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The unusual size effect of eutectic Sn/Pb alloys in the micro regime: Experiments and modeling

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ABSTRACT

Eutectic alloys are widely used as solder-joint materials due to their suppressed melting points, but when used in micro-devices with small dimensions, their characteristic lamellar microstructure may lead to an internal length scale that affects strength. Here, we report an unusual 'smaller-being-weaker' phenomenon in eutectic Sn/Pb alloys with fine lamellar microstructure, namely, in the specimen-size regime close to and slightly larger than the interphase lamellar spacing, the strength decreases with decreasing size, while above this regime the strength tends towards the bulk value. Theoretical modeling indicates that in a fine lamellar microstructure, high contents of dislocations are retained, so that strength is governed by mutual dislocation interactions, rather than by dislocation starvation. Therefore, in smaller samples, fewer interphase lamellar boundaries are present to block dislocations, thus resulting in a 'smaller-being-weaker' behavior. In samples a lot larger than the lamellar spacing, significant strengthening arises from Taylor hardening and mutual dislocation interactions as a result of significant dislocation retention by the interphase boundaries, so that strength does not depend on specimen size anymore. In a course lamellar microstructure, however, even a larger micro-specimen may contain insufficient interphase boundaries to significantly retain dislocations, and strength may be governed by the starvation effect due to significant loss of dislocations at free surfaces. In this case, the size effect of strength may become a lot milder, or even exhibit the conventional "smaller-being-stronger" behavior. The results here supplement conventional knowledge on size effects in micro-scaled crystalline materials, and provide important implications on solder-joint design in micro-devices.

1. Introduction

In the last decade, size effects of micro-scaled metallic materials have been intensively investigated [1–3], and a 'smaller-being-stronger' phenomenon, reported for the first time in metallic whiskers in the 1950's [4], has now been established as the norm behavior of monolithic metals of micro sizes. Such a deformation behavior obeys a power law of size [5–8] and has a jerky, stochastic nature [9–11]. In small crystals, mobile dislocations can glide to and annihilate freely at free surfaces without significant accumulation or multiplication, leading to a sustained dislocation-starved state with a high flow stress [3,12], and the quantized emission of new dislocations to maintain the plastic flow leads to discrete strain bursts of a stochastic nature [10,13]. In larger, but still micro-sized, crystals, a 'source truncation' mechanism operates in which single-armed dislocation sources truncated by free surfaces govern the flow stress, and since the source arms in smaller specimens are generally shorter, their operational stress and hence the flow stress is higher [14–16]. As the loss of dislocations is the cause for the strain bursts and jerky deformation [9,13,17–19], to improve the deformation behavior a number of attempts have been made to control the dislocation density in micro specimens, via means including pre-straining [13,20,21], surface coating [21,22], inserting grain boundaries [23–25]. In fact, the majority of real-life metallic materials for engineering applications are alloys with complicated microstructures, such as second-phase precipitates, grains, inclusion particles, twins or solutes. Hence, understanding the size effect of metals with merely a monolithic, single-crystalline structure would be insufficient and indeed irrelevant for real applications, and for alloys in real use their complicated microstructures should introduce an internal scale length that would couple with the external specimen size to affect the mechanical behavior [26–30]. For example, an unusual phenomenon of a "weakest size" in the

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micro-regime was recently discovered in duralumin with a precipitated microstructure, due to the interplay between the dislocation-starvation mechanism controlled by the external sample size in the micro range, and the precipitation-hardening mechanism controlled by the internal length scale of the precipitate spacing [31].

Eutectic Sn/Pb solders have been widely used for electrical and structural joints due to their low melting points, good wettability, acceptable electrical conductivity and good plasticity [32–35]. The typical microstructure of eutectic Sn/Pb alloys features alternant lamellae of the two constituent phases with an average spacing that is sensitive to the solidification conditions of the alloy [36,37]. Recently, the length-scale dependent plasticity in metallic multilayers fabricated by successive physical deposition has been studied [38–40], and the results also conform to the "smaller-being-stronger" rule in that strength monotonically increases as the multilayer thickness decreases. In lamellar Ti-Al alloys in which the lamellae are formed naturally in the wrought process, the lamellar spacing rather than the grain size dominates the yield strength according to a dependence similar to the Hell-Patch relation [41]. However, the notion of "smaller-being-stronger" for lamellar materials has been challenged by the observation that the finer lamellar microstructure leads to lower tensile flow strength in cast Al-based alloys [42]. In summary, although the plasticity of small-size materials coupled with multiple length scales of the internal microstructures has received a lot of attention [43–46], understanding on how the strength of micro-sized eutectic alloys depends on their lamellar layers is still lacking.

In this article, we report a significant 'smaller-being-weaker' size effect of strength in micro-sized eutectic Sn/Pb alloys with a characteristic lamellar microstructure. By controlling the lamellar spacing through different solidification treatments, the effect of the internal length scale on the deformation behavior of Sn/Pb micropillars is studied. Theoretical modeling based on a continuum dislocation model and two-dimensional dislocation dynamics simulations is carried out to understand the reason of such an unusual size effect of strength.

2. Experimental

An as-received eutectic Sn/Pb alloy (63 wt % Sn-37 wt % Pb) was used as the starting material. A large piece of the specimen was cut into two which were then melted by heating over the eutectic point of 183 °C. Afterwards, one piece of the specimen was air cooled (AC) at room temperature (RT ~ 20 °C), whilst the other was slowly cooled within the furnace (FC). The oxide layers of the sample surfaces were removed by mechanical polishing. The samples were then left to fully age at RT for more than two weeks to achieve a stable microstructure [33]. Then, the AC and FC samples were mechanically ground and fine-polished with 1 μm diamond paste to achieve mirror-like surfaces. They were then further vibration-polished with a Buehler VibroMet 2 polisher using a MasterSet 0.06 μm colloidal silica suspension with anhydrous alcohol to achieve a stress-free surface state.

The eutectic microstructure of the samples was examined using a Hitachi S4800 FEG scanning electron microscopy (SEM). Micropillars were milled using focused ion-beam (FIB) milling by an FEI Helios Nanolab 600i dual beam FIB/SEM system operated at 30 kV ion beam voltage. Since both the AC and FC samples were cut from the same as-received alloy and melted and cast in the same way albeit cooled differently after reheating to above the eutectic temperature, they contained large grains oriented close to the ~ [001] direction. Thus, in both the AC and FC samples, a large grain with an orientation ~ [001] and size larger than 500 μm, as detected by electron backscattered diffraction (EBSD), was selected for milling the micropillars. For the FIB process, initially, a Pt layer of ~100 nm thick was FIB-deposited homogenously on the selected specimen surface to protect lamellar microstructure from ion damage. The procedure of FIB milling consisted of a series of concentric annular pattern milling steps using a current from 60 nA for the initial coarse milling step, to 50 pA for the last fine milling step. Specifically, pillars of diameter ranging from 1 to 7.5 μm were milled, with the height-to-diameter ratio kept as 2.5: 1.

Compression of the fabricated micropillars was performed at RT (~20 °C) by an Agilent G200 Nanoindenter equipped with a flat-end diamond punch in a load-controlled method. Before the compression tests, thermal drift was controlled to below 0.1 nm/s and was re-measured to check for consistency during the unloading stage. Loading and unloading were programmed at a constant rate of ~2 MPa/s, with the resultant strain rates of all micropillars within the order of ~10^{-3}/s, and the maximum loads were set at values corresponding to engineering stresses of ~150 MPa for the AC micropillars and ~55 MPa for the FC ones. The morphology of the deformed micropillars was imaged using SEM. Furthermore, TEM (transmission electron microscopy) examination was carried out on longitudinal sections of the deformed micropillars, which were prepared by first FIB milling along the height direction of the micropillar to a thickness of ~1 μm, followed by cutting from the bulk substrate and welding onto an Omniprobe by tungsten deposition. The TEM specimens were then finely milled to achieve thicknesses below 150 nm after removal from the Omniprobe and welding onto copper TEM grids. TEM examination was carried out in an FEI Tecnai G2 20 Scanning TEM at 200 kV.

3. Experimental results

Fig. 1(a) and (c) show typical microstructures of the eutectic Sn/Pb alloys. Alternate lamellae of β-Sn (dark phase) and Pb (light phase) are homogenously distributed. Using the line-intercept method, the lamellar thicknesses of β-Sn in the air-cooled (AC) and slow furnace-cooled (FC) alloys were measured to be ~1.5 and ~3.5 μm, respectively, and those for the Pb phase were ~0.45 and ~1.05 μm, respectively. The AC microstructure exhibits finer lamellar structures than the FC, which agrees with previous findings that the cooling rate significantly determines the lamellar structure of Sn/Pb alloys [47]. Fig. 1(b) and (d) show the nominal strain-stress curves of the AC and FC eutectic Sn/Pb micropillars along the direction ~[001]. In both cases, significant stress avalanche occurred with specimen sizes below 3.5 μm, where the applied stress rapidly dropped with prominent strain increments of ~0.15. Larger specimens exhibit smoother deformation with reduced strain. The 2% proof strength $\sigma_{0.02}$ of the eutectic Sn/Pb micropillars measured from the strain-stress graphs are plotted in Fig. 2(a). It can be seen that as the specimen size D increases, the 2% proof $\sigma_{0.02}$ strength of both AC and FC eutectic micropillars increases in the size regime from 1 to ~3.5 μm, which corresponds to an unusual 'smaller-being-weaker' size effect compared with the conventional "smaller-being-stronger" behavior in monolithic metals [2,5,29,48]. As the specimen size increases beyond ~3.5 μm, the strength of both types of Sn/Pb micropillars becomes insensitive to specimen size and tends towards the bulk value. The strength-size trend even in the sensitive regime is not describable by the conventional power law, even with a positive power exponent of size. Fig. 2(a) further indicates that the strength of AC eutectic Sn/Pb with a finer lamellar microstructure is larger than that of the FC state with a coarser lamellar microstructure, with strengths at ~87 MPa and ~30 MPa at D ~7.5 μm, respectively. Meanwhile, the 'smaller-being-weaker' size effect of the FC samples with coarser lamellar structure is much milder than the AC case. Fig. 2(b) shows typical morphologies of deformed AC and FC