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Lead–bismuth eutectic recrystallization studies for the Megapie target

A. Zucchini [a], P. Agostini [b], E. Baicchi [b,*]

[a] *ENEA, FIS/MET, Centro Ricerche 'E.Clementel', via Don Fiammelli 2, 40128 Bologna, Italy*
[b] *ENEA, FIS/ING, Centro Ricerche Brasimone, 40032 Camugnano (Bologna), Italy*

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Abstract

The expansion behaviour after freezing of the lead–bismuth eutectic (LBE) with 44.5% lead and 55.5% bismuth is described according to the reported theory. The issue of the vessel structural integrity after LBE recrystallization was dealt with by experimental and numerical studies performed in the frame of the Megapie (Megawatt Pilot Experiment) project. We have identified the important elements which, in the case of LBE solidification inside the Megapie target, play a role in the reduction of the possible vessel over-stressing; among them, the LBE yield strength has been tested under significant experimental conditions. The resulting suggestions can also be related to the design and to the freezing procedures for other LBE technology facilities.

1. Introduction

In 2001, some structural damage to LBE vessels was observed at ENEA (Italian Board for New Technologies, Energy and the Environment) due to ageing of the solid alloy inside them. Considering the Italian plans to operate large LBE facilities, the necessity of performing in-depth studies to deal with recrystallization problems was envisaged. For these reasons, a study of the literature was carried out followed by related experimental tests and numerical modelling referring to the Megapie project as a first application. Different experimental studies have been performed by PSI (Paul Scherrer Institut, Villigen, Switzerland) in parallel [1–3].

2. Chemistry of the phenomenon

As reported in the Russian literature [4], recrystallization is an atomic-level phenomenon which tends to an equilibrium state. Also in our case, different phases are segregated in adjacent grains and form crystal cells with different interatomic distances. With varying temperature, atoms can migrate through grain boundaries and pass from one to the other crystalline form. After solidification, LBE presents a mixture of two phases (Fig. 1):

- a β-phase (intermetallic compound),
- a γ-phase (solid solution of Pb in Bi).

The excess γ-phase precipitates with time and at constant temperature. This gives rise to a volume increase, as this phase is richer in bismuth, which tends to increase the atomic distances when passing from the liquid to the solid phase. The precipitation of the γ-phase continues

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[*] Corresponding author. Tel.: +39 534 801276; fax: +39 534 801225.
E-mail address: elio.baicchi@brasimone.enea.it (E. Baicchi).

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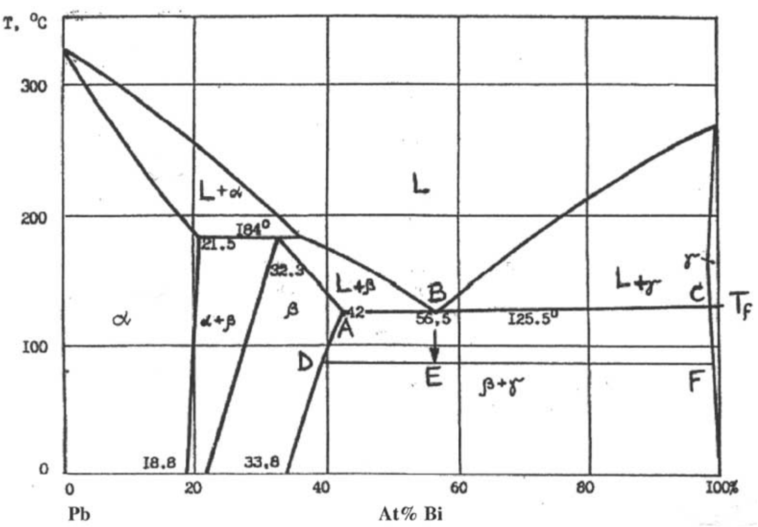

Fig. 1. Phase diagram of the lead–bismuth alloy.

at each temperature until the equilibrium condition is asymptotically reached. The equilibrium condition is essentially determined by the position of the phase boundary between the β and β + γ-regions of the Pb–Bi phase diagram. The temperature effect accelerates the process which corresponds to a volume increase measured by several researchers [5]. Considering all the cooling procedures so far tested, the maximum observed asymptotic volume increase was over 1% [1,4].

With reference to Fig. 1, it can be seen that the content of the γ-phase at the eutectic temperature T_f is AB/AC ≈ 25% and at the generic temperature T it is DE/DF ≈ 29%. Since the slope of the interface line between the γ- and β + γ-regions is steeper than the interface line between the β and β + γ-regions, it can be seen that DE/DF > AB/AC. Consequently, the excess γ-phase precipitates with time and at a constant temperature $T < T_f$.

3. Stress analysis of LBE expansion

The main concern about the freezing of LBE inside a vessel after an experiment such as in the Megapie project (Fig. 2) is related to the risk of a rupture of the target hull when the frozen LBE starts to expand by recrystallization.

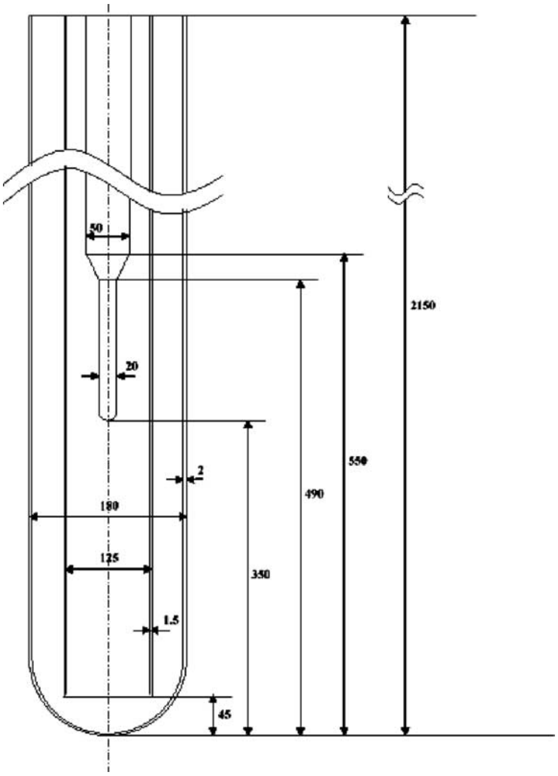

Fig. 2. Geometry and dimensions of the model.

A study of the parameters affecting the stress level in the vessel produced by expansion of the solidified LBE was necessary. The stress analysis of the target during possible freezing procedures has been performed with the FEM code ABAQUS. The lower part of the target including the internal guide tube (A), which separates the upward flow (B) in the centre of the target from the downward flow (C) in the outer annulus, and the central instrumentation rod (D) have been simulated in the 2-D axial-symmetric FE model shown in Fig. 3. The mechanical model is loaded with an isotropic expansion (linear strain ε_0) applied to the LBE solidified in the bottom of the target, generated by means of a fictitious increase of the temperature of the layer and an appropriate thermal expansion coefficient. The stress analysis is based on full elastic–plastic FEM simulations, with elastic-perfectly plastic materials (no strain hardening). The guide tube is assumed to be of AISI 316 stainless steel and the target hull of T91 martensitic steel (French designation). The material properties assumed in the analysis are: $E_{316} = E_{91} = 200\,\text{GPa}$, $E_{\text{LBE}} = 2.5\,\text{GPa}$; $\nu_{316} = \nu_{91} = 0.3$, $\nu_{\text{LBE}} = 0.38$; $\sigma_y^{316} = 400\,\text{MPa}$, $\sigma_y^{91} = 550\,\text{MPa}$, $\sigma_y^{\text{LBE}} = 1\text{–}4\,\text{MPa}$, where E = Young modulus; ν = Poisson modulus; σ_y = yield stress.

The results are plotted as stress distributions in the model vs. the expansion (ε_0) of the LBE layers. The model for the behaviour of the LBE–structure interfaces is a non-slip (perfect bond) contact that was chosen because of the lack of available experimental data for the friction coefficient under the actual experimental conditions. This approach is conservative because any constraint to axial LBE expansion, such as higher shear stresses at the interfaces, will increase its radial expansion and consequently the stress state in the containment structures.

The stress analysis shows that the field of stresses in the Megapie target vessel is mainly affected by the height of the solid LBE level, the degree of expansion of the contained alloy, the yield stress of the solid LBE and the presence of internal structures.

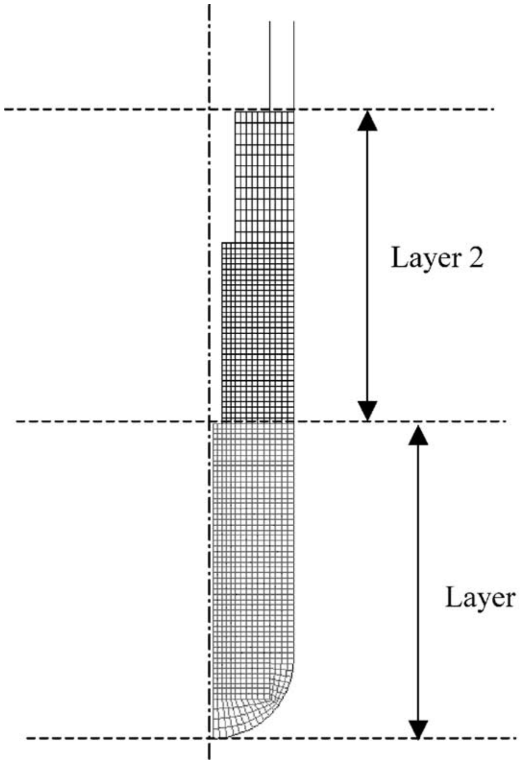

Fig. 3. FEM mesh of Megapie target. The two solidification layers are shown in different grey levels; the presence of internals can be deduced from different mesh.

The height of each solid LBE layer (Fig. 3) in the container turns out to be a significant parameter which needs to be accurately chosen to avoid over-stressing of the vessel. We assume that the liquid volume resting above the level of the solid substrate is free to expand towards the top of the vessel without any constraint. It was proposed in case of tall containers, to carry out freezing of the LBE in consecutive batches of appropriate height, a procedure which was also numerically simulated. In the case of the Megapie target it has been found that the maximum stress at the bottom (or in the window) of the target varies from 230 to 300 MPa, assuming 1 MPa of LBE yield strength, by increasing the solid LBE height in the first expansion step from 350 to 550 mm. The results of the two-step simulations are drawn together in Fig. 4, where the left side shows the Mises stresses behaviour during the expansion of the first layer (ε_0^1) in different positions of the Megapie target, the right side the evolution of the same stresses through the subsequent expansion of the second layer (ε_0^2). The results are in agreement with the concept of vertical extrusion of the radially constrained LBE mass: a larger thickness of solid LBE means a higher shear force opposing vertical movement and therefore higher internal stresses at the bottom. Fig. 4 shows also that the stresses in the window increase again at the end of the two-step analysis. In general, the stress level in a layer can still be affected by the expansion of a second LBE layer above it.

The degree of expansion of the recrystallization process depends on its duration and temperature. It is evident from Takeda's experiments [1] that a first period of faster expansion which ranges from 2 h (110 °C) to three months (20 °C) is followed by a second period of lower expansion rate. In any case the final volume expansion hardly exceeds 1.3% in one year. The assumed volume expansion was 1.2% in the numerical study reported in Fig. 3, for the case of the Megapie target. It has also been found that the maximum stress of the Megapie window would increase by about 15% if the final expansion is 1.5% instead of 1.2%.

The LBE yield strength turned out to be a very important parameter. Figs. 5 and 6 show the Mises stress at two different positions of the window for LBE yield stress in the range between 1 and 4 MPa. The calculated maximum stress increases from 230 to 440 MPa. A higher yield stress means that the internal forces needed to extrude axially the material in a plastic flow are higher, consequently increasing the load on the containment structures. According to Takeda's experimental tests [1] the LBE free strain rate is affected by temperature, while Dai [3] has found that the LBE yield stress is a function of temperature and strain rate. The

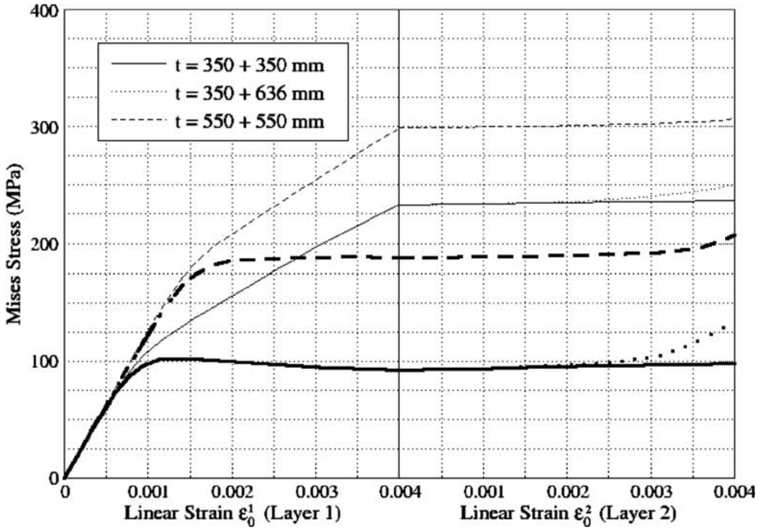

Fig. 4. Influence of the solid LBE height. $\sigma_y = 1\,\text{MPa}$, $\varepsilon_0 = 0.4\%$, where h_1 and h_2 are the heights of two LBE layers frozen and solidified in sequence from bottom to top; σ_y is the LBE yield strength; ε_0 is the linear expansion ($\varepsilon_0 = 0.4\%$ corresponding to 1.2% volume expansion). The thick lines refer to the junction between cylindrical and hemispherical vessel parts. The thin lines refer to the centre of the hemispherical shell.

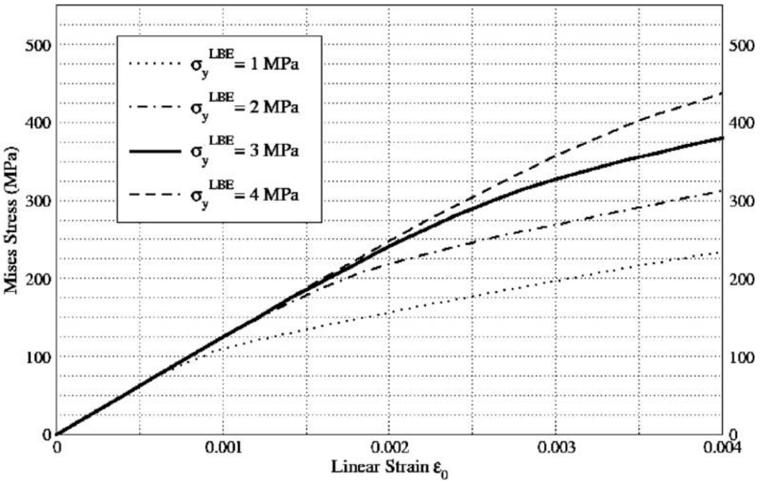

Fig. 5. Influence of the yield stress. Stress at the junction between cylindrical and hemispherical vessel parts with $h_1 = 350\,\text{mm}$, $\varepsilon_0 = 0.4\%$, where h_1 is the height of the bottom LBE layer; σ_y is the LBE yield strength; ε_0 is the linear expansion ($\varepsilon_0 = 0.4\%$ corresponding to 1.2% volume expansion).

LBE yield stress could therefore provide a means of reducing the stress level in the walls by controlling the temperature of the solidified LBE. Such a possibility suggested further numerical and experimental research work, whose results are presented in Section 4. The presence of internal structures which interrupt the extrusion flow path, such as the central instrumentation cluster, clearly results in a wall stress increase, as verified with the stress analysis simulation.

Further considerations involve the shrinkage and thermal expansion. During the transition from liquid to solid, a significant shrinkage (5%) takes place; unfortunately the voids are concentrated in the centre of the free surface and do not allow a significant volume recovery during recrystallization. The linear thermal expansion coefficient of LBE at 100 °C is $21 \times 10^{-6}\,\text{K}^{-1}$, while that of stainless steel is $16 \times 10^{-6}\,\text{K}^{-1}$; this means that the contraction of LBE during cooling is larger than

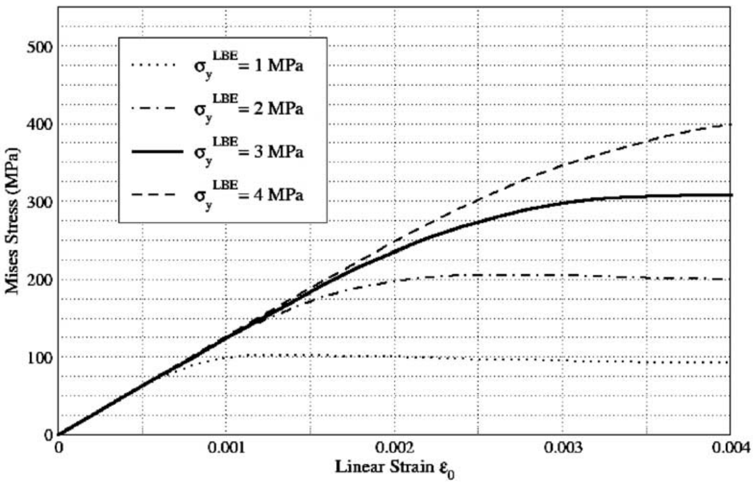

Fig. 6. Influence of the yield stress. Stress in the centre of the hemispherical shell with $h_1 = 350$ mm, $\varepsilon_0 = 0.4\%$, where h_1 is the height of the bottom LBE layer; ε_0 is the linear expansion ($\varepsilon_0 = 0.4\%$ corresponding to 1.2% volume expansion).

that of its container and 0.15% of the volume becomes available after reaching room temperature. Such a value is about one-tenth of the volume expansion due to recrystallization.

4. Experimental tests

The numerical observation about this parameter can be explained by considering that the plastic flow toward the free surface is made easier and takes place without exerting large forces on the walls when the LBE is very 'soft' (lower yield stress). However, when the LBE is 'hard' (higher yield stress) its flow towards the top requires large internal forces. These forces can definitely damage the vessel. For this reason it would be useful to deal with 'soft' LBE during freezing – de-freezing operations. To establish quantitative ranges for the calculations, the execution of some tests of LBE specimens at controlled temperature was compulsory.

Compression tests were preferred for two reasons:

- there is a better correlation of the results, since in the actual case the LBE is compressed;
- it was easier to fabricate, control and test the specimens.

The interface between specimens and testing machine was refurbished following the ASTM standard E 9-81 for compression tests [6–8]. Each specimen was a small cylinder of LBE obtained by the following procedure: pouring the liquid alloy at about 180 °C into glass tubes (moulds), breaking these tubes after solidification, cutting the metal rods into pieces and polishing the latter at both ends. Each specimen was 26 mm in diameter and 20 mm long. During each test the LBE specimen was immersed in diathermic oil, whose temperature was continuously monitored by thermocouples. The comparison test parameter was the 0.2% yield strength recorded as an average value of five valid tests in each condition. Experiments have shown that the chosen parameter is strongly influenced by the test temperature, as illustrated in Fig. 7.

The second parameter affecting the LBE yield strength is the strain rate. When this parameter is combined with high temperatures close to the melting point, there is a complex micro-physical phenomenology involving relaxation, phase segregation, matrix deformation and grain boundary sliding. The net result is a positive strain rate sensitivity of the material, which is clearly shown in Table 1.

It should be noted that the importance of the LBE creep increases dramatically with temperature: according to the Russian literature [4] the creep extent above 70 °C is about four orders of magnitude higher than at 0 °C. The velocity of stress application in the actual case is close to the strain rate of the free LBE expansion. The maximum LBE expansion rate, from Takeda's experiments, was about $5 \times 10^{-6}\,s^{-1}$ at 110 °C. Since the strain rate effect of a constrained expansion has not yet been studied, we assume the former figure as the most realistic strain rate for laboratory compression tests.

The third parameter, whose influence was evident, is the ageing time after solidification. Two different sets of compression specimens were tested at $5 \times 10^{-6}\,s^{-1}$ strain rate and 90 °C and 110 °C (Figs. 8–11). The first set of specimens had been produced three months before testing, namely the 'aged' condition. The second set had

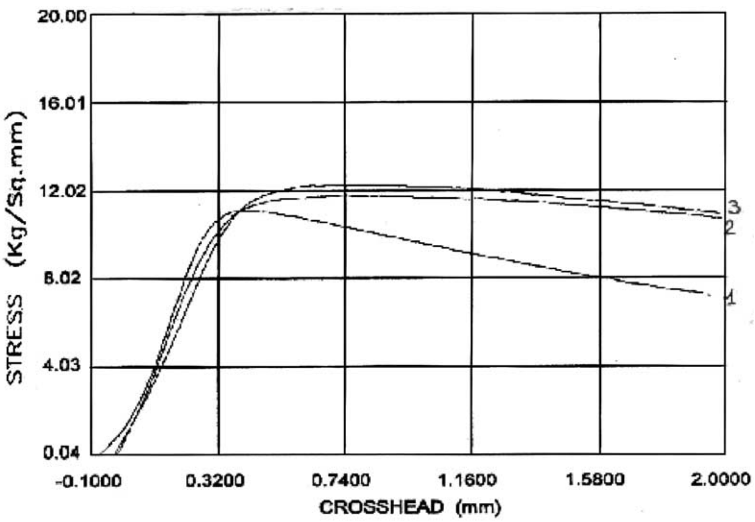

Fig. 7. Influence of temperature in the stress–strain diagram of Pb–Bi. Diagram No. 1 was obtained at 110 °C. Diagram No. 2 was obtained at 100 °C. Diagram No. 3 was obtained at 90 °C.

Table 1
Positive strain-rate sensitivity of the material and comparison between 'fresh' and 'aged' samples

Strain rate (s^{-1})	Temperature (°C)	Maximum age	0.2% Yield strength (MPa)	Fresh/aged
5×10^{-6}	90	3 months	6.1	Aged
5×10^{-6}	110	3 months	3.3	Aged
5×10^{-6}	90	5 h	5.2	Fresh
5×10^{-6}	110	5 h	3.0	Fresh
1×10^{-4}	86	3 months	11	–
1×10^{-4}	104	3 months	8.8	–

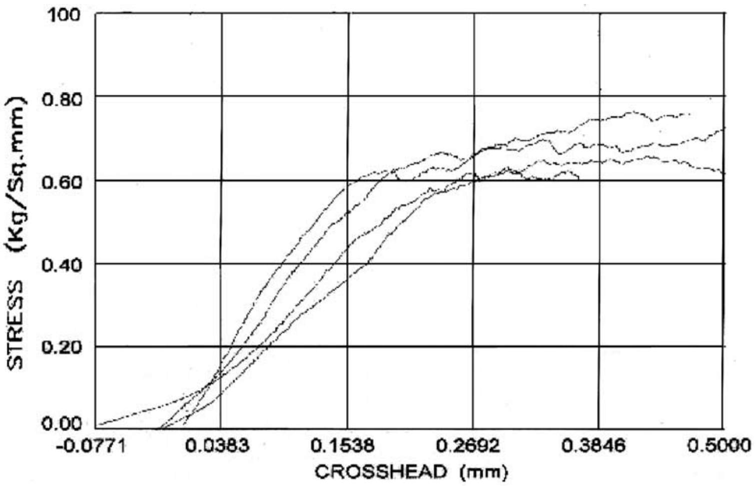

Fig. 8. Stress–strain curves for aged Pb–Bi specimens at 90 °C.

been produced less than 5 h before testing, namely the 'fresh' condition. The comparison between 'fresh' and 'aged' samples, as reported in Table 1, reveals a yield stress reduction of up to 18% for the 'fresh' LBE. It must

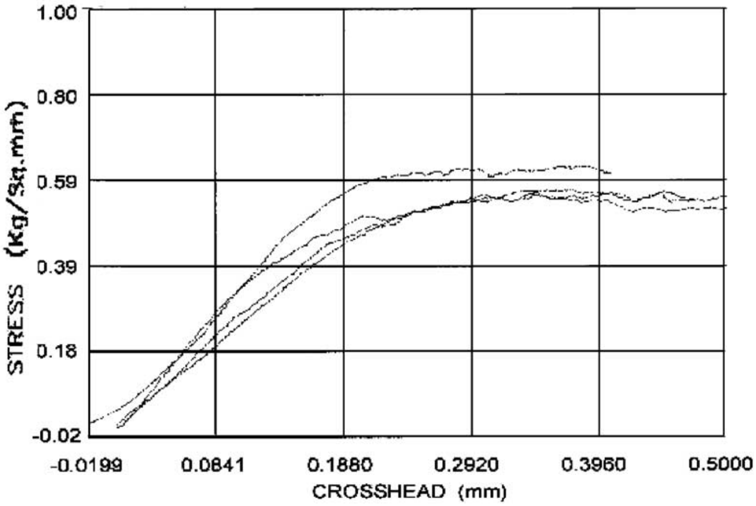

Fig. 9. Stress–strain curves for 'fresh' Pb–Bi specimens at 90 °C.

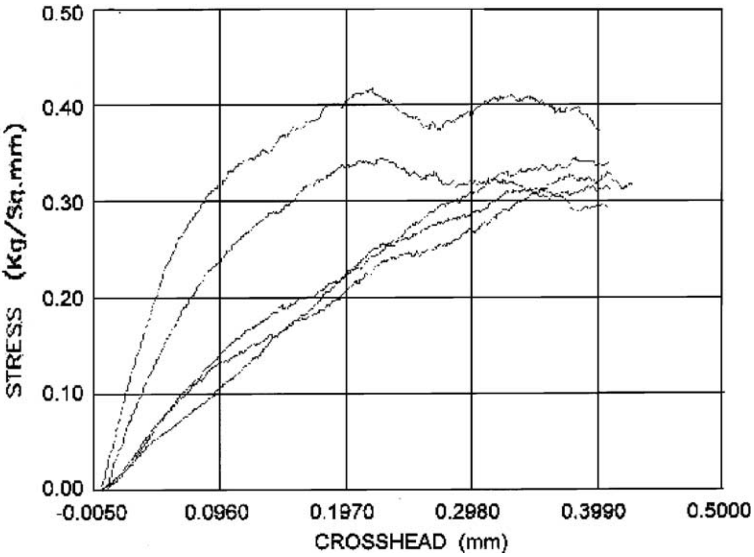

Fig. 10. Stress–strain curves for aged Pb–Bi specimens at 110 °C.

be pointed out that the case of freshly frozen samples is the most representative of the actual situation, where the expansion takes place immediately after the solidification.

The yield stress reduction observed on the 'fresh' samples is due to the lack of γ phase segregation, while in the 'aged' alloy it constitutes a significant resistance to the relative motion of dislocations. The most favourable situation, in terms of low yield stress, is the case with 'fresh' LBE expanding at high temperature and low strain rate, which can be achieved by maintaining the containing vessel at 110 °C for about one week after solidification. More than 80% of the total expected volume expansion occurs, asymptotically, during this time period, closely approaching saturation [1].

5. Conclusions

The numerical and experimental studies described in this paper show that over-stressing due to LBE recrystallization and expansion in containment vessels such as in the Megapie target must be considered during the design phase of the containment structures and

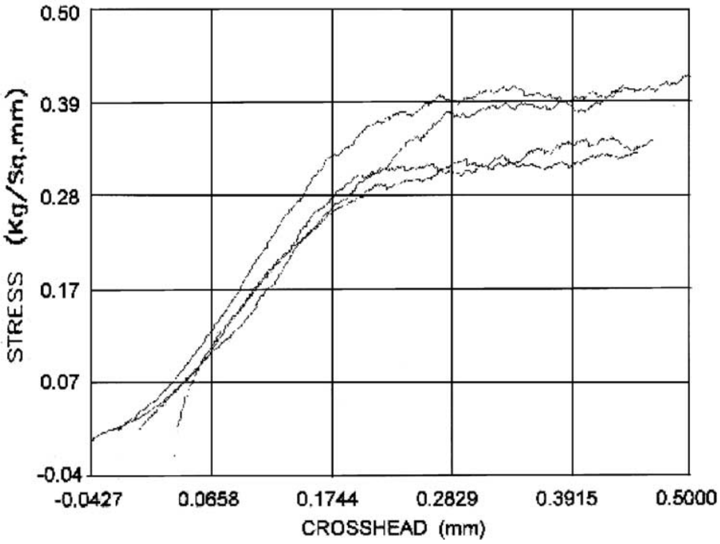

Fig. 11. Stress–strain curves for 'fresh' Pb–Bi specimens at 110 °C.

can be managed by means of engineering rules. To avoid over-stressing of structures it is necessary to reduce:

- the height of each solid LBE layer,
- the presence of internal structures,
- the LBE yield strength.

Reduction of the LBE yield strength can be attained by raising the temperature at which recrystallization takes place. The height of expanding LBE can be reduced by an appropriate freezing procedure, where the vessel is solidified in consecutive steps from bottom to top. Another important factor significantly affecting the yield stress is the strain rate. It has been shown that a realistic combination of the former effects can lead to a significant reduction of the LBE yield strength down to values of 3 MPa. As for the effect of γ-phase segregation, we could not evidence a significant difference between 'aged' (3 months) and 'fresh' (5 h) samples. Further steps have to be made to compare the numerical approach with a validating experience and to formulate specific indications for the design.

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