrystallization initiates the growth of the Pb(2 0 0) at the fourth scan and then the Pb(2 2 0) as observed at the seventh scan. Worthy of notice is that, the peaks of Pb (1 1 1) and (2 0 0) returned to the original position after current stops. The displacement of Pb lattice parameter is recovered during the recrystallization process. The recovery of the Pb lattice dimension closes the excess dissolution allowance for the second phase. Consequently, the Sn(2 1 1) starts to precipitate along with the recrystallization of the Pb matrix. The recrystallization observed herein is a consequence of the stabilization of the meta-stable Pb matrix induced by electrical current stressing. Such recrystallization is thus termed electrorecrystallization. This process is quite different from the reported electrical current related recrystallization process [18,19]. Recrystallization may be induced by incorporating electrical pulsing on a deformed metal [18,19]. The electrical current induces local temperature rise which triggers the recrystallization of the deformed area [18,19] at the expense of the strain energy, that is different from the electrorecrystallization described in the present study.

The dissolution of the Sn second phase upon current stressing is further evidenced by the synchrotron EXASF (Extended X-ray Absorption Fine Structure) spectra. Fig. 3 shows the Sn K-edge spectra of the as-prepared 97Pb3Sn, 95Pb5Sn solder strips, current stressed and thermal heated 95Pb5Sn solder strips. The time and temperature of the thermal heating were controlled the same as that of the current stressing in this experiment for comparison. The inset shows the wide spectrum of the EXAFS spectra. The maximum absorption edge arises from an electric transition from 1s to 5p states. The absorption edge indicates the same valence state of Sn atom. The oscillations beyond the absorption edge are the EXAFS which represents the environmental surroundings of the Sn atom. The EXAFS of the thermal heated 95Pb5Sn is not smooth due to the magnetic field or electric field induced by the heating coil. Nevertheless, the overall spectra shows the EXAFS oscillations of the current stressed 95Pb5Sn and as-prepared 97Pb3Sn are similar. The oscillation of thermal heated 95Pb5Sn is similar to that of

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