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The re-crystallization issue in lead–bismuth technology

P. Agostini, E. Baicchi *, A. Zucchini, G. Benamati

ENEA C.R. Brasimone, 40032 Camugnano (Bologna), Italy

Abstract

Numerical and experimental studies were performed to investigate the behaviour of lead–bismuth eutectic (LBE) after solidification. Re-crystallization of LBE is the main phenomenon to consider; it may lead to serious over-stressing of structural materials. The conditions for the target vessel of MEGAwatt PIlot Experiment (MEGAPIE) were especially considered. Some general recommendations were deduced in order to help avoiding dangerous events.

1. Introduction

During the year 2001, in ENEA (Italian Board for New Technologies, Energy and the Environment) some structural damages, due to the lead–bismuth eutectic (LBE) expansion (re-crystallization) produced by solidification and aging of the alloy inside containers and pipes were observed (see Fig. 1). Considering the Italian plans to operate large LBE facilities, the need to set a strategy for solving the re-crystallization related problems was envisaged. In order to perform a correct analysis a bibliographic research has been started, but the reported information turned out to be incomplete and only qualitative [1]. Subsequently test activities were started. Experimental studies have been, in parallel, performed and diffused by PSI (Paul Scherrer Institut, Villigen, Switzerland) [2].

On the basis of these works, experimental and numerical studies have been made in Italy to provide recommendations to design LBE vessels.

* Corresponding author. Tel.: +39 534 801276; fax: +39 534 801225.

E-mail address: elio.baicchi@brasimone.enea.it (E. Baicchi).

2. Theoretical explanation

Re-crystallization is a phenomenon that takes place at the level of crystal grain atoms, passing from a state of non-equilibrium towards a state of equilibrium. In adjacent grains different phases are segregated; these form crystal cells with different inter-atomic distances. When temperature varies with time, the atoms migrate through the grain boundaries, passing from one crystal cell to another. Solidified lead–bismuth eutectic (LBE) is a mixture of 2 phases (see Fig. 2):

- a β phase, which is an inter-metallic compound with about 40% Bi;
- a γ phase, which is a solid solution of Pb in Bi consisting of about 99% Bi with a small amount of Pb.

As a consequence of the lever rule in the phase diagram (Fig. 3), when one passes from point B to point E, the solid alloy gets richer in phase γ, and with time, at a generic temperature T lower than T_f (the melting temperature), the excess γ phase precipitates. The γ phase is richer in Bi than the phase β, and bismuth (unlike lead) expands on solidification. So the γ phase precipitation and the atoms' migration from grain to grain (i.e. re-crystallization) tends to generate a volume

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Fig. 1. Typical plastic deformations due to LBE freezing.

increase. An application of this work was referred to the target vessel of MEGAwatt PIlot Experiment (MEGAPIE) [3].

3. Yield strength of LBE

After freezing the LBE alloy inside a vessel, high stresses can arise in the containment wall. Their maximum value can be numerically related to the measured yield stress of the contained LBE. The yield strength of solid LBE is strongly dependent on temperature, age of the alloy and strain rate. Some experiments were performed to evaluate the dependence on temperature and age of the alloy (Fig. 4). The yield strength of solid LBE, at a temperature close to the melting point, ranges from 2.6 to 6 MPa at a strain rate of $5\times10^{-6}\ s^{-1}$.

In Fig. 5, the effects of LBE yield strength on the MEGAPIE vessel stresses are shown in two different cases, as calculated by a numerical simulation:

(1) at LBE yield strength of 1 MPa, the maximum stress is about 300 MPa;
(2) at LBE yield strength of 4 Mpa, the maximum stress is about 550 Mpa.

The MEGAPIE vessel is to be made of standard T91 steel, whose yield stress at room temperature is of the order of 450 Mpa. Numerical simulations for re-crystallization conditions seem to lead to stresses close to the T91 yield strength.

4. Suggestions to the designer

A stress state, induced by the LBE expansion in structures, has been studied in ENEA by means of a finite element model (FEM). The stress analysis is based on full elastic–plastic FEM simulations, which produce the stress distributions in the model as a function of LBE expansion. The model was implemented in the ABAQUS thermal-mechanical code and employed to verify the target vessel of MEGAPIE. The result indicates that some design features

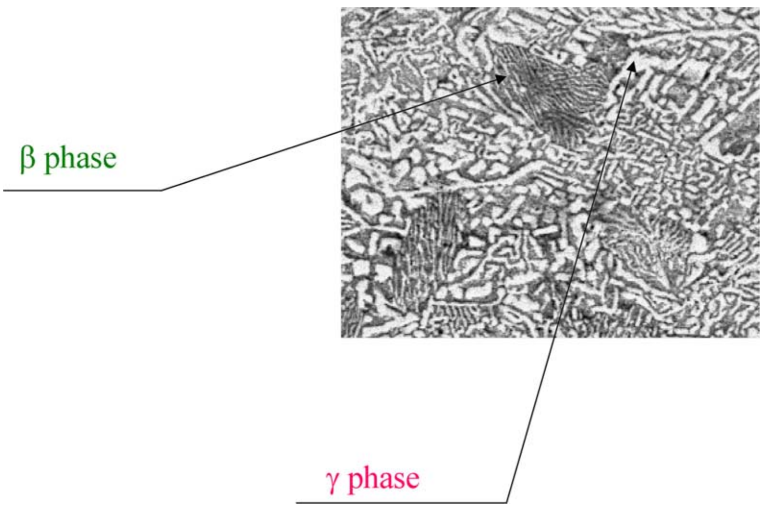

Fig. 2. Phases of the solid LBE.

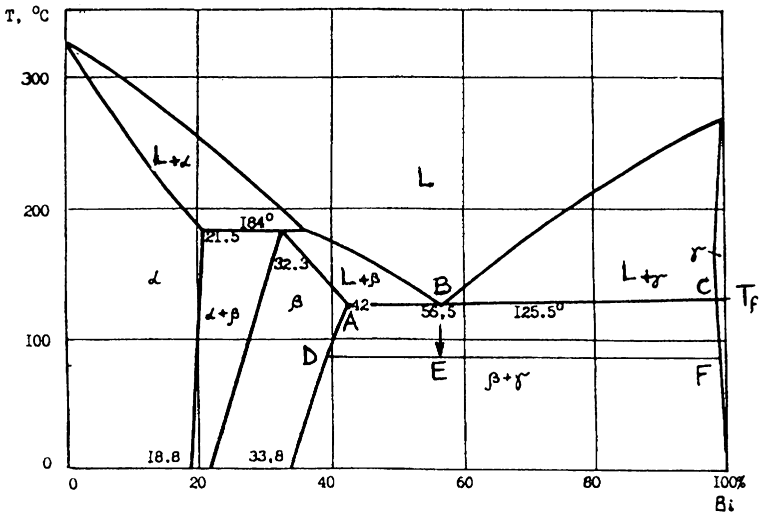

Fig. 3. Lead–bismuth phase diagram.

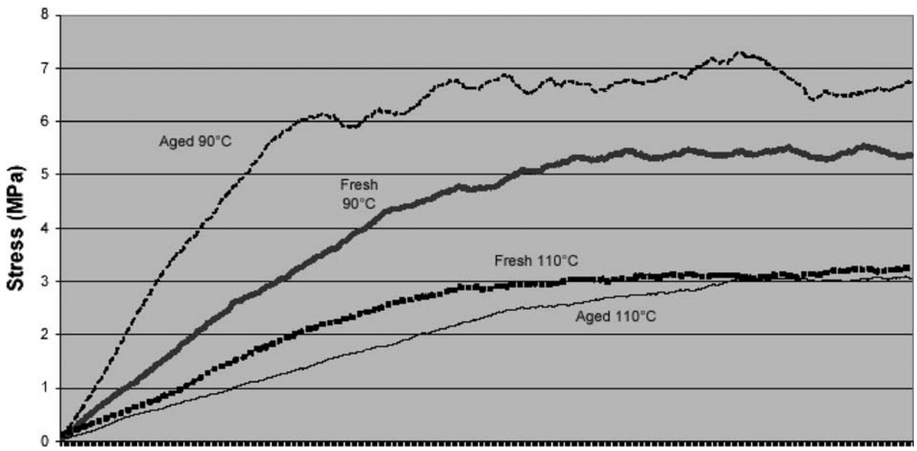

Fig. 4. Engineering stress–strain plot for fresh and aged specimens at 90 and 110 °C.

are important to limit, as far as possible, the risk to damage LBE vessels and pipes in case of alloy freezing. They are:

- reduced height of the frozen volume: so only regions of shells of uniform curvature can be interested, and moreover one can take better advantage of LBE shrinkage volumes;
- presence of adequate crush volumes: besides LBE shrinkage volumes, other voids can be artificially introduced which would 'implode' under LBE solidification;
- limited presence of internals: smaller tubes or instrumentation arrays inside the vessel should clearly complicate and worsen the stress patterns.

As for managing the LBE facilities, it is suggested, in the case of solidification, to maintain the LBE vessels and pipes in a temperature range (90–120 °C) close to the melting point. The advantages are:

(1) to minimize the bismuth super-saturation in the β phase and
(2) to take advantage of the lower yield stress of LBE at higher temperatures.

In Fig. 6 the favorable effect of a crush volume in the MEGAPIE vessel is illustrated.

It was found that LBE yield strength (σ_y) depends also on the elapsed time since solidification, at least at the lower temperature tested. Samples solidified less

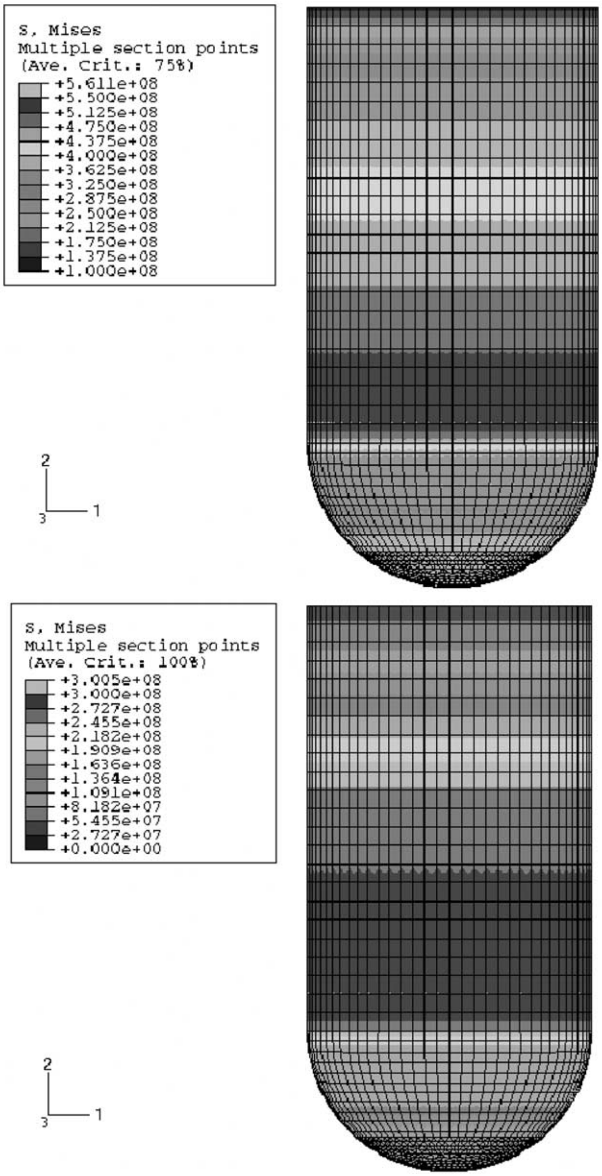

Fig. 5. Vessel V. Mises stresses at $^{\sigma_y}$LBE = 1 MPa (upper) and $^{\sigma_y}$LBE = 4 MPa (lower).

than 5 h before testing and compressed at 90 °C showed a lower σ_y than similar samples solidified 3 months earlier. At 110 °C such a difference is so small as to become indistinguishable from experimental errors, since the alloy super-saturation in the γ phase is minimal.

5. Conclusions

Some accidental events and cracks in lead–bismuth eutectic (LBE) experimental facilities for ADS reactors induced to experimental and numerical studies about the alloy solidification phenomenon. The eutectic mixture re-crystallization is the key process that must be understood in all its aspects in order to avoid similar harmful and dangerous events. From compression tests and numerical analyses we deduced some useful recommendations for the designers:

(1) it is advisable to cool LBE in batches from bottom to top, according to the geometry of the container (tall or flat). The height of the first layer must be kept as low as possible.

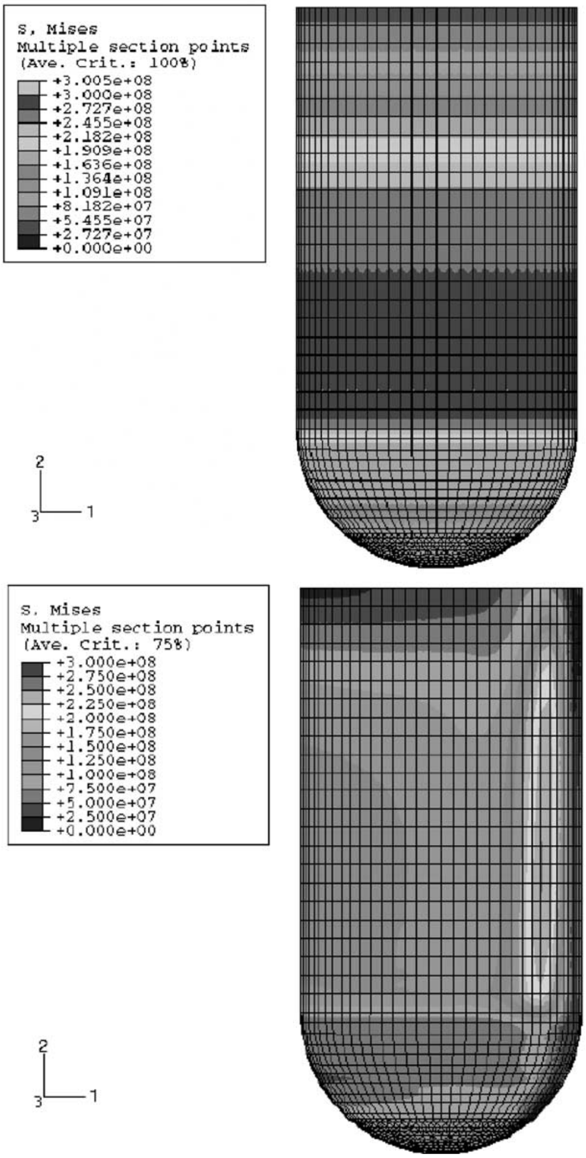

Fig. 6. MEGAPIE vessel V. Mises stresses: without crush volume (upper), with crush volume (lower).

(2) The presence of internal structures in the tanks must be reduced to a minimum.
(3) It is necessary to work in conditions of the minimal LBE yield stress (σ_y).

In particular, results showed that the higher the temperature, the lower the yield stress. At 110 °C, for instance, the yield stress is noticeably lower than at 90 °C.

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