“Cryogenic *in situ* microcompression testing of Sn”

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Abstract

Characterizing plasticity mechanisms below the ductile to brittle transition temperature (DBTT) is traditionally difficult to accomplish in a systematic fashion. Here, we use a new experimental setup to perform *in situ* cryogenic mechanical testing of pure Sn micropillars at room temperature and at -142 °C. Subsequent electron microscopy characterization of the micropillars shows a clear difference in the deformation mechanisms at room temperature and at cryogenic temperatures. At room temperature, the Sn micropillars deformed through dislocation plasticity while at -142 °C they exhibited both higher strength and deformation twinning. Two different orientations were tested, a symmetric (100) orientation and a non-symmetric (4 5 1) orientation. The deformation mechanisms were found to be the same for both orientations.

Keywords: Tin, Solder, DBTT, small-scale testing, EBSD, transmission EBSD, TEM, cryogenic testing

1. Introduction

The use of small-scale mechanical testing techniques provides a number of advantages for evaluating material in extreme environments. For example, one advantage is that small single crystals can be milled from polycrystalline samples so that similarly-oriented samples can be systematically tested over different conditions. A further advantage is that small-scale testing gives us the ability test samples in situ in an electron microscope in order to observe deformation mechanisms directly as well as monitor the quality of the tests. One environmental extreme of interest for aerospace applications is ultracold temperatures. For space applications in particular, it is important to understand the mechanical performance of components at the extreme temperature conditions seen in service for an accurate assessment of a components performance. For solder alloys used in microelectronics, cryogenic temperatures can prove especially problematic. At low temperatures Sn-based solders undergo a ductile to brittle transition that leads to brittle cracks, which can result in catastrophic failure of electronic components, assemblies and spacecraft payloads.

Sn-Pb solder joints are used extensively in packaging of electronics. These joints provide both electrical connections and mechanical support for the electronic package. Differences in thermal expansion coefficient of materials used in electronic packaging cause cyclical strains in the solder joints upon thermal cycling [1]. The mechanical properties of Sn are of particular interest since Sn is the major constituent in most solders used by industry. Reliability of the solder joint depends upon a number of mechanical and microstructural evolutionary processes that occur interactively during its service lifetime [2]. Approximately 70% of failure in electronic circuitry is solder related,

therefore understanding the deformation mechanisms associated with Sn is increasingly important [2]. At room temperature Sn has a body centered tetragonal (BCT) structure. Sn exhibits an allotropic phase transformation from the BCT structure to a diamond cubic structure at 13.2 °C, commonly known as tin pest. The transformation is accompanied by a volume increase of 27% [3]. However this transformation is rather sluggish and in pure Sn, requires nucleation and growth with an incubation period requiring months and even years for completion [4-5]. Since most space applications are thermal cycled from hot to cold temperature extremes over a relatively short time frame (hours), the primary structure of interest is the BCT structure of Sn and the corresponding DBTT that this structure exhibits. The crystal orientation and microstructure of the Sn phase will have significant effects on the mechanical properties of solder joints [6]. This structure gives rise to several unique deformation mechanisms that change as a function of temperature, strain rate and orientation. At room temperature and relatively slow strain rates, Sn deforms by dislocation-mediated slip. The primary slip systems at room temperature have been identified as $\{100\}\langle 001\rangle$, $\{110\}\langle 001\rangle$ and $\{100\}\langle 010\rangle$ [7,8]. While these are the preferred slip systems, depending on the crystal orientation, Sn can access up to ten different slip systems [9]. Conversely, at higher strain rates, one of the more recognizable deformation mechanisms at room temperature is tin cry [10]. Tin cry is a result of twinning, or shearing of crystallographic planes in Sn. Chalmers [11] first identified the primary twinning system at room temperature to be the $\{301\}\langle 103\rangle$. Additional studies have also identified $\{101\}\langle 101\rangle$ as a secondary twinning system [12]. In the cold temperature regime, solder has been primarily characterized by charpy impact testing [13]. Only limited studies have been performed below room temperature but

above the DBTT of -125°C [14]. While the charpy test is very useful for identifying the transition from ductile to brittle behavior, it does not shed light on the mechanisms that are driving these transitions. Previous studies have demonstrated the use of *in situ* small-scale cryogenic indentation testing [15], however no similar systems have been reported that could reach a low enough temperature to characterize the DBTT of Sn. There has yet to be a systematic study that explores the deformation mechanisms below the DBTT and at strain rates that do not induce fracture, which is representative of solder joints that are thermal cycled in this regime. The objective of this study is to better understand the change in deformation mechanisms responsible for the ductile to brittle transition in Sn. This study discusses the use of a new experimental apparatus- an *in situ* scanning electron microscope (SEM) cryogenic mechanical testing system and its initial application to characterize the plasticity of Sn both at room temperature and below the DBTT.

2. Experiments and Methods

2.1 Specimen fabrication

Micro-compression specimens (micropillars) with a square cross-section of 3×3 microns in width were fabricated from a polycrystalline Sn sample using a dual beam focused ion beam (FIB) microscope. Prior to micropillar fabrication, the sample was mechanically-polished on two adjacent faces. During polishing the Sn was mounted directly next to a thin piece of steel in order to prevent deformation and rounding near the edge of the Sn. The samples were planarized using SiC grinding paper with water as a lubricant and were then polished with $0.3\mu\text{m}$, $0.1\mu\text{m}$ alumina polishing solutions, and $0.05\mu\text{m}$ colloidal silica polishing solution.

Prior to ion beam machining, Electron Backscattered Diffraction (EBSD) was used to identify grains for micropillar locations. Two grains were chosen for micropillar fabrication in order to investigate any effect of orientation on deformation behavior below the DBTT. The first grain had a (100) orientation with respect to the applied load direction and the second grain was rotated approximately 52° from the first orientation with a (451) orientation. Fabrication of the micropillars was similar to the method demonstrated by Reinhold et al. [16] using a FEI Quanta dual beam FIB/Scanning Electron Microscope (SEM) with a Ga^+ ion source operated at 30keV. Micropillars were fabricated close to the polished edge using an ion current of 1nA for coarse milling and 10pA for a final polish of the pillar faces. The sample taper was controlled by polishing all four sample sides individually with adjusted tilt angles on the order of 2 degrees. A final polish was done on the top surface of the micropillar by tilting the sample to 90° in order to ensure that top of the micropillar is flat and uniform. These steps were taken in order ensure a homogeneous stress distribution during loading. A total of seven micropillars were manufactured in the (100) grain and four micropillars were manufactured in the (451) grain.

2.2 Cryogenic Micromechanical Testing

Micropillar tests were performed on the (100) and (451) oriented specimens at both room temperature at -142°C . Testing was performed *in situ* in the FIB using a Hysitron PI-85 nanoindenter with a flat-tip diamond punch. To perform cryogenic testing, a custom built cryogenic cooling system manufactured by Hummingbird Scientific was used in conjunction with the Hysitron PI-85 nanoindenter (see Figure 1).

The cryo system cooled the tip and sample simultaneously in order to minimize thermal drift. Figure 1a shows a schematic of the cooling system and the nanoindenter. Figures 1b and 1c show temperature data taken by attaching a thermocouple directly to the sample platform to characterize the stability of the cooling system. We want to emphasize that accurate sample temperature and good sample-tip temperature calibration is critical for environmental testing such as this. As an example of the stability achieved by our system, Figure 1b is a cooling curve showing that the system stabilizes approximately 2000 seconds after cooling is initiated. Figure 1c demonstrates the thermal stability of the system when the tip contacts the sample (in the case of the calibration study, the thermocouple was the sample). A Type K thermocouple was used for this study which is rated from -200 °C to 900 °C. A small deviation in the temperature recording can be seen around 25 seconds corresponding to the point at which contact to the thermocouple was made by the diamond indenter tip. After this initial contact the temperature stabilized almost immediately while the tip remained in contact with the thermocouple. It is necessary to perform these tests under vacuum since otherwise ice will form on the specimen from the air humidity.

Cryogenic testing of the Sn micropillars was performed when the sample and tip stabilized at -142 °C (± 2.5 °C) as measured by a thermocouple attached to a pure Sn sample that was mounted on the sample platform. A flattened conical diamond tip with a diameter of 5 μm was used to compress the micropillars. Sample loading was performed in displacement control with a strain rate of 10^{-3} s^{-1} . The load and displacement data as well as SEM images were captured during the test.

2.3 EBSD and Transmission EBSD of the Micropillars

As mentioned in section 2.1, EBSD was initially used to identify grains for micropillar fabrication. After the micropillars were compressed both at room temperature and at -142 °C, EBSD was used to characterize the orientation of the micropillars. Following EBSD analysis, a representative room temperature and cryo-compressed micropillar were selected from each grain tested. These selected micropillars were lifted out of the bulk sample using an FEI Quanta Dual Beam FIB with Kleindiek nano manipulators. Micropillars were mounted to a standard TEM copper half grid. A platinum layer was deposited prior to any ion milling to protect the micropillar microstructure. Micropillars were thinned to electron transparency in the FIB with a final polish of 5keV and 44pA in order to minimize FIB damage.

The technique of transmission EBSD (t-EBSD) [17] was utilized to characterize the microstructure of the micropillars. The primary advantage of this technique is the improved spatial resolution in comparison to traditional EBSD, which can be below 10nm [18]. T-EBSD was used to characterize the microstructure of both room temperature and cryo-compressed micropillars from each grain using the conditions outlined by Kacher, *et al.* [19]. Analysis was performed using Oxford/HKL data collection software at an accelerating voltage of 30 kV. The samples were analyzed at a working distance of 5mm using a custom-built stage that positioned the TEM grid normal to the electron beam. A step size of 50nm was used for each sample. The collected data were used to construct inverse pole figure (IPF) maps. A low level of data interpolation was used with a criterion of 7 neighbors for extrapolation. Wild spikes and zero solutions were also removed. All maps were compared with band contrast maps to verify that the noise reduction did not lead to any spurious results.

2.4 Transmission Electron Microscopy of Micropillars

All of the micropillars that were examined with t-EBSD were also characterized using Transmission Electron Microscopy (TEM) in order to verify the t-EBSD results and to analyze the underlying defect structure. Characterization of the lift-out samples was performed in a JEOL 3010 LaB₆ TEM operated at 300 kV.

3. Results

3.1 Micro-mechanical testing results

Figure 2 shows the mechanical testing data for the micropillars compressed in the (100) orientation both at room temperature and at -142 °C, which is well below the DBTT of Sn. Micropillars that were compressed at room temperature exhibited slip, as evident in both the stress-strain behavior and slip traces that could be seen on the SEM micrographs of the micropillar surface. The cryo-compressed micropillars exhibited a significant increase in strength and different deformation behavior consisting of uncontrolled strain bursts. Figure 3 shows images from representative micropillar tests from both a sample strained at room temperature and one at -142 °C. Unlike the room temperature micropillars, which kept in constant contact with the diamond tip during the entire test, the cryo-compressed micropillars deformed suddenly and catastrophically and lost contact with the tip after reaching maximum strength. This loss of contact is reflected in the unloading curves of the cryo-compressed micropillars. The observed difference in the mechanical response between the micropillars tested at room temperature and at -142 °C suggests that a different deformation mechanism is activated above and below the

DBTT. Additional characterization techniques such as EBSD, t-EBSD and TEM were employed in order to identify and understand these different mechanisms.

3.2 Characterization of micro compressed micropillars

EBSD was used to characterize both groups of micropillars directly following compression testing. As can be seen in Figure 4, all of the cryo-compressed micropillars showed a different orientation than the parent grain after compression. Further analysis revealed that all of the cryo-compressed micropillars exhibited twinning. Each cryo-compressed (100) micropillar was twinned to a new orientation that was related to the base orientation by a 60 degree rotation about the [100] direction, as can be seen in the t-EBSD scan shown in Figure 5. This twinning behavior is consistent with the formation of the {301} and {101} twin in Sn [20,21]. The twinning angles associated with these two twins are similar, 62.8° and 57.2°. The 60° observed rotation results from misfits between segments, which is consistent with observations of low angle tilt boundaries in the TEM. However, the t-EBSD analysis of the cryo-compressed micropillar, shown in Figure 5, clearly resolves two twins within the micropillar and enabled direct characterization of each individual twin. The microstructure shown in Figure 5 has two twin orientations. The first twin, labeled as twin 1 in Figure 5 is consistent with the {301} twin with a 62.9 degree rotation about the <001>, and second twin is rotated 58.9 degrees from the first twin, also about the <001> which is consistent with the {101} twin.

TEM analysis was performed on the micropillars in order to verify the t-EBSD results and look for evidence of any other underlying deformation mechanisms. Figure 6 shows a TEM micrograph of the cryo-compressed micropillar. It can be clearly seen

from the TEM micrograph that the twin boundaries directly correlate to the t-EBSD results. TEM analysis also revealed the presence of low angle tilt boundaries. Figure 7 shows one of the twin boundaries as indicated in Figure 6 at higher magnification and with selected area diffraction patterns. It is interesting to note that the interiors of both the parent (untwinned) and twinned grains are relatively free of defects, consistent with the deformation being accommodated exclusively through twinning.

Figure 8 shows the t-EBSD scan of a micropillar compressed at room temperature. Here, the orientation remains unchanged from the parent grain, consistent with dislocation plasticity and no twinning at room temperature. The observed orientation gradient across the length of the micropillar in the EBSD map suggests an accumulation of geometrically necessary dislocations. TEM was also performed on the room temperature compressed micropillar, which confirmed the t-EBSD results as shown in Figure 9. The room temperature compressed micropillar did not show any evidence of twinning, rather the microstructure was characterized by a high density of dislocations. Due to the high dislocation density, upon thinning the room temperature compressed micropillar bent considerably preventing reliable characterization of the Burgers vectors of the dislocations.

3.3 Characterization of Micropillars from a (451) oriented grain

In order to verify that the observed twinning behavior at cryogenic temperatures was not related to the highly symmetric (100) orientation, the experiment was repeated on a (451) oriented grain, which is rotated approximately 52° from the (100) orientation. Presumably, this orientation can be considered random and would not result in any

preferred axis for twinning. Similar to the first orientation, the (4 5 1) oriented micropillars compressed below the DBTT exhibited a significantly higher strength and very different deformation behavior than micropillars compressed at room temperature. Figure 10 summarizes the stress-strain behavior for micropillars compressed in the (451) orientation. EBSD and t-EBSD of these micropillars after compression was performed, and again the cryo-compressed micropillars exhibited twinning and the room temperature micropillars exhibited dislocation slip. Similarly both twin types were observed in the (451) orientated grain. As the stress vs. strain data shows, for the (451) orientation both the cryo-compressed micropillars and the room temperature micropillars lost contact with the tip upon yielding and demonstrated large strain bursts. The reason that the (451) room temperature case also lost contact with the tip upon loading in this orientation is presumably due to large dislocation avalanches along a limited number of systems. Whereas the symmetric (100) orientation has multiple dislocation slip systems with similar Schmid factors, the (451) orientation has a few highly-preferred slip systems.

3.4 Discussion

By examining the deformation behavior of Sn within a single grain at both ambient conditions and below the DBTT of Sn, the deformation behavior can be examined as a function of temperature. As the temperature is lowered, dislocation motion tends to become more limited due to the lack of thermal assistance for dislocations to overcome the Peierls barrier. In BCC systems the DBTT is observed at temperatures similar to those at which plastic deformation by twinning becomes more important than dislocation slip [22]. Deformation twins provide high stress

concentrations that have been recognized as potentially important for crack nucleation [23]. Similar to BCC systems, the BCT structure of pure Sn exhibits deformation twinning below the DBTT. As deformation twinning was observed in both the (100) orientation and (451) orientation, this suggests that this mechanism is a direct result of temperature and not orientation.

4. Summary

In order to design engineering applications that can withstand extreme environments, we must first understand the underlying deformation mechanisms that can hinder material performance. Prior to this study, deformation mechanisms of pure Sn below the DBTT were not well characterized due to a lack of experimental methods for doing so. In this study, we have developed a novel *in situ* cryogenic apparatus that has made it possible to systematically study plasticity mechanisms down to -142 °C. Through EBSD and TEM characterization of the twinning behavior at cryogenic temperatures, we have gained a greater understanding of the deformation mechanisms that are active below the DBTT.

5. Acknowledgments

Part of this research was carried out at the Jet Propulsion Laboratory, California Institute of Technology, under a contract with the National Aeronautics and Space Administration. AL was supported by a NASA GRP Fellowship and also by Boeing, Inc. We would like to thank both Hummingbird Scientific, Inc. and Hysitron, Inc. for help with the design and fabrication of the cryogenic testing apparatus. The TEM analysis was performed at the National Center for Electron Microscopy at Lawrence Berkeley National Laboratory, which is supported by the U.S. Department of Energy under Contract # DE-AC02-05CH11231.

Figure 1. Overview of the *in situ* cryogenic testing system. (a) Schematic of the cooling system and the nanoindentation stage. (b) Cooling curve showing that the system stabilizes approximately 2000 seconds after cooling is turned on. (c) Demonstration of the thermal stability of the system by using a thermocouple as a sample. A small deviation in the temperature recording can be seen around 25 seconds corresponding to the point at which contact to the thermocouple was made by the diamond indenter tip. After this initial contact the temperature stabilized almost immediately while the tip remained in contact with the sample.

Figure 2. Summary of the stress-strain behavior of the Sn micropillars from the (100) grain, compressed both at room temperature and at -142 °C . The cryogenic micropillars show both higher strength and large strain bursts at yielding.

Figure 3: SEM micrographs from in situ microcompression testing at room temperature (top row) and -142 °C (bottom row). (a) Image near the start of the room temperature test, (b) image towards the end of the room temperature test, (c) image from a different angle after the room temp test.). (a) Image near the start of the cryogenic test, (b) image directly after a large deformation event in the cryogenic test, (c) image from a different angle after the cryogenic test.

Figure 4: The seven (001) Sn micropillars corresponding to the mechanical data shown in Figure 2. (a) SEM image prior to compression, and (b) the corresponding EBSD map (ipf Z) after all seven micropillars have been compressed. The 4 micropillars at the left were compressed at room temperature, while the 3 micropillars deformed at -142C are on the right. As can be seen from the EBSD map, the cryogenically-tested micropillars have a different orientation than the parent grain below after compression.

Figure 5: Transmission EBSD maps of a cryogenically-compressed micropillar from the (100) orientation showing the presence of 2 twins. (Maps are shown as IPF Z, Y and X)

Figure 6: TEM micrograph of cryo-compressed micropillar 6, the boxed region corresponds to the boundary shown in Figure 7.

Figure 7: (a) TEM micrograph of a twin boundary from the cryogenically-compressed micropillar shown in Figures 5 and 6. Selected area diffraction pattern of the boundary (b), left of the boundary (c), right of the boundary (d). The zone axes are labeled in (c) and (d).

Figure 8: Transmission EBSD map (IPF X) of a (001) micropillar compressed at room temperature showing that the micropillar maintained its original orientation after compression. Even though the entire micropillar is nominally the same orientation as it was before compression, a slight color gradient can be seen corresponding to geometrically-necessary dislocations arranged in the micropillar. The boxed region corresponds to the region imaged in Figure 9.

Figure 9: TEM micrographs of the deformed room temperature compressed micropillar shown in Figure 8. The sample is heavily dislocated after deformation, unlike the cryogenically-compressed pillar shown in Figure 6.

Figure 10: (a) Stress vs. strain data from the (451) oriented micropillars tested at room temperature (red) and -142 °C (blue). (b) T-EBSD of a (451) micropillar compressed at -142 °C, showing that the entire micropillar has reoriented through twinning. (c) An *in situ* SEM image before a twinning event in a micropillar compressed at -142 °C, (d) corresponding image after twinning event.

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