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Dynamic and Static Restoration Behaviors of Pure Lead and Tin in the Ambient Temperature Range

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Dynamic and static restoration behaviors of pure lead and tin were investigated by compression tests, and the deformation temperature and strain rate were varied in the range from 223 to 348 K and from 2×10^{-3} to $1\,s^{-1}$, respectively. Lead and tin used had two purity levels of 99.999% (5N) and 99.9% (3N), and 5N and 4N, respectively. The hot working simulator was reformed so as to enable compression tests at low temperatures ranged from 223 to 273 K. S-S curves observed in lead and tin were those in dynamic recrystallization and dynamic recovery types of metals, respectively, of which activation energies were 92 to 119 kJ/mol in lead and 49 to 52 kJ/mol in tin. Steady state flow stress in lead with 5N purity was lower than that of tin with 5N purity. A reduction of purity level from 5N to 3N in lead significantly increased flow stress, but difference in purity level of 5N and 4N in tin exerted tiny influence on flow stress at strain rate below $1 \times 10^{-1}\,s^{-1}$. Static recrystallization in lead with 5N purity completed in the holding time of less than 600 s even at 273 K, while tin with the same purity showed a slightly retarded recrystallization progress. A reduction of purity level in both metals extended the time period for completion of recrystallization by more than 2 orders. [doi:10.2320/matertrans.MRA2007078]

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Keywords: *pure lead, pure tin, lead plug, seismic energy absorption, dynamic recrystallization, static recrystallization, softening ratio*

1. Introduction

One of advantageous properties of lead is high energy absorbing property, and an example of its practical applications is a lead plug for seismic isolation.[1] In a lead plug, a cylindrical shape of lead is introduced under a pressurized condition into a hollow column made in the mid-center of a multi-layered natural rubber component. One of the main utilization fields in a lead plug is a bridge construction, where it is set up in the bottom of pillars supporting a bridge. Once an earthquake occurs, a plug can absorb seismic energy by a large plastic deformation in lead in conjunction with a multi-layered rubber, and then collapse of the bridge is possibly avoided. The other advantage of lead in this application is that properties of lead subjected to a seismic deformation can be restored by strain relief due to static recrystallization taking place immediately after deformation at the ambient temperature, which makes possible to use a plug continuously and repeatedly after suffering a number of earthquakes. That is, recrystallization capability at the ambient temperature in lead is effectively utilized in a lead plug for seismic isolation.

Since lead was recognized to be a toxic metal to a human body, animals and other living things, various metals and alloys substituting for lead or without containing lead have been developed.[2,3] A lead plug itself is closed system, having a very few possibility to contact directly with a human body or other living things during and after construction of bridges or other structures with this system. However, workers in manufacturing of lead plugs may be possibly exposed to atmosphere containing lead vapor, and in addition, possibility of elution of lead ion from a plug during a long term use due to such a cause as acid rain is also concerned. Thus, it is expected to develop a new plug material used for seismic energy absorption in place of lead.[1]

Mechanical properties required for a plug material are relatively low strength and such high ductility as to be tolerable to a large amount of deformation caused by an earthquake. In addition, recrystallization temperature in a plug material is needed to be around the ambient temperature, which enables a repeated use of a plug as noted above. The melting point of lead is very low (600 K), and thus, deformation at the ambient temperature corresponds to the hot working temperature region in this metal, inducing occurrence of recrystallization at the ambient temperature in lead.

As metals with the low melting point appear to have the low recrystallization temperature, one of candidate materials which can substitute for lead is tin with the low melting point of 505 K. Basic studies of static recrystallization and creep in metals with the low melting point such as lead or tin were mostly conducted before 1980's,[4–9] but systematic investigations of both dynamic and static restoration processes in these metals which occur in deformation at the higher strain rate over that in creep have not been conducted. The ambient temperature range in a plug used for seismic isolation may be from 233 to 315 K which covers the arctic region to very hot region considering a world wide construction of bridges and other structures. The most past studies on creep and recrystallization behavior of lead and tin were conducted at the temperature above 293 K without covering the low temperature below 273 K.

The present study was conducted to investigate dynamic and static restoration behaviors in pure lead and tin in the ambient temperature range and to examine possibility of substitution of tin for lead as a plug material used for seismic isolation. Both of strength and recrystallization temperature of pure metals are markedly affected by purity, which was varied in two levels in a respective metal. As noted above, the ambient temperature in pure metals with a low melting point corresponds to the hot working temperature, suggesting that

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Table 1 Chemical compositions of lead and tin used in this study (ppm)

Specimen	Pb	Sn	Ag	Bi	Cd	Cu	Fe	Mg	Mo	Ni	Sb	W	N	O
5N–Pb	Bal.	—	—	0.5	0.1	0.1	—	—	—	—	—	—	12.8	76.4
3N–Pb	Bal.	—	—	100	—	100	77	2	40	24	—	—	17.1	76.0
5N–Sn	—	Bal.	0.6	—	0.17	0.12	0.1	—	—	0.1	—	0.1	12.8	76.4
4n–Sn	—	Bal.	—	—	—	2	4	—	—	—	6	—	12.8	89.5

strength or flow stress possibly varies widely depending on the deformation temperature and strain rate.[10–14)] Thus, dynamic restoration behaviors or variations in the true stress-true strain (S-S) curve with the deformation temperature and strain rate were investigated by compression tests using a hot working simulator.[13)] The deformation temperature and strain rate were varied in the range from 223 to 348 K, and 2×10^{-3} to $1\,s^{-1}$, respectively. The hot working simulator was reformed so as to enable compression tests at the low temperature below 273 K. Dynamic restoration process in both metals is investigated and discussed based on analysis using Zener-Hollomon parameter[15)] and the activation energy in a respective dynamic restoration process.

The typical method to investigate the progress of static restoration or static recovery and recrystallization is the microstructural observation. However, the microstructural observation of lead is no easy, because this metal is very soft and is apt to be subjected to plastic deformation or creep deformation during cutting and polishing in preparation of the specimen for metallographic observation. Thus, static restoration kinetics in both metals was investigated by means of the mechanical test.[12,16)] In this method, a variation of flow stress in two S-S curves with the holding time is obtained by an intermittent two-stage compression test, and the progress rate of static recovery and recrystallization is evaluated based on a variation of softening ratio with the holding time. The other advantage of this method is that static restoration progress proceeding during a very short hold time of less than 60 s can be evaluated. Finally, through comparison between dynamic or static restoration behavior in lead and tin as well as the effect of purity level on these behaviors, possibility of substitution of tin for lead as a plug material for seismic isolation is discussed.

2. Experimental Procedure

Chemical compositions of lead and tin used in this study are listed in Table 1. All of them are commercial annealed bar products, of which diameters were 10 mm in lead with purity of 99.9% (noted as 3N hereafter) and tin with purity of 99.99% (4N), 8 mm in lead with purity of 99.999% (5N) and 6 mm in tin with purity of 5N. Impurity elements contained in lead with 3N purity are Bi, Cu, Fe, Mo and Ni, and tin with 4N purity contains Pb and Sb. Both metals contain impurity oxygen of 76 to 90 ppm. Numerous compression test specimens with a column shape were prepared from these bars using a cutter with a thin diamond wheel. The diameter (D) and height (H) in the specimen size were varied depending on a diameter of bars, and these were $10^D \times 15^H$, $8^D \times 12^H$ and $6^D \times 9^H$ mm. All specimens were annealed at 373 K for 300 s.

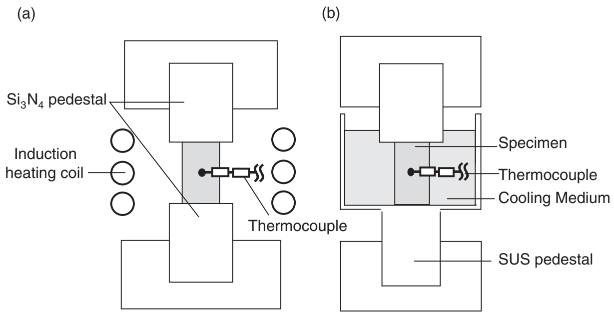

Fig. 1 Schematic diagrams of heating or cooling portion in a hot working simulator. (a) and (b) show heating and cooling of the compression specimen, respectively.

Compression tests were conducted using a hot working simulator of THERMEC MASTER-Z which was developed by one of the authors.[13)] This simulator enables one to conduct hot compression test in wide ranges of strain rate and the temperature. In conventional use of this equipment, the specimen is set between top-side and bottom-side pedestals made by ceramics such as Si_3N_4 and is heated by an induction heating coil, which is schematically drawn in Fig. 1(a). In the present investigation, it is needed to conduct the compression test at the low temperature ranged from 223 to 273 K, and so the reservoir for a cooling medium which is united with the bottom-side pedestal was made as shown in Fig. 1(b). A material of the reservoir and the pedestal was austenitic stainless steel with a grade of the A 316. The specimen was dipped into a cooling medium and was set between both pedestals. A cooling medium used was the mixture of ethanol and dry ice, and the mixed ratio of these materials was optimized to attain the objective temperature. The temperature was measured and controlled by a thermocouple connected to the surface in the mid center in the vertical direction of the specimen.

In dynamic restoration study of lead and tin, the specimen was cooled or heated at the temperature ranged from 223 to 373 K, and was hold for 300 s at a respective temperature, followed by a compression test. Strain rate was varied in the range from 2×10^{-3} to $1\,s^{-1}$ under a constant true strain of 0.69, and the S-S curve in a respective deformation condition was obtained.

In investigation of static restoration behavior in both metals, an intermittent two-stage compression test with various holding time periods was conducted. Test temperatures were 273 K, 298 K and 348 K, and the specimen was hold for 60 s at a respective temperature, followed by the compression test. Strain rate and the strain in the first compression test were $1\,s^{-1}$ and 0.4, respectively. After termination of the first stage test, load was released. Then, the deformed specimen was hold for various time periods ranged

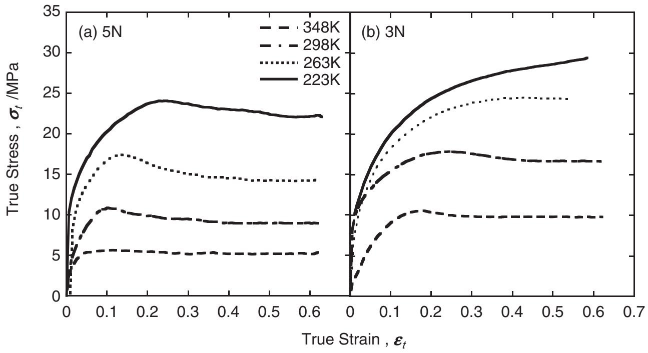

Fig. 2 Variations of Stress-Strain curves with the deformation temperature at strain rate of $2 \times 10^{-3}\,s^{-1}$ in lead. Purity of lead is 5N in (a) and 3N in (b).

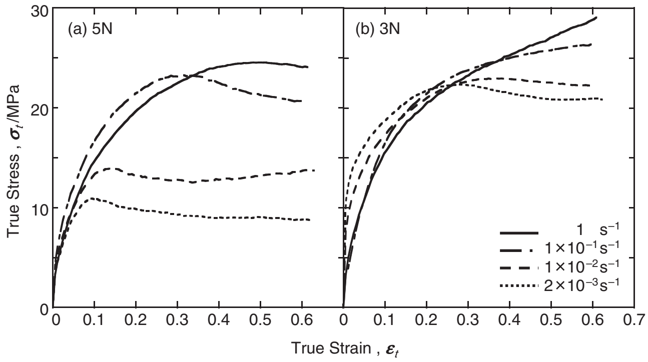

Fig. 3 Variations of Stress-Strain curves with strain rate at the deformation temperature of 298 K in lead. Purity of lead is 5N in (a) and 3N in (b).

from 0.5 s to 600 s, followed by the second-stage compression test at strain rate of $1.0\,s^{-1}$ and in a strain of 0.2. Softening ratio (Xs) which corresponds to the progress ratio in static recovery and recrystallization is obtained based on flow stress values in two S-S curves, and it is expressed by eq. (1)[12,16)]

$$X_S = (\sigma_m - \sigma_y)/(\sigma_m - \sigma_0), \quad (1)$$

where σ_0 and σ_m are 0.2% flow stress and flow stress at strain of 0.4 in the first-stage compression test, respectively and σ_y is 0.2% flow stress in the second-stage test. No progress in static recovery and recrystallization yields $X_S = 0$ for $\sigma_y = \sigma_m$, and completion of recrystallization results in $X_S = 1.0$ for $\sigma_y = \sigma_0$.

3. Experimental Results

3.1 Dynamic restoration behavior in lead and tin

Variations of the S-S curve with the temperature and strain rate in lead are shown in Figs. 2 and 3, respectively. Figure 2(a) shows variations of the S-S curve with the temperature in high purity lead with 5N purity, and these show a typical dynamic recrystallization type of the S-S curve at all temperatures ranged from 223 to 348 K. That is, after yielding peak stress (noted as σ_p) in a short strain, flow stress decreases with the increase of strain, followed by steady state flow stress (noted as σ_s) with the further extension of strain. Both values of σ_p and σ_s markedly increase with a reduction of the temperature. On the other hand, it is found from S-S curves in lead with 3N purity shown in Fig. 2(b) that S-S curves at the low temperatures of 223 K and 263 K show continuous work hardening without reaching to peak stress with the increase of strain. An elevation of the temperature above 298 K gives rise to a dynamic recrystallization type of the S-S curve. It is found from comparison of S-S curves in Figs. 2(a) and (b) that flow stress in lead with 3N purity is much higher than that in lead with 5N purity in a respective deformation condition.

Figure 3 shows variations of S-S curves with strain rate at 298 K in lead. As shown in Fig. 3(a), the S-S curve in lead with 5N purity obtained by deformation at strain rate of $1\,s^{-1}$ shows continuous work hardening with straining. A typical S-S curve observed in a dynamic recrystallization type of metals is seen at strain rate below $1 \times 10^{-1}\,s^{-1}$, and peak

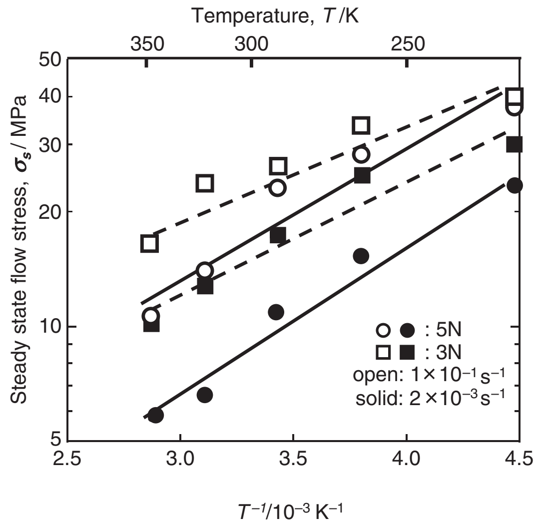

Fig. 4 Variations of steady state flow stress on a reciprocal of the absolute temperature in lead.

strain and steady state flow stress decrease with a reduction of strain rate. On the other hand, in lead with 3N purity shown in Fig. 3(b), the continuous work hardening type of the S-S curve is observed at strain rate above $1 \times 10^{-1}\,s^{-1}$, and steady state flow behavior after yielding peak stress is seen at the lowest strain rate of $2 \times 10^{-3}\,s^{-1}$. Lead with 3N purity results in much higher flow stress at a respective strain rate compared with that of lead with 5N purity, and this increment of flow stress with a reduction of purity in lead becomes more marked at lower strain rate. Adiabatic heating occurring in deformation at higher strain rate tends to become more pronounced in the higher strength metal, which may reduce flow stress in the higher strain. This appears to cause the reduced increment in flow stress observed at the higher strain rate with a reduction of purity from 5N to 3N in lead. Variations in S-S curves with the temperature and strain rate shown in Figs. 2 and 3 were very similar to results observed at temperatures above 285 K in lead with 2N purity reported by Sakai *et al.*[17)]

Figure 4 shows variations of steady state flow stress in a log-scale with a reciprocal absolute temperature in lead. A linear relationship between them is observed at both strain rates and in both purity levels of lead. The higher strain rate

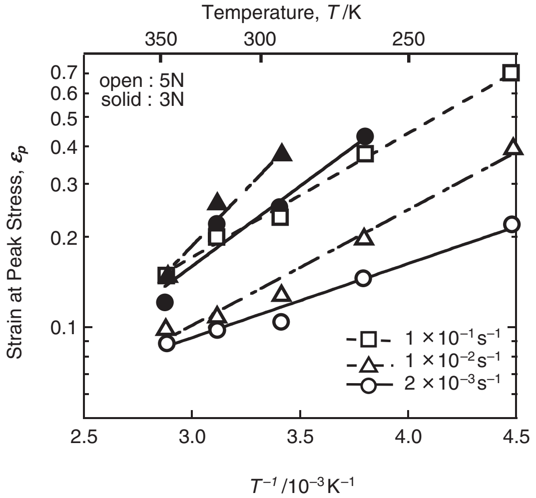

Fig. 5 Variations of peak strain with a reciprocal of the absolute temperature in lead.

and the lower purity level of lead result in the higher flow stress at all temperatures. Figure 5 shows variations of strain at peak stress (noted as ε_p) in a log-scale with a reciprocal absolute temperature in lead. It has been well confirmed that dynamic recrystallization starts at strain of around 0.8 or $0.9 \times \varepsilon_p$[13,18] and so ε_p is the strain almost corresponding to the onset of dynamic recrystallization. Figure 5 clearly shows that this strain varies depending on not only the deformation condition such as the temperature or strain rate, but also the purity level in lead. A reduction in the temperature or the increase in strain rate increases this strain. A reduction of purity from 5N to 3N in lead markedly increases ε_p, or retards the onset of dynamic recrystallization. Under the maximum strain of 0.69, dynamic recrystallization does not start at the temperature below 273 K and strain rate over $1 \times 10^{-2}\,s^{-1}$ in lead with 3N purity.

Figures 6(a) and (b) show variations of the S-S curve with the temperature in tin with purity of 5N and 4N, respectively. The temperature is varied in the range from 223 to 348 K and strain rate is $2 \times 10^{-3}\,s^{-1}$. Differently from results obtained in lead, difference in the purity level between 5N and 4N in tin exerts very tiny influence on the S-S curve and flow stress. The S-S curve obtained by deformation at 223 K yields the continuous work hardening with straining, and all S-S curves at the temperatures above 263 K exhibit a typical dynamic recovery type of the S-S curve, which yields work hardening in the early stage of straining, followed by steady state flow stress.

Figures 7(a) and (b) show variations of the S-S curve with strain rate at 298 K in tin with 5N and 4N purities, respectively. Both purity levels of tin exhibit a typical S-S curve observed in dynamic recovery type of metals, and steady state flow stress increases with the increase of strain rate. Flow stress in lower purity tin is higher than that of high purity tin at higher strain rate over $1 \times 10^{-1}\,s^{-1}$, but purity effect on flow stress is very small at lower strain rate below $1 \times 10^{-2}\,s^{-1}$, which is the same with results shown in Fig. 6. Difference in impurity contents between 5N and 4N purities in tin is much smaller compared with that of lead with 5N and 3N purities, which appears to exert very tiny influence on

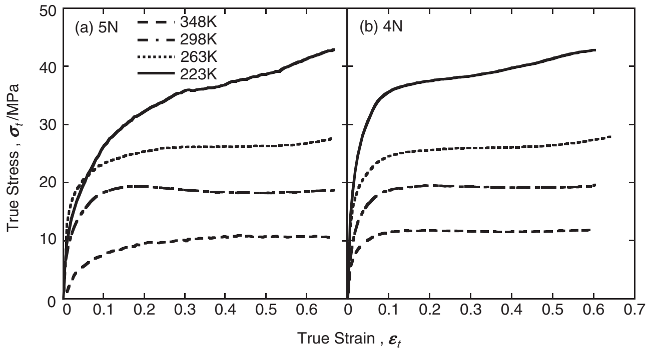

Fig. 6 Variations of Stress-Strain curves with the deformation temperature at strain rate of $2 \times 10^{-3}\,s^{-1}$ in tin. Purity of tin is 5N in (a) and 4N in (b).

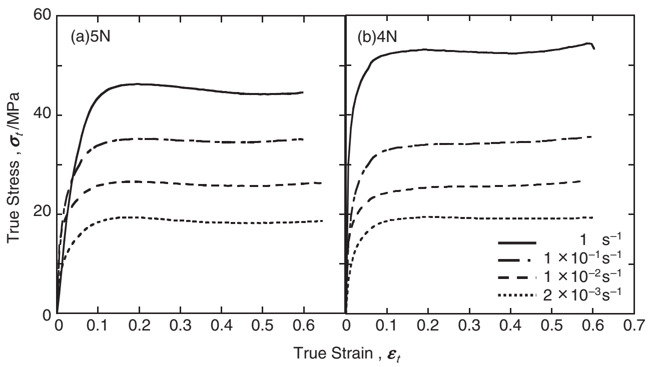

Fig. 7 Variations of Stress-Strain curves with strain rate at the deformation temperature of 298 K in tin. Purity of tin is 5N in (a) and 4N in (b).

flow stress in deformation at slower strain rate. However, in deformation at high strain rate such as $1\,s^{-1}$, the progress of dynamic recovery during deformation may be disturbed by impurities contained in tin with 4N purity, which causes the increase of flow stress. It is found from S-S curves in tin shown in Figs. 6 and 7 that the onset strain of steady state flow stress is not varied by the temperature and strain rate. This is different from results obtained in other dynamic recovery type of metals such as ferritic steel or ferrite in pure iron and HSLA steel, where the onset strain increases with the increase of the steady state flow stress which is caused by the increase of strain rate or the decrease of the temperature.[14,19,20]

Figure 8 shows variations of steady state flow stress with a reciprocal absolute temperature in tin. A linear relationship between them is obtained, and a slope value in this relation is the same in two levels of strain rate. The higher strain rate results in the higher flow stress at a respective temperature, and the effect of the purity level in tin on flow stress is not seen. Tin transforms from α phase with a diamond structure to β phase with a tetragonal structure at the temperature of 286 K, but any significant change in a slope value in relation of flow stress vs. the reciprocal absolute temperature is not observed at this transformation temperature.

3.2 Static restoration behavior in lead and tin

Examples of S-S curves obtained by an intermittent two-stage compression test in lead are shown in Figs. 9(a) to (d). Results obtained by deformation at 273 K and 348 K in lead

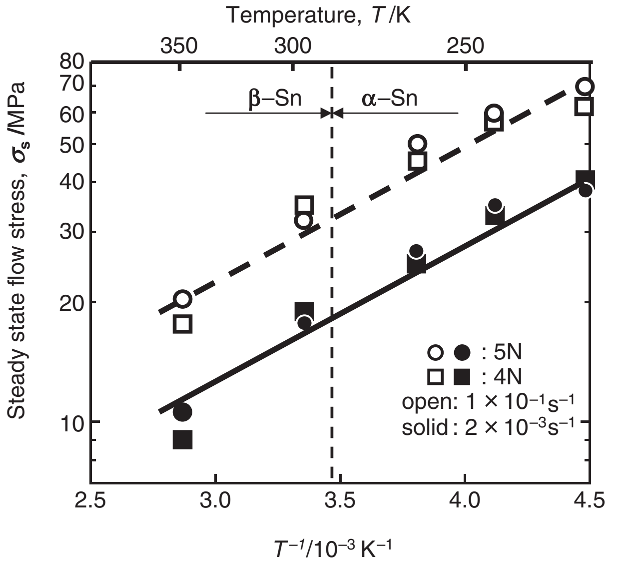

Fig. 8 Variations of steady state flow stress with a reciprocal of the absolute temperature in tin.

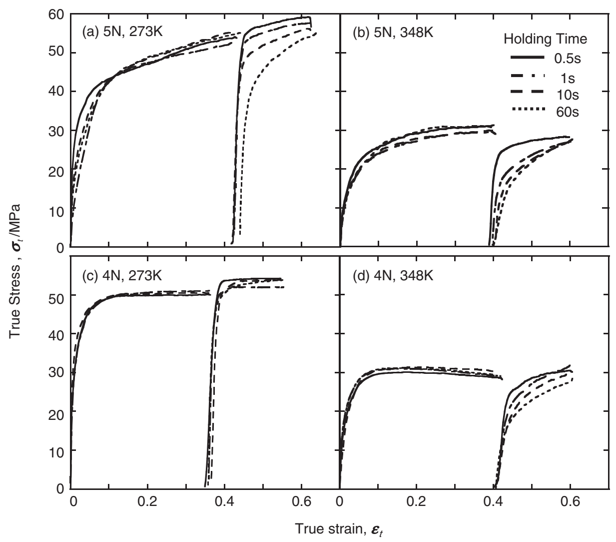

Fig. 10 Stress-Strain curves of tin obtained by an intermittent two-stage compression test. Purity of tin is 5N in (a) and (b), and 4N in (c) and (d). The deformation temperatures are 273 K in (a) and (c), and 348 K in (b) and (d).

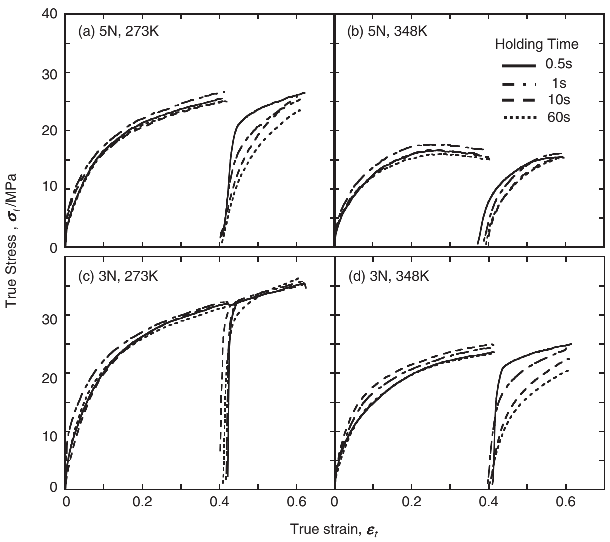

Fig. 9 Stress-Strain curves of lead obtained by an intermittent two-stage compression test. Purity of lead is 5N in (a) and (b), and 3N in (c) and (d). The deformation temperatures are 273 K in (a) and (c), and 348 K in (b) and (d).

with 5N purity are shown in Figs. 9(a) and (b), respectively. The S-S curve obtained in the first stage deformation at 273 K shows continuous work hardening in a strain up to 0.40, and flow stress in this strain (σ_m) is around 26 MPa. This indicates no occurrence of dynamic recrystallization, and so the microstructure in lead evolved immediately after deformation appears to be a fully pan-caked one. S-S curves obtained by the second-stage deformation after holding for several time periods show the continuous reduction in 0.2% flow stress (σ_y). This change of σ_y values indicates the continuous progress of static recovery and recrystallization for holding time periods from 0.5 s to 60 s. On the other hand, the S-S curve obtained in the first stage of deformation at 348 K shown in Fig. 9(b) shows a drop of flow stress after yielding the peak stress, indicating the some amount of progress in dynamic recrystallization. That is, the microstructure evolved immediately after the first-stage deformation appears to be consisting of as-deformed one and the some amount of dynamically recrystallized one. The S-S curve obtained by the second-stage deformation is almost the same among various holding time periods, all showing a large drop in σ_y value from σ_m value. This suggests that static recrystallization has markedly progressed during the shortest holding time period of 0.5 s at this temperature.

Figures 9(c) and (d) show S-S curves in lead with 3N purity obtained by an intermittent two-stage compression test at 273 K and 348 K, respectively. S-S curves obtained by the first stage of deformation show continuous work hardening in a strain up to 0.40 at both temperatures. At the temperature of 273 K shown in Fig. 9(c), σ_y values are the same with σ_m value for a holding time period of less than 10 s, which displays two individual S-S curves just like a single and continuous S-S curve. This indicates no progress in static recovery and recrystallization for the holding time period of less than 10 s at this temperature. The value of σ_y slightly decreases after holding for 60 s. On the other hand, σ_y value continuously decreases with an extension of the holding time period at 348 K as shown in Fig. 9(d), and σ_y value approaches to σ_0 value for the holding time period over 10 s, indicating completion of static recrystallization.

Figures 10(a) to (d) show results of tin with purities of 5N and 4N obtained by an intermittent two-stage deformation. As shown in Figs. 10(a) and (b), S-S curves obtained by the first stage deformation at temperatures of 273 K and 348 K in tin with 5N purity exhibit continuous work hardening in a strain up to 0.40. The value of σ_y at 273 K continuously decreases with an extension of the holding time period. On the other hand, this value at 348 K largely decreases for the holding time of 0.5 s and it approaches to σ_0 value for the holding time of 1.0 s. Results of tin with 4N purity are shown in Figs. 10(c) and (d). The value of σ_0 in this purity of tin obtained by the first stage deformation at 273 K and 348 K

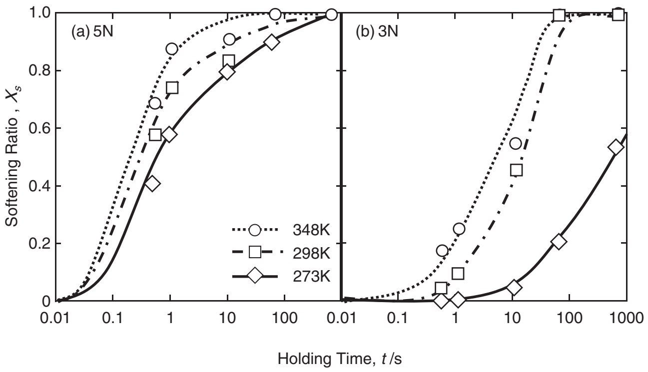

Fig. 11 Variations of softening ratio with the holding time in lead. Purity of lead is 5N in (a) and 3N in (b).

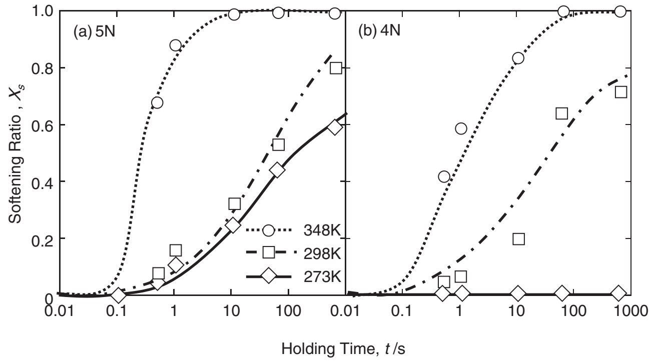

Fig. 12 Variations of softening ratio with the holding time in tin. Purity of tin is 5N in (a) and 4N in (b).

increases compared with that of tin with 5N purity, and the work hardening rate becomes much smaller, flow stress being almost constant in a strain up to 0.40. The values of σ_y at 273 K become slightly higher than σ_m value. However, these are considered to be the same with σ_m value for all holding time periods in the range of the experimental error, indicating no progress of static recovery and recrystallization. This value at 348 K decreases continuously with an extension of the holding time, and it approaches to σ_0 value after holding for 60 s.

Values of softening ratio denoted in eq. (1) were calculated using σ_0, σ_m and σ_y values which were obtained from a respective S-S curve in an intermittent two-stage deformation. Variations of softening ratio with the holding time in lead are shown in Fig. 11. The holding time period was varied in the range from 0.1 s to 600 s. It is found from Fig. 11(a) that softening ratio in lead with 5N purity increases rapidly for very short holding time at all the temperatures, and it reaches to 0.90 at 348 K and 0.60 at 273 K for the holding time period of 1 s. On the other hand, as shown in Fig. 11(b), the progress rate of softening ratio in lead with 3N purity is retarded by almost two orders in the holding time period compared with that of lead with 5N purity. Values of softening ratio at 348 K and 273 K reach to 1.0 and 0.30 for the holding time of 100 s.

Variations of softening ratio with the holding time period in tin are shown in Fig. 12. As shown in Fig. 12(a), the progress in softening ratio at 273 K and 298 K in tin with 5N purity is much retarded compared with that of the same purity lead, while very similar softening behavior is observed at 348 K. On the other hand, softening progress in tin with 4N purity shows marked temperature dependence. Softening in this purity of tin proceeds very similarly to that in lead with 3N purity at 348 K, but no softening variation takes place at 273 K for the holding time up to 600 s.

4. Discussion

4.1 Dynamic restoration behavior in pure lead and tin

In deformation of pure lead and tin in the ambient temperature range, first, both metals showed very strong dependence of flow stress on the deformation temperature and strain rate regardless their purity levels, and secondly, a variation in S-S curve behavior with the deformation condition made clear that lead with F.C.C. structure and tin with a tetragonal or diamond type of a structure were a type of metals belonging to dynamic recrystallization and dynamic recovery, respectively. Regarding on the latter result, the most metals and alloys with F.C.C and B.C.C. structures belong to dynamic recrystallization and dynamic recovery types of metal, respectively, although some exceptions exist like aluminum with F.C.C. structure which is a typical dynamic recovery type of metals. Tin exhibited a dynamic recovery type of the S-S curve in both α and β phases, indicating to have relatively high stacking fault energy in both phases. The former results indicate that the temperature range investigated in this study corresponds to that of hot working in both metals nevertheless it is the ambient temperature range. In general, the temperature region of hot working is characterized by marked dependence of flow stress on the deformation condition and the prominent progress in dynamic restoration process. The authors assumes that the temperature region of hot working may be the region satisfying $T/T_M > 0.55$, where T is the working temperature and T_M is the melting point of metal or alloy. The temperature range investigated in this study was from 223 to 348 K, which yields T/T_M from 0.37 to 0.58 in lead and from 0.44 to 0.69 in tin. That is, the temperature region investigated covers both the warm and hot working temperature regions in both metals. In the following, results of S-S curve behavior and flow stress in the ambient temperature region in both metals are analyzed based on Zener-Hollomon parameter of Z value expressed by eq. (2)[15] and the activation energy (Q) in dynamic restoration process. Relationship among steady state flow stress, the temperature and strain rate, and relationship between Z value and flow stress are shown in eqs. (3) and (4), respectively.

$$Z = \dot{\varepsilon} \exp(Q/RT) \quad (2)$$

$$\dot{\varepsilon} = A\sigma_s^m \exp(-Q/RT) \quad (3)$$

$$Z = A\sigma_s^m, \quad (4)$$

where $\dot{\varepsilon}$ is strain rate, T is the temperature, R is gas constant, σ_s is steady state flow stress, and A and m are constant values which do not depend on the temperature. Figure 13 shows the relationship between $1/T$ and $\ln \dot{\varepsilon}$ in lead with 5N purity and tin with 5N purity under a fixed stress of 12.5 MPa and 16.5 MPa, respectively, and data plots are approximated by a straight line in a respective metal. Activation energy values

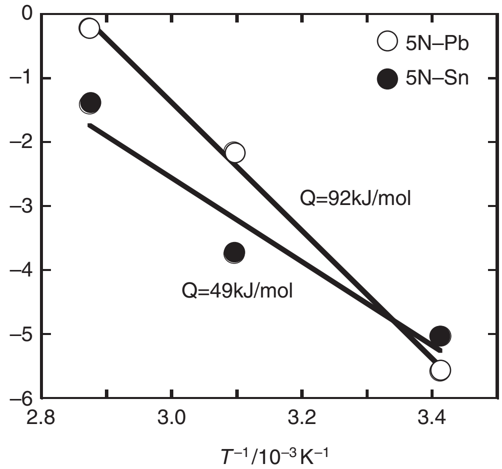

Fig. 13 Arrehenius plots of strain rate under a fixed flow stress of 12.5 MPa in lead and 16.5 MPa in Tin. Purity of both metals is 5N.

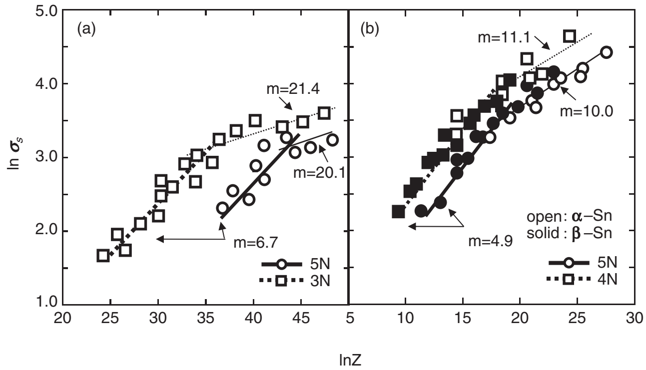

Fig. 14 Variations of steady state flow stress with Z value in (a) lead and (b) tin.

obtained from both slopes in this relationship were 92 kJ/mol in lead and 49 kJ/mol in tin. Activation energies of lead with 3N purity and tin with 4N purity obtained by similar analysis were 119 kJ/mol and 52 kJ/mol, respectively. The activation energy in dynamic restoration process which has been obtained in many metals and alloys was very similar to the activation energy in self diffusion, and the latter values in lead and tin are 102 kJ/mol and 94 kJ/mol, respectively.[21,22) Both values in lead are roughly the same, but the activation energy of tin obtained by the present deformation study was almost one half of the activation energy for self diffusion in this metal. Suh *et al.* studied high temperature creep of tin, reporting that the activation energy for creep at the temperature above and below 423 K($0.84T_M$) in tin was 98 to 118 kJ/mol and 40 to 52 kJ/mol, respectively.[6) Weertman *et al.* also reported the similar change in the activation energy for creep in a single crystal of tin, and the creep activation energy of 46 kJ/mol below this temperature.[9) That is, the activation energy obtained by the present study is consistent with the activation energy for creep obtained in the low temperature region reported by these authors.

The low activation energy for creep in low temperature region in tin had been attributed to high speed diffusion through a dislocation core.[5,6,9) The microstructure evolved by hot deformation in a dynamic recovery type of metal is constituted by subgrains which are formed by dislocation climb.[14) Dynamic recovery process in tin is controlled by such a high speed diffusion as grain boundary diffusion or pipe diffusion.[23,24) Shaby *et al.* reported that the activation energy for dislocation core self-diffusion was a function of the absolute melting point which is shown by eq. (5).[24)

$$Q = 11RT_M \quad (5)$$

The value of Q in tin calculated from this equation is 46 kJ/mol, which nearly coincides with the activation energy value obtained in this study.

The stress exponent of m in eq. (4) is obtained from the slope in relationship between $\ln Z$ vs. $\ln \sigma_s$ using Q values obtained in a respective purity of lead or tin. Figures 14(a) and (b) show variations of steady state flow stress with Z values in lead and tin, respectively, and the reciprocal slope value in a respective straight line is m value. As seen in Fig. 14(a), the slope of the straight line in lead with 5N and 3N purities changes in Z value of around 44 and 36, respectively, and m value in both purities of lead obtained in the region with Z value below these values is about 6.7. In the region with high Z value, m value is over 20. The region of low Z value corresponds to the deformation condition at the high temperature or low strain rate, which induces marked progress in dynamic recrystallization. In these deformation conditions, m value obtained in a dynamic recrystallization type of metals is mostly over 5.0, which coincides with the present result of m value in lead. Variations of steady state flow stress with Z value in tin are shown in Fig. 14(b). The flow stress difference between 4N and 5N purities in tin is much smaller compared with difference between 3N and 5N purities in lead, and difference of α and β phases does not affect flow stress. The slope in both purities of tin changes in Z value of around 18, and m value in the region of low Z value is around 4.9. The most of a dynamic recovery type of metals yield m value below 5.0, and the stress exponent value in tin obtained in the past creep studies were 4.6 to 6.6,[5,8,9) and these results coincide with m value obtained in the present study. S-S curves and their variations with the deformation condition in tin evidently indicate that tin belongs to a dynamic recovery type of metals, and so relatively high m obtained in the present study or other creep studies may possibly associate with very lower activation energy for hot deformation or creep compared with that of self diffusion of tin as noted above, although it is not easy to explain how to relate m value and the activation energy.

4.2 Static restoration behavior in lead and tin

Static restoration behavior in lead and tin investigated by the mechanical test method made clear that the recrystallization temperature of both metals was around the ambient temperature, although it was varied by purity level of both metals. Static recrystallization of lead with 5N purity completed for the holding temperature of 100 s even at the temperature 273 K, and softening ratio of 3N purity lead reached to over 0.5 for the holding time of 600 s at this temperature. On the other hand, the recrystallization kinetics in tin with 5N and 4N purities was slightly retarded compared with lead with 5N and 3N purities, respectively. Consequently, static recrystallization kinetics of tin appears to be similar to that of lead with one grade lower purity.

In general, static recovery and recrystallization kinetics at a particular temperature is affected by both deformation conditions and metallurgical factors. The increase of strain or strain rate, the finer initial grain size and the increase of purity of metals tend to increase the progress rate in static restoration process. The present investigation of static restoration behavior in lead and tin was conducted under a given deformation condition, and the deformation temperature and purity of both metals were primarily varied. The first stage deformation was conducted at relatively high strain rate of $1\,s^{-1}$ and in a strain of 0.40. This deformation condition evolved work hardened and dynamically recovered microstructure in both lead and tin except lead with 5N purity, which yielded partially dynamically recrystallized structure in the first stage deformation. The dynamically recrystallized microstructure is stable under deformation, but unstable after deformation, and so static restoration after deformation may proceed rapidly compared with that of work hardened microstructure. Difference in a variation of softening ratio in lead between 5N and 3N purities shown in Figs. 9(a) and (b) includes this effect beside the direct effect of purity on static recrystallization kinetics. Occurrence of dynamic recrystallization depends on the temperature, strain rate and the amount of strain. It can be noted that lead may be more rapidly statically recrystallized over tin, once dynamic recrystallization occurs partially or fully.

4.3 Substitution of tin for lead as a plug material for seismic isolation

Possibility of substitution of tin for lead in use of a plug material for seismic isolation is discussed by comparison of dynamic and static restoration behaviors in both metals. It was found that dynamic restoration process was different in both metals, but that flow stress or strength at a particular temperature could be controllable by selection of purity level in both metals. Static recrystallization in tin took place at the ambient temperature similarly to lead, and a slight retardation in static recrystallization kinetics in tin was found to be optimized by an appropriate selection of purity level in this metal. Thus, tin possibly substitutes for lead as a plug material for seismic isolation.

There are a couple of concerned matters in substitution tin for lead. As found from comparison of flow stress shown in Figs. 14(a) and (b), flow stress of tin tends to increase over lead in deformation condition with a high Z value such as low temperature or high strain rate. Static recrystallization behavior at the temperature below 273 K in both metals was not investigated in this study, but marked retardation in static recrystallization in the low temperature region may occur in low purity tin as estimated from the result of the temperature of 273 K shown in Fig. 12(b), where no variation in softening ratio was observed in tin with 4N purity. Another concern for this substitution of metals is possibility of brittle facture at the low temperature in tin. While lead with F.C.C. structure has no possibility of brittle fracture, tin with a tetragonal structure has that possibility. Consequently, it appears to need investigations of various properties of tin at the temperature below 273 K, when the ambient temperature in a practical application of a tin plug covers such a low temperature.

Finally, it is important to note about the hot working simulator used in this study. Since this simulator was developed in 1980,[13] it has been used widely in a world-wide and various research sectors as an experimental tool for a basic study of hot deformation strength, thermo-mechanical processing, hot workability and so in a wide variety of metals and alloys. In this study for metals with the low melting temperature, this simulator was reformed to enable a hot deformation study at the low temperature ranged from 223 K to 348 K, and it was confirmed that both of dynamic and static restoration behaviors in this temperature range in lead and tin could be investigated similarly to hot deformation behavior in other metals and alloys with the higher melting temperature. It appears to be very advantageous that deformation study can be performed using the same simulator under a wide deformation conditions, covering both temperature regions above 293 K by heating and below 273 K by cooling, and a wide range of strain rate. Consequently, it was confirmed by the present study that the utilization field of this simulator was further expanded.

5. Conclusion

Dynamic and static restoration behaviors of pure lead and tin with two purity levels were investigated by compression tests in the ranges of the temperature from 223 to 348 K and strain rate from 2×10^{-3} to $1\,s^{-1}$. Static restoration progress with the holding time period was investigated based on softening ratio evaluated by an intermittent two-stage compression test. The following results were obtained.

(1) S-S curve behaviors observed in lead and tin were those of dynamic recrystallization and dynamic recovery types of metals, respectively. The activation energies for dynamic restoration process in lead and tin with 5N purity were 92 kJ/mol and 49 kJ/mol, respectively, and the latter value was almost one half of the activation energy for self diffusion of tin.

(2) Steady state flow stress in lead with 5N purity was lower than that of tin with the same purity, and a reduction of purity from 5N to 3N in lead markedly increased flow stress. Purity difference of tin between 5N and 4N yielded very tiny change of flow stress at strain rate below $1 \times 10^{-1}\,s^{-1}$, but flow stress of tin tended to become higher than that of lead in deformation at the low temperature or high strain rate.

(3) Static recrystallization in lead with 5N purity completed for the holding time of less than 600 s even at 273 K, and it was slightly retarded in tin with the same purity. A reduction of purity in both metals retarded the time period for completion of recrystallization by more than 2 orders.

(4) Consequently, regarding on dynamic and static restoration behaviors, tin with the higher purity grade over lead purity can substitute for lead as a plug material for seismic isolation.

(5) A hot working simulator of THERMEC MASTER-Z in which a reservoir for coolant to cool the specimen was installed in place of an induction heating coil enabled to

conduct compression tests at the low temperatures ranged from 223 to 273 K and at a wide range of strain rate.

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