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Internal friction associated with the melting of Pb nanoparticles in an Al matrix

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Abstract

Pb nanoparticles embedded in an Al matrix were prepared by ball milling and the internal friction behavior associated with the melting of Pb nanoparticles was studied by means of dynamic mechanical analyzer. The height H of internal friction peak was found to be dependent on the frequencies and heating rates, which indicates a first-order phase transition of Pb melting. The internal friction behavior of Pb nanoparticles is discussed with respect to the surface melting.

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Keywords: Internal friction; Melting transition; Nanoparticles; Ball milling

1. Introduction

Low-dimensional materials research became more and more attractive in recent years because they exhibit different properties from those of the counterpart bulk materials. The melting behaviors of nano-structured materials such as nanoparticles embedded in a continuous matrix have been widely studied by means of various methods including differential scanning calorimetry [1–3], X-ray diffraction [4,5], transmission electron microscopy [6–8] and so on. The results show that most of nano-structured materials demonstrate size dependent melting properties and the melting temperature can either be lower or higher than the melting point of counterpart bulk materials [3,6,9]. Although many experiments have been done, a complete understanding of the melting of nanoparticles remains unclear. So exploring a new method for studying the melting properties of nanoparticles is highly necessary.

Internal friction technique has been widely used to study crystal structures, defects, atomic diffusion, grain boundary relaxation and phase transitions in solids [10–12]. Most recently, the internal friction behavior on fusion of pure metals has been investigated, showing that internal friction is also sensitive to solid–liquid transitions [13–16]. In this Letter, internal friction behavior associated with the melting of Pb nanoparticles embedded in an Al matrix was studied. The system of Pb nanoparticles embedded in an Al matrix is selected because Pb is insoluble with Al and has low melting point (327 °C). In this study, we wish to demonstrate that the internal friction technique could provide a special means for studying the melting behavior of nano-structured materials.

2. Experimental

In order to obtain Pb nanoparticles, 3 g solid mixtures of pure Al (99.999%) and pure Pb (99.999%) with the composition of Al-5 at%Pb were sealed in a steel vial with two large (8.3 g for each) and four small (1 g for each) hardened steel balls in a nitrogen atmosphere, and milled for 12 hours

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with a SPEX 8000 shaker mill. For internal friction measurement, the milled powders were immediately consolidated by means of a specifically constructed die with dimensions of 1 mm × 3 mm × 30 mm under a pressure of 0.9 GPa at ambient temperature for 10 minutes.

Internal friction measurement was performed at different frequencies and different heating rates using a Perkin–Elmer Pyris Diamond dynamic mechanical analyzer (DMA) in a bending mode with single cantilever geometry. During DMA testing, the sample was held between a probe and an end clamp. The sample was subjected to a sinusoidal stress with constant amplitude of 1 N through the probe and the resultant strain developed in the sample was measured simultaneously. The internal friction $\tan\delta$ was determined, where the angle δ is the phase angle difference between the stress applied and responding strain. All data acquisition and processing were completely controlled by a computer.

The morphologies of the samples were observed with scanning electron microscopy (SEM, HITA x-650) in backscattered electron mode. X-ray diffraction (XRD) experiment was carried out on a Philips XPert-PRO X-ray diffractometer with Cu Kα radiation. The differential scanning calorimetry (DSC) experiment was carried out with the Perkin–Elmer Pyris Diamond DSC with the heating rate of 20 °C/min.

3. Results and discussion

A typical microstructure of the as-prepared Al–Pb sample obtained after the consolidation of the milled powders is shown in Fig. 1. It can be found that the Pb particles were refined into nanometer size by ball milling and uniformly dispersed in the Al matrix. The XRD diffraction pattern for the as-prepared Al–Pb sample is shown in Fig. 2, from which we can see that only diffraction peaks for pure Al and pure Pb can be observed and no trace of other intermediate phases is found. The average grain size of Pb nanoparticles was estimated from the diffraction peaks of Pb (111) and (200) plane by using the Scherrer formula [17]: $d = 0.89\lambda/B\cos\theta$, where λ is the X-ray wavelength, B full width at half maximum (FWHM) of the XRD peak and θ the diffraction angle. The average grain size of Al matrix was also estimated from the diffraction peaks of Al (111) and (200) plane. In estimating the average grain size, the effect of the internal strain on the line broadening was neglected [1,18]. The results show that the average grain size of Pb and Al are about 36 and 64 nm, respectively. It can also be found that there exist pores in the sample from the TEM image [16].

Fig. 3a shows the internal friction ($\tan\delta$) change as a function of temperature (T) ($\tan\delta$–T curves) from 280 to 350 °C

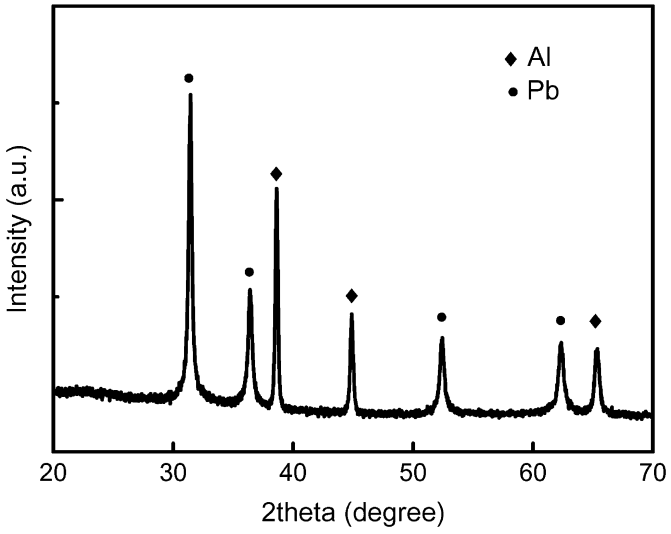

Fig. 2. X-ray diffraction patterns for the Al–Pb sample.

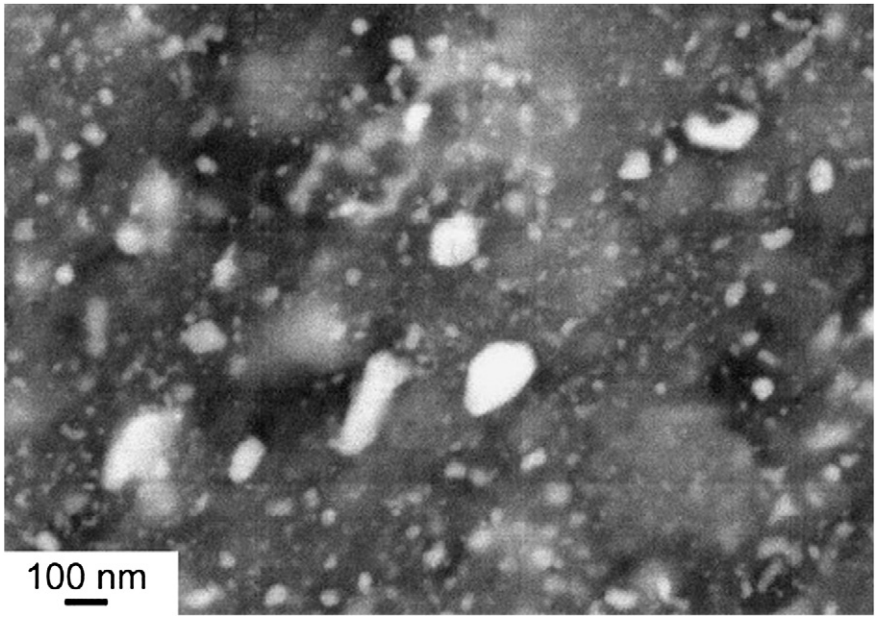

Fig. 1. Backscattered SEM image of the as-prepared Al–Pb sample showing the typical distribution of the Pb nanoparticles embedded in an Al matrix. Bright particles are Pb.

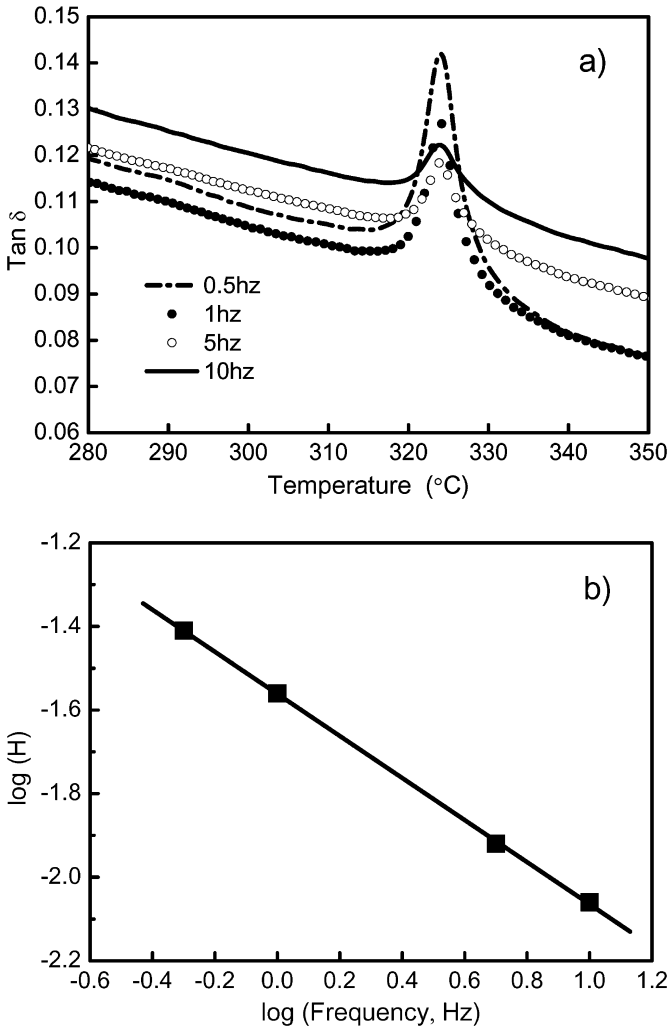

Fig. 3. (a) Internal friction-temperature $\tan\delta$–T curves for the Al–Pb sample with different frequencies, at the heating rate of 2 °C/min. (b) A log–log plot of the internal friction peak height H with frequency.

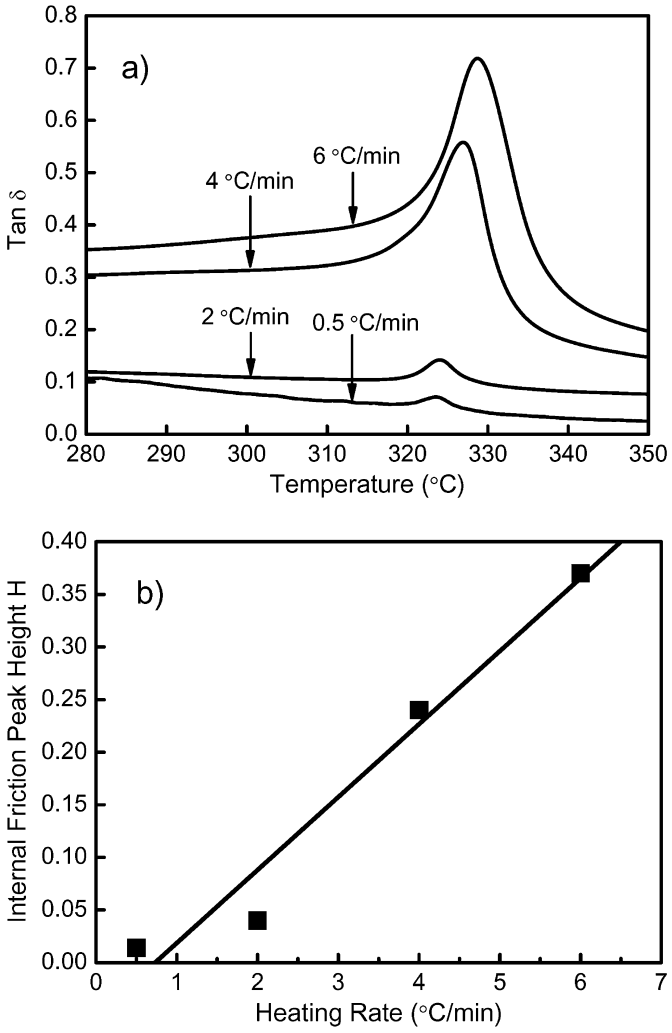

Fig. 4. (a) Internal friction–temperature $\tan\delta$–T curves for the Al–Pb samples with the frequency of 0.5 Hz, at the different heating rates. (b) A plot of the internal friction peak height H vs. heating rates shows a linear correlation to the heating rates.

with the frequencies of 0.5, 1, 5 and 10 Hz at the heating rate of 2 °C/min. At each fixed frequency an obvious internal friction peak occurs around the Pb melting point (327 °C) in the $\tan\delta$–T curve. It can be seen that the position of the peaks does not alter with the frequencies, but the height of the peaks decreases with the frequency increasing. We defined the internal friction peak height as $H = (\tan\delta)_{\max} - (\tan\delta)_{\min}$, here, $(\tan\delta)_{\min}$ refers to the minimum value of $\tan\delta$ before the peak and $(\tan\delta)_{\max}$ is the maximum value of the peak. A plot of $\log(H)$ versus log frequency shows that H is inversely proportional to frequency as shown in Fig. 3b. Fig. 4a shows the $\tan\delta$–T curves for the Al–Pb samples with the frequency of 0.5 Hz at the heating rate of 0.5, 2, 4 and 6 °C/min. It can be seen that the peak temperature and the peak height H are raised as the heating rate $(\mathrm{d}T/\mathrm{d}t)$ was increased from 0.5 to 6 °C/min (Fig. 4b). These features are perfectly in accordance with those of the first-order phase transitions in crystalline solids, which have been theoretically interpreted by Delorme and Belko models [19,20]: $H = K(\mathrm{d}T/\mathrm{d}t)/\omega$, where K is a constant for a given material, $\mathrm{d}T/\mathrm{d}t$ is the heating rate, ω equals $2\pi f$ and f is the cyclic oscillation frequency. The linear relation as shown in Fig. 3b and Fig. 4b means a first-order phase transition of the Pb melting. In addition, it is confirmed that the internal friction peak is associated with a melting transformation of Pb nanoparticles as demonstrated by comparing with DSC curves, on which an endothermic peak around the Pb melting point indicating the Pb melting was observed (Fig. 5).

It is well known that melting is caused by a vibrational instability in the crystal lattice when the root-mean-square displacement between the atoms reaches a critical fraction of the distance between them [21]. The atoms on the surface have a reduced number of bonds, so melting transition will first occur on the surface of materials. In experiment, it has been observed that the Pb particles embedded in an Al matrix melt first at the boundary between Pb particles and Al matrix [6,22]. That is to say, at the heating process the melting transition will first happen on the surface of Pb nanoparticles so that a layer of liquid will surround the solid core of the Pb nanoparticles in our experiment. It is the surface molten fraction of a particle that causes

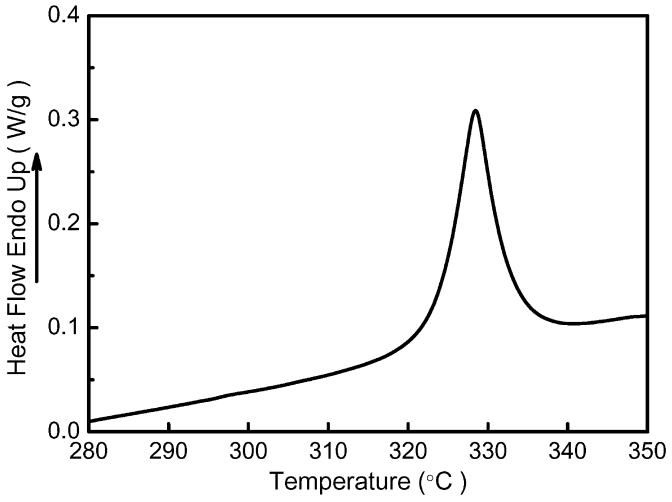

Fig. 5. The DSC results for the Al–Pb sample as a function of temperature showing an endothermic peak associated with the melting of Pb nanoparticles.

the vibration energy dissipation and induces the internal friction peak [16].

Another contribution to the vibration energy dissipation may be the existence of the grain boundary, dislocation and porosity effects in the matrix [23–25]. In our experiment the pure Al sample was also prepared with the same procedure and the experiment results show that there is no internal friction peak for the pure Al sample [16]. However, there appears an internal friction peak for the Al–Pb sample. This fact suggests that the internal friction peak for the Al–Pb sample is not associated with the grain boundary sliding in Al, but is related to the melting of the Pb particles. Although there exist many grain boundary, dislocation and pores in the sample, we think the vibration energy dissipation caused by these factors can only drive up the background and do not have an influence on the internal friction peak.

4. Conclusions

An internal friction peak related to the Pb melting was observed for the Pb nanoparticles dispersed in an Al matrix. The internal friction peak height increased with increasing the heating rates, while decreased with the increasing frequencies. It is suggested that the internal friction peak was caused by the surface melting of Pb nanoparticles. The present study proves that the internal friction technique is effective in studying the melting of nano-structured materials.

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