Investigation of metal-matrix composite containing liquid-phase dispersion

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Abstract
Al-Pb binary system is a suitable model system for testing liquid phase dispersion strengthening in bulk materials for structural applications. Liquid Pb islands can be finely dispersed in still solid Al matrix due to the substantial difference of melting points. The Al-Pb system prepared by means of Equal Channel Angular Pressing (ECAP) process was investigated by Small-Angle Neutron Scattering technique (SANS) which enables in-situ measurement of size and morphology parameters of Pb inclusions at elevated temperatures. It was observed that the lead particles were elongated roughly in the direction of ECAP. During the subsequent in-situ thermal cycle RT-400°C-RT, the elongated Pb particles transformed to nearly spherical shape. The change of scattering contrast during melting of Pb mapped the transform of the confined lead particles to the liquid phase. The center of the transition region is around 342°C (615K) for both the as-cast and the ECAP deformed samples, which is a significant shift with respect to the free Pb melting point 327°C (600K). For the ECAP sample, the transition is not sharp, indicating a broad size distribution of lead particles.

1. Introduction
Over many decades, a large number of investigations were devoted to find ways to harden metallic materials by introducing a dispersion of second phase particles through precipitation or as dispersoids [1]. To obtain maximum strengthening at ambient temperatures, strong particles with large resistance against cutting by dislocations are required. However, at elevated temperatures, this aspect becomes less important as the dislocations are able to circunvent the particles by climb. The theory predicts that soft particles are particularly attractive because they facilitate relaxation of the dislocation stress field at the particle, thus ensuring a large attractive interaction between particle and dislocation [2-4]. In fact, this concept is utilized in potassium doped tungsten wire filaments, where evaporation of potassium leads to the formation of vapor-filled bubbles about 9 nm in diameter at service temperature [5]. Similarly, one would expect effective dispersion strengthening in bulk materials for structural applications if stable voids or a liquid phase could be finely distributed in the solid matrix.

It is, however, not trivial to test this hypothesis, as a fine and stable dispersion of liquid phase in a metal matrix is not easy to obtain. Al-Pb binary system can be a model system in this respect and was chosen to test this hypothesis. Pb (fcc, lattice constant a_Pb = 0.495 nm) has negligible solubility in Al (fcc, a_Al = 0.405 nm) in the solid state and no intermediate phases form in Al-Pb binary system [6]. Importantly, the melting points of Al (T_M=660.32°C) and Pb (T_M=327.46°C) are very different, therefore at high temperatures (>400°C) Pb can remain as a liquid dispersion in solid Al matrix. It has been reported that T_M of Pb increases in the confinement of the Al matrix and this increase is particle
size dependent [7]. The melting of Pb nanoparticles was studied by means of dynamic mechanical analyzer / internal friction measurements [8] and transmission electron microscopy (TEM) [6].

Structural, thermodynamic and diffusion properties of crystalline particles completely embedded in a metallic matrix have been investigated in detail in a variety of homogenous systems of immiscible elements for the past two decades. The monotectic binary Al-Pb system, which has a large miscibility gap in the liquid phase and extremely limited mutual solubility in the solid phase, has been a key-type system for such investigations. Shapes and sizes of nanoscale Pb inclusions and their melting in Al have been studied by several authors, see e.g. [9, 10]. Nevertheless, neutron scattering carrying bulk information was not – to the authors’ knowledge – yet employed.

However, Pb particles are very coarsely dispersed in Al matrix in the as cast Al-Pb system. For any interaction with the dislocations, a fine dispersion of the Pb particles within the grains is needed. Also for mechanical property tests, this dispersion should be available in bulk materials and not only on thin films. Equal Channel Angular Pressing (ECAP) was used to produce a uniform and fine dispersion of Pb in the present study.

As cast and ECAP deformed Al samples containing 4wt.% of Pb were investigated by small-angle neutron scattering (SANS) both ex situ and in situ at temperatures in the vicinity of the Pb melting point. The aim was (i) to test a possibility to investigate microstructure of such system in situ by SANS, (ii) to confirm $T_m$ shift of the confined Pb particles and (iii) eventually conclude on details of microstructure during thermal cycle around solid – liquid transition temperature of lead particles.

2. Experimental

Al-4wt.%Pb system was melted in a vacuum arc furnace (VAF) with non-consumable tungsten electrode. High purity iron free aluminum (99.9995%) and lead (99.8%) was used for melting. It was seen that even a very small Fe impurity in Al resulted in a Fe-Pb eutectic formation in the cast system. The microstructure of the as cast Al-4%Pb is shown in Fig. 1 and it can be seen that the Pb particles are coarse (in μm scale) and dispersed mainly at the grain boundaries and at triple points (i.e. where three grains touch). In order to refine the Pb particle dispersion, cast bars of 10 mm x 10 mm square cross-section 50 mm long were processed by equal-channel angular pressing. ECAP was done in a die with 90° angle ($\Phi$) following the so called route B (sample rotated 90° between each pressing [11]. Load of ~50 kN was needed for the pressing. The bars were smeared with Molykote BR2+ lubricant prior to pressing.

![Figure 1. Dispersion of Pb in the as cast Al-4%Pb system.](image)

Fig. 2 shows scanning electron microscopy (SEM) images of a Al-2.5%Pb system with increasing number of identical ECAP passes (up to 40). It can be seen that the Pb particles are elongated after ECAP. The particle sizes were measured from the micrographs assuming an ellipsoidal shape. The
variation in particle sizes with increasing number of ECAP passes are shown in Fig. 3. It shows that ECAP considerably refines the as cast particle size and also particles are dispersed more homogenously. However, saturation is reached after ~20 passes (in the case of Al-2.5\%Pb system) and further ECAP processing actually increases the particle size. In total, 8 ECAP passes were adopted in the Al-4\%Pb system for optimal refinement. The microstructure after 8 passes is shown in Fig. 4.

ECAP deformed Al-4\%Pb and pure Al samples were investigated by small-angle neutron scattering (SANS) both ex situ at RT and in situ at temperatures in the vicinity of Pb melting point. The measured samples are reported in Table 1.

![Figure 2. Pb dispersion in as cast sample and after different number of ECAP passes in Al-2.5\%Pb system. The particle dispersion is shown with increasing ECAP passes. a) as cast, b) 4 pass, c) 8 pass, d) 16 pass and e) 40 pass.](image)

![Figure 3. Evolution of Pb particle size versus increasing number of ECAP passes in Al-2.5\%Pb system: (a) major axis of the ellipsoid, (b) minor axis of the ellipsoid.](image)
Table 1: Samples used for SANS measurements in different orientations

| Sample                  | ECAP | Measurement mode (ex-, in- situ at HT) | thermal treatment: orientation | Size (Å)          |
|-------------------------|------|---------------------------------------|---------------------------------|------------------|
|                          |      |                                       |                                 | smaller particles | larger particles |
| as cast Al-4%Pb No. 2    | No   | ex situ                               | no thermal treatment 1, 2       | 1340             | 5670             |
| ECAP Al-4%Pb, bar 3      | Yes  | ex situ                               | no thermal treatment 1,2,3      | 1490 (short axis), 1970 (long axis) | 4790 8410 |
|                          |      |                                       |                                 | 1480             | 5270             |
| ECAP Al-4%Pb, bar 3, A   | Yes  | ex situ                               | RT-400°C, 1 cycle, prior to SANS| 1480             | 5270             |
|                         |      |                                       |                                 | 1480             | 5870             |
| ECAP Al-4%Pb, bar 3, B   | Yes  | ex situ                               | RT-400°C, 2 cycles, prior to SANS| 1480             | 5270             |
| Pure Al with ECAP        | Yes  | ex situ                               | no thermal treatment 1, 2       | 1630             | 4770             |
|                         |      |                                       |                                 | 2200             | 7370             |
| As cast Al-4%Pb, bar 2, | No   | in situ at HT, 1 cycle                | RT-400°C, 1 cycle               | 1340             | 5670             |
| ECAP Al-4%Pb, bar 3, A   | Yes  | in situ at HT, 1 cycle                | RT-400°C, 1 cycle               | 1480             | 5270             |
|                         |      |                                       |                                 | 1480             | 5870             |
| ECAP Al-4%Pb, bar 3, B   | Yes  | in situ at HT, 2 cycles               | RT-400°C, 2 cycles              | 1480             | 5270             |
|                         |      |                                       |                                 | 1480             | 5870             |

Orientation 1: beam parallel to ECAP direction
Orientation 2: beam perpendicular to ECAP direction
Orientation 3: beam perpendicular to ECAP direction; turned by 90° (χ angle, i.e. around the ECAP direction) with respect to the orientation 2
Neutron scattering measurements were carried out at the SANS-II instrument of SINQ user laboratory of Paul Scherrer Institute (PSI), Villigen, Switzerland [12]. Samples in the form of cubes (approximately 10 x 10 x 10 mm$^3$) were used and a vacuum furnace was employed for the in-situ heating. The form of the sample enables its rotation by 90° around the sample axes and thus measurements at different orientations with respect to ECAP direction. The sample thickness of 10 mm is neither a problem from the transmission point of view (Al matrix practically does not absorb neutrons), nor from the multiple scattering point of view (scattering contrast is low and thus the multiple scattering does not play a role).

The scattering data were collected at several geometries; the sample-to-detector distance (SDD) varied from 1.2 m to 6 m and the neutron wavelength $\lambda$ from 4.55 Å to 19.6 Å. The full covered range of the scattering vector magnitude $Q = |\mathbf{Q}|$ was approximately $2\times10^{-3}$ Å$^{-1}$ to 0.2 Å$^{-1}$ (i.e. $2\times10^{-2}$ nm$^{-1}$ < $Q$ < 2 nm$^{-1}$), where the magnitude $Q = |\mathbf{Q}| = |\mathbf{k} - \mathbf{k}_0|$, $\mathbf{k}_0$ and $\mathbf{k}$ being the wave vectors of the incident and scattered neutrons, respectively, and $|\mathbf{k}| = |\mathbf{k}_0| = 2\pi/\lambda$ due to the elastic scattering. The measured raw data were corrected for background scattering and calibrated to absolute cross section. Due to the relatively large size of Pb particles (hundreds of nanometers) - from the point of view of SANS - the important measurements were the ones at the lowest $Q$ values, i.e. with SDD equal to 6 m and $\lambda$ = 19.6 Å. This experimental geometry allowed – due to the rather low primary beam intensity – to remove the beamstop without a danger of overloading the detector. The used sample background subtraction ([13], see section 3.4 and scheme in Fig. 1 therein) enables evaluation of the scattered data even in the region of the primary beam. Scattering in or near this region provides information even on the large particles of size several hundreds of nanometers, although no details about size distributions can be extracted. It cannot be excluded, that there exist still larger particles (of micrometer size), scattering of them cannot be, however, distinguished (by scattering curve profile) from the primary beam.

The scattered intensity of the neutrons is proportional to the volume fraction as well as to the scattering contrast ($\Delta p_{\text{SLD}}$)$^2$ (i.e. the square of the difference between scattering-length densities (SLDs) of the particles and the matrix, see e.g. [14]). The SLD of the particles and matrix depends on their composition, which is known precisely in this case, and on their densities ($p_{\text{Al}} = 2.702$ g/cm$^3$ at 293 K, $p_{\text{Pb}} = 11.34$ g/cm$^3$ at 293 K). At RT, the SLD of Pb is $31.02\times10^{9}$ cm$^{-2}$ and SLD of Al is $20.77\times10^{9}$ cm$^{-2}$. Therefore, the scattering contrast at RT ($\Delta p_{\text{SLD}(293K)}$)$^2$ is equal to $1.051\times10^{20}$ cm$^{-4}$.

3. The cause of the possible SANS pattern change

There are expected changes in the mass density and shape of Pb particles during thermal cycle applied on Al-Pb composite (both as cast and ECAP processed). These changes can affect both the measured intensity and also the shape of 2D scattering curves.

The dominant cause for a change of scattered intensity is the change of the scattering contrast. The other possibilities (change of the specific surface, change of the size of particle due to the thermal expansion during heating or change of the size during phase transformation) are estimated to be small in comparison to this effect.

Change of the scattering contrast is connected to the density changes in dependence on temperature due to (i) thermal expansion coefficient difference (23.1 $\mu$m$^2$·m$^{-1}$·K$^{-1}$ for Al and 28.9 $\mu$m$^2$·m$^{-1}$·K$^{-1}$ for Pb at 25°C) and (ii) step-like change of average spacing of Pb atoms during phase transformation (density of liquid Pb at melting point is 10.66 g/cm$^3$).

The scattering contrast change due to the different thermal expansion coefficients of Al and Pb is estimated to be altogether not larger than 3% in the investigated high temperature range 300-400°C.
On the other hand, a stepwise expansion of Pb inclusion at its melting temperature (Fig. 5) causes large volume (and thus also density) change between the solid and liquid Pb. This volume change needs to be accommodated by plastic or elastic deformation (or both) of the surrounding Al matrix. It may eventually also need diffusional rearrangement of atoms in the Al matrix in case of particle shape changes. It is known [6] that thermal vacancies become mobile in Al at temperatures above 200°C, which enables such rearrangement.

![Figure 5. Scheme of the Pb particle expansion on solid-liquid transformation.](image)

For pure Pb not confined into any matrix, the density change would be of the form shown in Fig. 6. The step in the dependency is at the melting point. The scattering contrast then changes (also shown in Fig. 6) by 28% in the measured temperature range RT-400°C. Out of this change, 19% is due to the step-like change at melting point and 9% due to thermal expansion coefficient difference. As lead particles are confined in the solid matrix, their expansion to the full extent on heating may not occur. Nevertheless, also partial thermal and transformational expansion leads to a certain lowering of Pb density with respect to the unmelted particle at RT. Therefore, although the change of scattering contrast of Pb confined in Al can be expected to be lower than for free Pb, there should be a change of...
the contrast and at least its stepwise part at the melting point should be visible in SANS intensities. The above mentioned magnitudes of the scattering contrast change can be taken as an upper limit of the expected scattering intensity change.

It should be also pointed out that different sizes of confined particles could possess slightly different melting points [6, 7] which would cause smearing of the step in the scattering contrast – and thus in the scattered intensity – dependency.

The quantity, which is reported later in the text and figures, is “volume fraction times the scattering contrast”. Although scattering contrast is known at RT, its evolution with temperature T is unknown (a precise measurement of lattice parameter evolution with temperature using diffraction for both Al and confined Pb would be necessary [15] to determine this). Therefore, we can report only the compound parameter $c_{v_{f}} \times (\Delta \rho_{SLD}(T)^{2}/(\Delta \rho_{SLD}(293K))^{2}$ where $c_{v_{f}}$ is the volume fraction of Pb particles seen by SANS. It is normalized in such a way that the parameter magnitude at RT is equal to the volume fraction. The further evolution of the compound parameter at higher temperature is, however, practically exclusively driven by the scattering contrast change with temperature.

On cooling back to RT, Pb particles should shrink on liquid-solid phase transformation. It could – in principle – produce voids between the Pb particle and the Al matrix in case the matrix deformation on heating was (at least partially) plastic.

The original shape of the solid lead inclusion is ellipsoid rather than sphere in the case of ECAP deformed material. Then, the shape of Pb particles can possibly change during the phase transformation (Fig. 7) as a consequence of Pb expansion described in the previous paragraphs. Due to this expansion, hydrostatic pressure on Pb particles increases and they have a tendency to change shape from ellipsoidal to spherical. This change in particle shape would influence the shape of 2D SANS profile.

![Figure 7. Scheme of the Pb particle shape change in ECAP deformed sample on solid-liquid transformation.](image)

4. RT measurements at several orientations

4.1. The as cast sample

SANS measurement in two orientations on the as-cast sample exhibited no significant anisotropy. The scattering can be modeled well by two types of particles, which are most likely the parts of the same size distribution. The scattering near $Q=0$ is well described by spheres of mean diameter 5670 Å. In order to describe well the scattering also at larger $Q$-values, the model has additionally included spheres of mean diameter 1340 Å (fitted sizes are reported in Tab. 1). We decided to split the size distribution into these two parts in order to observe easily size changes during in-situ thermal cycle (see later in the text) by observing volume fraction evolutions of these two fixed particle sizes. The data does not allow detailed extraction of more parameters (e.g. size distribution width).
4.2. The ECAP samples

The ECAP samples were tested at different orientations both before and after in situ heat treatment. Before heat treatment, the ECAP sample shows significant anisotropy, mainly when the ECAP direction is perpendicular to the neutron beam. It means that the Pb particles are rather prolate ellipsoids than spheres in this case. The size of the individual axes found by SANS is written in Tab. 1.

![Figure 8. SANS data (differential cross section dΣ/dΩ(Qx,Qy) measured at RT for ECAP Al-4%Pb sample at low Q-values before the in-situ thermal cycle. The grey scale map (logarithmic scale) shows measured 2D data versus Qx and Qy after raw-data treatment and the white equi-intensity lines depict the fitted curves. The data for three orientations (definition see Tab. 1) of the cube-shaped sample are shown. The white parts of the PSD were masked as there were regions on the PSD with blind spots of the detector efficiency. Leaving the spots unmasked would significantly harm the evaluation.](image)

![Figure 9. The depiction of the models resulting from the optimum fits for the ECAP Al-4%Pb sample. The microstructure was modeled in 3D. The used depiction procedure resumes imaging through transmission electron microscopy. The model is imaged using slices of the 3D model with the thickness equal to the mean precipitate size. The model of smaller (left) and larger (right) particles is projected to 2D and a partial transparency of the particles is enabled. As the precipitates are of various sizes and positions, this procedure results in the different grey color brightness of the imaged particles. Ellipsoidal shape of particles can be recognized.](image)
The fitting of the 2D SANS patterns for three different orientations (Fig. 8) was performed at once (i.e. a 3D fit was in fact carried out). Therefore, it was found by SANS that the larger axis of the ellipsoid does not lie exactly in the direction of ECAP, but it is tilted in two directions always by approximately 15°. This fact explains orientation of anisotropy in the 2D SANS data measured at two different orientations of the sample with neutron beam perpendicular to ECAP direction (Fig. 8, orientations 2 and 3). The third orientation of the measured ECAP sample (beam parallel to ECAP) provides nearly isotropic scattering (Fig 8, orientation 1). The models of both types of particles are displayed in Fig 9.

The measurement at two sample orientations on the ECAP sample after in situ thermal cycle (RT-400°C-RT) (Fig. 10) shows that the scattering is again nearly isotropic. However, the orientation 2 still exhibits small elongation of the pattern in the direction perpendicular to ECAP. This is possibly caused by small retained elongation of the large Pb particles along ECAP. Nevertheless, the change from the large anisotropy before thermal treatment to the small anisotropy after the treatment means, that the solid-liquid transition of Pb particles strongly influenced the shape of the Al cavity in which Pb is confined.

Figure 10. SANS data \(\frac{d\Sigma}{d\Omega(Q_x,Q_y)}\) measured at RT for ECAP Al-4%Pb sample at low \(Q\)-values after the in-situ thermal cycle.

4.3. The pure Al sample after ECAPs

The last sample which was tested ex situ was pure Al (i.e. without any Pb) after ECAP procedure. Surprisingly, it exhibits also rather strong scattering, moreover anisotropic (see Fig. 11). As the original material contained no impurities, the only explanation is that the elongated pores were created in the material during the ECAP procedure. Unfortunately, there was no virgin Al sample without the ECAP procedure measured by SANS, therefore this can be taken only as a speculation. Nevertheless, any other explanation for the scattering would be difficult to comprehend. If we assume scattering contrast of pores, then the measured volume fraction would be 0.06% for smaller (135 nm) and 0.11% for larger (570 nm) pores. At this point, it cannot be excluded that such pores are present also in the ECAP Al-4%Pb sample. However, it is highly probable that all such potential pores are filled by lead in the ECAP Al-4%Pb sample. Otherwise, the high temperature scattering (particularly around the Pb melting point) which will be described in the next section would be different.
5. In situ measurements

All the in situ measurements were performed in orientation 1, i.e. beam parallel to the ECAP direction. The thermal cycle used for in situ SANS measurement was basically RT-300°C -400°C-RT, where the temperature increase was paused with 5 or 10K temperature steps between 300 and 400°C in order to collect the neutron scattering data.

5.1. As cast Al-4%Pb sample

From the evaluation of the in-situ measurement (RT-400°C-RT cycle) of the as cast Al-4%Pb sample, the temperature dependence of volume fraction times scattering contrast of Pb particles can be deduced (Fig. 12). The following conclusions can be drawn:

Figure 11. SANS data (dΣ/dΩ(Q_x,Q_y)) measured at RT for pure Al after ECAP at low Q-values.

Figure 12. The temperature dependence of volume fraction times scattering contrast of the as cast Al-4%Pb sample during the thermal cycle.
1) The total amount of Pb particles is the same before and after in situ thermal cycle, which is in accordance with the expectations.

2) The total amount of Pb particles times scattering contrast is roughly constant during heating RT -> 613 K (340°C). A small decrease in this temperature range is possibly due to the small decrease of scattering contrast.

3) However, the proportion between smaller and larger particles changes: amount of smaller particles decreases while amount of larger Pb particles increases during heating to 613K (340°C). Small increase of size of Pb particles due to the thermal expansion is not the cause for this effect because then a corresponding decrease in size during cooling would also be present (i.e. such effect would be reversible). However, it is not the case: the amount of small particles is different at RT before and after the cycle. The same is valid for large particles. Further, when we exclude dissolution of smaller Pb particles (remembering Al and Pb are immiscible), then a possible explanation is joining of the very neighboring particles. Another, even more probable explanation, is smoothing of the surface of particles (wrinkles smoothing) which increases scattering to low angles on expense of large-angle scattering. It can be easily accommodated in the matrix as thermal vacancies are mobile in Al above 200°C [6].

4) Between 613K (340°C) and 618K (345°C), a step-like decrease of the scattered intensity occurs. It is due to the sudden scattering contrast change on the solid to liquid transition. Pb expands in a stepwise manner at this point and its scattering length density is lowered. Temperature $T_M = 615\pm3$K (342±3°C) can be thus deduced as the melting point of Pb in confinement in Al. The observed melting is rather sharp: transformation occurs over a narrow temperature range (within 5K). The large majority of Pb particles melted within the given range 613 to 618K (340-345°C). The temperature of the transformation is 15K higher than for the free lead, and agrees with previous observations on confined Pb particles [6, 7].

5) The total volume fraction of Pb after the thermal cycle returns back to the original value observed before thermal cycle. There is also no indication of creation of additional scattering intensity at large $Q$-values which could indicate creation of small voids at Al-Pb interface. Therefore, the Al matrix deformation on heating was highly probably only elastic and no voids were thus produced on cooling.

5.2. Al-4%Pb ECAP sample, one thermal cycle

The same evaluation was performed for Al-4%Pb ECAP sample which underwent one thermal cycle (RT-400°C-RT). The resulting volume fractions are plotted in Fig. 13.
The amount of small particles at RT before the in-situ cycle is larger than for the as-cast sample. Nevertheless, the overall character of the volume fraction evolution with temperature is the same as for the as-cast sample. There is again a visible change of proportion between small and large particles up to the melting point very similar to the one observed by the as cast sample. The amount of small particles is also different at RT before and after the heating cycle. The same is observed for large particles as well.

However, there exist some differences in details between the ECAP and the as cast samples. Firstly, on temperature increase, there is no visible step down observed in the “volume fraction times scattering contrast” parameter of large particles in ECAP sample (blue line and triangles in Fig. 13) around the melting point. This parameter is nearly constant above 607K. It differs from the large-particle line for as cast sample in Fig. 12. It means that - for the ECAP sample - the increase in volume fraction of larger particles (or wrinkles smoothing) above 607K still acts and completely balances (in terms of the scattered intensity) the gradual decrease of the scattering contrast of large particles during melting.

Secondly, the change of scattering contrast in the vicinity of melting point is not as sharp as in the as cast sample. This can be seen clearly in Fig. 14, where both the as cast and the ECAP data are presented. In the ECAP sample, the transition region ranges from 607 to 633K (334-360°C), which is a 26 K broad range and is significantly broader (probably due to the broader Pb particle-size range) than the transition region of the as cast sample (5K). The center of the transition region is at 343°C (616K), i.e. practically at the same position as the center for the as cast sample (342°C or 615K) and thus 16K higher than melting point of free Pb.
The stepwise change of the scattering contrast which can be deduced from Fig. 14 is around 17\% for the as cast and around 22\% for the ECAP deformed sample. Taking into account the experimental uncertainties, this is in good agreement with the theoretical value for scattering contrast change of a free lead particle (19\%) shown in Fig. 6. This agreement means that – although confined in Al matrix - the Pb particle expands to its full extent (i.e. as a free particle) on the solid-liquid transition.

5.3. Al-4\%Pb ECAP sample, two thermal cycles

In order to see evolution of the microstructure during the thermal cycling, two cycles RT-400°C-RT-400°C-RT were applied on another ECAP deformed Al-4\%Pb sample. For this sample, however, less number of points on the temperature curve was measured due to the time restrictions. The evolution of the volume fraction of Pb particles during cycling is plotted in Fig. 15. It can be seen that the first cycle corresponds well to the previous Al-4\%Pb sample (Fig. 13), also with respect to the proportion of smaller and larger particles. The second cycle, however, does not follow the same dependency. After the second cycle, the ratio between smaller and larger particles volume fractions returns to the same value as before the second cycle. That means that – unlike in the first cycle - there is no growth of particles (or smoothing of wrinkles) in the second cycle (within measurement errors) and that the microstructure is reversible. The largest influence on the morphology can be thus ascribed to the first cycle. The following thermal cycling would probably lead to much slower changes in the microstructure, as is usual in any cycling testing of metallic materials.
6. Conclusions

2D SANS data for the as-cast Al-4\%Pb sample shows no anisotropy. Anisotropy of Pb particles shape can be observed only after severe plastic deformation (ECAP). The lead particles were elongated roughly in the direction of ECAP.

Elongated pores in pure Al after ECAP processing were detected. They are of similar size and volume fraction like Pb particles in Al-4\%Pb ECAP sample. It indicates that empty cavities are always created during ECAP in aluminium, but - when Pb is present - this element fills these empty spaces.

During the in-situ thermal cycle RT-400\°C, morphological transformation of elongated Pb particles to nearly spherical shape during the solid-to-liquid transition of Pb for ECAP Al-4\%Pb can be deduced.

On heating up to the melting point, the increase of volume of large particles at the expense of smaller inhomogeneities can be observed. It is caused either by joining of the next neighboring particles or by smoothing of the surface of particles (wrinkles smoothing).

The change of scattering contrast during melting of Pb maps the transform of confined Pb particles to the liquid phase. The center of the transition region is around 342\°C (615K) for both as cast and ECAP deformed samples. This would mean a significant shift with respect to the free lead melting point (327\°C or 600K). Nevertheless, it is not surprising as such effect was already observed for confined particles [7].

For the ECAP sample, the transition is not sharp: 26 K broad range indicates that there is a broad size distribution of Pb particles. The inclusions of different sizes exhibit different melting points, which then gives rise to the scattering contrast change within the 26 K broad range. Also this is a confirmation of the previous observations, i.e. the dependence of the melting point of confined Pb on the particle size [7]. On the other hand, the transformation solid-liquid seems to be sharp for the as cast sample, probably due to the narrower size distribution.

Although confinement has an effect on the temperature of phase transformation, it seems to have no effect on thermal expansion of the lead particle during solid-liquid transformation. The Pb particle expands to its full extent (i.e. as a free particle).

From SANS data, it can be also deduced that the Al matrix surrounding Pb particle undergoes no plastic deformation. However, for the ECAP sample, there is a change of Al cavity shape on the phase
transform which remains unchanged on cooling. It would indicate that some diffusional process takes place at HT in Al surrounding the Pb particle which results in a permanent change of the cavity shape.

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