U.P.B. Sci. Bull., Series B, Vol. 86, Iss. 1, 2024. ISSN 1454-2331

ANALYSIS OF THE INFLUENCE OF EXPOSING AUSTENITIC 316L STAINLESS STEEL SAMPLES IN LIQUID LEAD ENVIRONMENT AFTER PRELIMINARY FATIGUE TESTS

Valentin OLARU[1], Vasile RADU[2], Livia STOICA[3]

This paper analyzes the influence of exposing 316L steel samples to liquid lead on the mechanical properties. Initially, mechanical fatigue tests are performed at room temperature on samples previously exposed in liquid lead environment. Secondly, Slow Strain Rate Tensile tests were carried out on these specimens, in both liquid lead and air for comparison, at a temperature of 400°C. The microstructural analyses revealed specific patterns of the fracture surfaces and grain size morphology for the specimens.

This study will contribute to understanding the effect of liquid lead corrosiveness on the candidate structural materials for Generation IV nuclear reactors.

Keywords: liquid lead, mechanical fatigue, SSRT, 316L stainless steel

1. Introduction

Research and projects on the use of lead and Pb-Bi alloy (LBE - Lead Bismuth Eutectic) as a cooling agent in fast nuclear reactors (LFR) were initiated in the early 1950s and operational since 1963 in Russia for military nuclear submarines. Recently, in the framework of Generation IV nuclear reactors development, there has been substantial interest in the design of both critical and subcritical reactors cooled with liquid lead (Pb) or lead-bismuth eutectic (LBE) [1].

Several experimental programs are currently being conducted all over the world to repurpose nuclear waste and build fast reactors with HLM cooling. These include the Advanced Fuel Cycle Initiative of the United States [2] and IP-EUROTRANS [3] [4], a four-year (2005–2009) integrated project of the European Commission for the TRANSmutation of High Level Nuclear Waste in an Accelerator Driven System. Additionally, there are many national-level programs running in Europe, including the MYRRHA project at SCKCEN in Belgium and

[1] Researcher at Department of Nuclear Materials and Corrosion, Institute for Nuclear Research, Romania, e-mail: valentin.olaru@nuclear.ro
[2] Senior Researcher at Department of Nuclear Materials and Corrosion, Institute for Nuclear Research, Romania, e-mail: vasile.radu@nuclear.ro
[3] Researcher at Department of Nuclear Materials and Corrosion, Institute for Nuclear Research, Romania, e-mail: livia.stoica@nuclear.ro

the GEDEPEON (Gestion de Déchets Radioactives par les Options Nouvelle) initiative in France. As an ADS, MYRRHA is being developed as a versatile neutron source for R&D purposes [5] [6] [7]. There is also the HYPER (ADS) and PEACER programs from South Korea [8] [9] [10], the Japanese program in relation to the creation of ADS and LFR [11] [12] [13] [14] and the Russian BREST [15] and SVBR [16] reactor programs.

Last but not least, one of six system concepts for continued development has been selected within the context of the Generation-IV Nuclear Energy Systems initiative: a class of Pb/LBE-cooled fast reactors (LFRs). The characteristics of Pb/LBE have opened up a wide range of new applications for LFRs, including the generation of hydrogen, the processing of nuclear waste, and small modular reactors with long-life cores for supplying heat and energy to isolated regions and/or developing countries.

In Romania, at RATEN ICN, this field is of great interest in view of the decision to develop and build the ALFRED experimental demonstrator, together with a number of support laboratories at this location.

The Advanced Lead Fast Reactor European Demonstrator (ALFRED) is the result of research into Europe's next generation of lead-cooled nuclear power plants. The main goal of ALFRED and Lead technology development is to maintain nuclear energy's important contribution to the development of a safe and low-carbon European energy system. ALFRED brings together industrial and research partners to develop so-called fourth-generation fast neutron reactor technology as one of the projects supported by the European Sustainable Nuclear Industry Initiative (ESNII) as part of the EU's Strategic Energy Technology Plan (SET-Plan) [17].

Degradation of mechanical properties by liquid metals can be classified as Liquid Metal Embrittlement (LME) [18], Liquid Metal Assisted Damage (LMAD) and Environmentally Assisted Cracking (EAC) mechanisms [19]. It involves a physical-chemical and mechanical process, the interpretation of which is largely based on the concept of wetting. In order to make a true comparison between these related phenomena, it is essential to correctly characterize the state of the surface in contact with the liquid metal at the appropriate scale. Once past this crucial phase, the metallurgical and mechanical parameters for LME, LMAD or EAC failure could be better defined.

The present paper presents some results from research programs which are in progress at RATEN ICN in the field of Generation IV nuclear reactors. It investigates the influence of exposing 316L stainless steel specimens in liquid lead environment on the mechanical properties. Firstly, specimens are exposed in liquid lead at 550ºC for 1000 hours and are then subjected to mechanical fatigue. In a second step, the specimens were subjected to SSRT (Slow Strain Rate Tensile) tests in liquid lead and air under temperature conditions of interest to the ALFRED demonstrator. The microstructural analyses complement investigations at the microscopic scale.

2. Test matrix and analysis

An overview of complex activities performed for the present paper can be seen in the flowchart given in Fig. 1. Also, the chapters dealing with specific matters from the flowchart are mentioned.

The samples were machined by electro-erosion cutting from 316L stainless steel (max 18% Cr, 13% Ni, 2% Mn, 2.5% Mo, 0.030% C, 0.045% P, 0.015% S, 0.75% Si) plate imported from Columbus Stainless®, South Africa. The appearance of specimens used in the tests, and also the sketch with the gauge length calibration zone is shown in Fig. 2.

The 316L specimens have a maximum length of 40 mm and a calibrated area of approximately 15 mm.

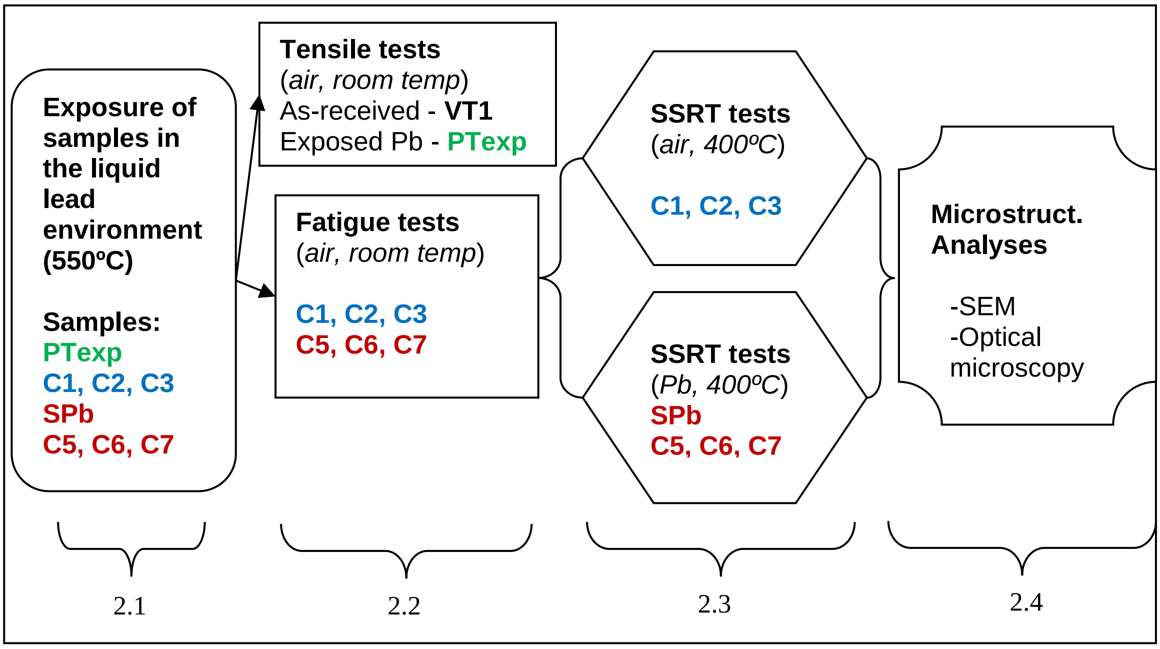

Fig. 1. The flowchart of investigation activities.

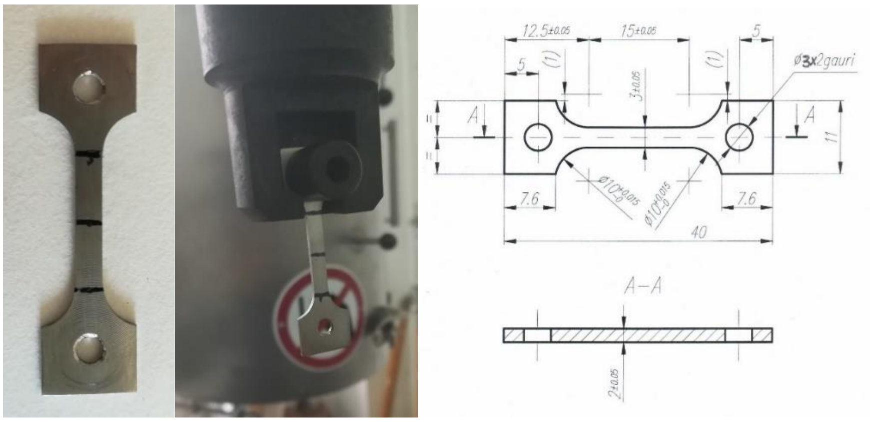

Fig. 2. Flat tensile specimens from austenitic 316L steel.

2.1 Exposure of samples in liquid lead environment

In order to pre-expose the samples in the liquid lead it was necessary to design a sample holder made of 316 and 304 steel (Fig. 3). The two steels were chosen because of their known resistance to nickel leech in molten lead environments. This was necessary as to not influence the liquid's chemistry during exposure. The central rod is made from 316L and the bolts and nuts from 304L.

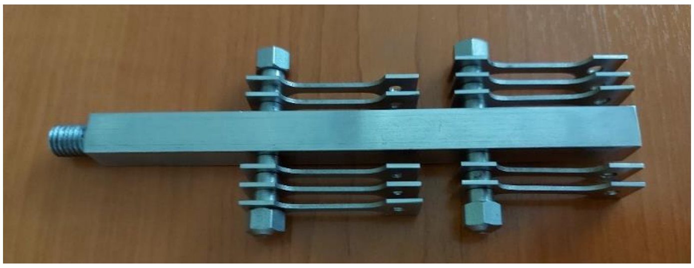

Fig. 3. The 316L sample holder for pre-exposure in the liquid lead environment.

Prior to exposure in the liquid lead environment, the specimens clamped in the holder were cleaned and chemically degreased in acetone to remove possible contaminants from the surface. The samples were then pre-exposed in a liquid lead corrosion test rig. This equipment is set up so that it can operate for an extended period of time under predetermined atmospheric and temperature conditions.

Pre-exposure was carried out in liquid lead with an oxygen concentration of about 10^{-3} mass percentage at a temperature of 550°C for 1000 hours. Samples after exposure in liquid lead are shown in Fig. 4.

Fig. 4. Samples after exposure in liquid lead environment for 1000 hours

2.2 Tensile tests and mechanical fatigue tests

In a first approach to assess the influence of liquid lead exposure on mechanical behavior, two tensile tests were carried out on a specimen in as-received condition, and also on an exposed specimen. Both tensile tests were performed at room temperature, and the stress-strain curve is displayed in Fig. 5.

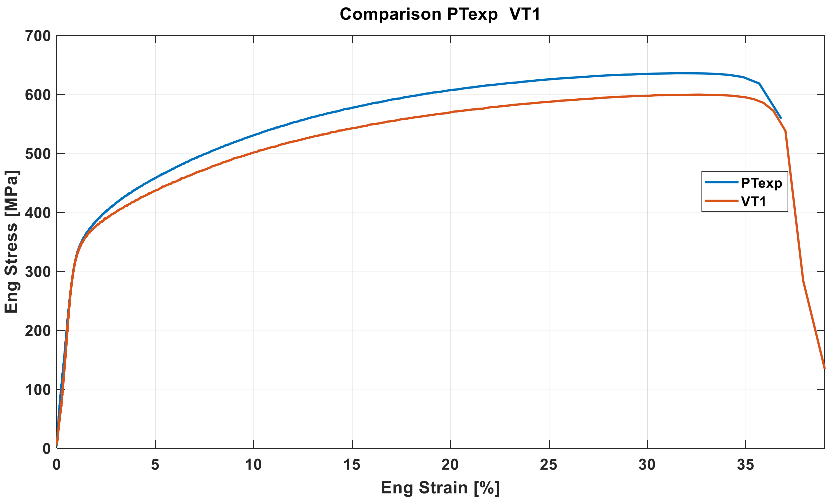

Fig. 5. Comparison of the stress-strain tensile curves in air at room temperature on 316L samples: exposed in liquid lead (PTexp) and as-received sample (VT1).

One may see a slight increase in ultimate tensile strength (UTS), and decrease of the fracture strain for the sample exposed to liquid lead (PTexp) compared to the as-received sample (VT1). The mechanical fatigue tests were performed on the specimens labeled C1, C2, C3, C5, C6 and C7 at room temperature, after their exposure to liquid lead. The load range was 50 N-1075 N and load amplitude was 512.5 N. The cycle frequency was 1 cycle/second, and the fatigue duration was about 10800s. The typical diagram of the mechanical cycling test is given in Fig. 6.

Fig. 6. The cycling test diagram in air at room temperature.

2.3 Slow Strain Rate Tensile Tests (SSRT)

The SSRT tests were carried out in air (C1, C2, C3) and in liquid lead (C5, C6, C7, SPb) at a temperature of 400°C, with a displacement rate of 0.06 mm/min, which corresponds to a strain rate of 10^{-5} s^{-1}. The specimen (SPb), was not subjected to mechanical fatigue, to provide a comparison of the influence of mechanical fatigue previously performed. The stress-strain curves, recorded during SSRT tests are displayed in Fig. 7.

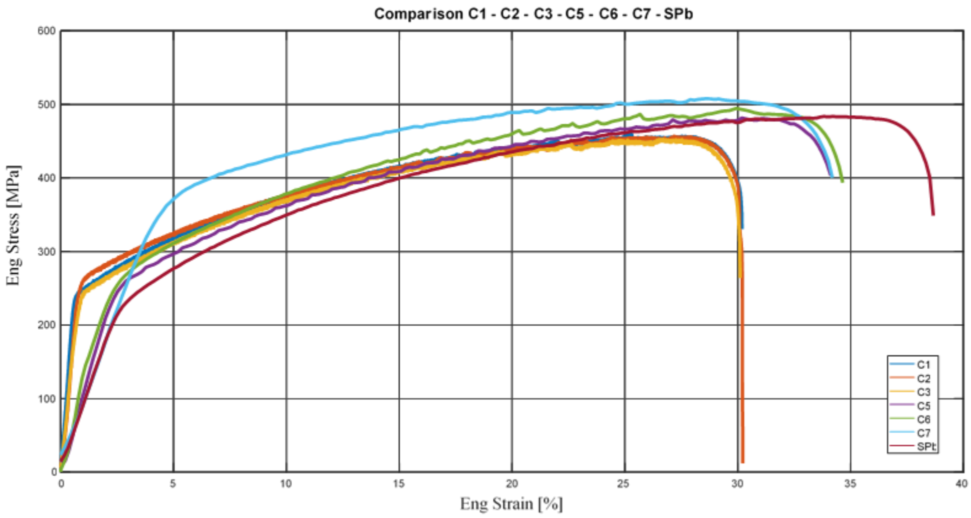

Fig. 7. The stress-strain curves from SSRT tests in liquid lead and air environment at 400°C.

At a first glance the following observations can be made:

- The mechanical fatigue strongly influences the mechanical behavior in liquid lead, for example, the SPb specimen, without fatigue, has the longest fracture strain in liquid lead from all tested specimens, also versus C5, C6 and C7 specimens, which are subjected previously to fatigue;
- The liquid lead environment influences the SSRT behavior, versus air environment, because the C1, C2 and C3 specimens have a fracture strain smaller by 5% than C5, C6, C7 specimens tested in liquid lead;
- One explanation could be given by the existence of internal stresses (residual stresses), which are diminished in the specimens during exposure in liquid lead at 550°C. This phenomenon could also be true for SSRT tests in liquid lead at 400 °C.

The SSRT test results are listed in Table 1.

Table 1

SSRT Test Results in liquid lead

Sample	No. Cycles	Cycling Range	Test environment	Fracture strain (ε_f)	UTS (σ_r)	Yield stress ($\sigma_{0,2}$)
C1	10800	50N-1075N	Air 400°C	30,66%	461 MPa	243,5 MPa
C2	10800	50N-1075N	Air 400°C	30,87%	455,8 MPa	261,9 MPa
C3	10800	50N-1075N	Air 400°C	30,23%	452,3 MPa	245,9 MPa
SPb	0	0	Lead 400°C	38,33%	483,6 MPa	218,3 MPa
C5	10800	50N-1075N	Lead 400°C	34,1%	481,4 MPa	245,4 MPa
C6	10800	50N-1075N	Lead 400°C	34,65%	494,7 MPa	238,8 MPa
C7	10800	50N-1075N	Lead 400°C	33,91%	507,9 MPa	318,4 MPa

From Table 1 we can observe that UTS and yield stress values obtained in liquid lead environment are, more or less, in the same domain.

According to section "11.1.2 Evaluation of EAC Resistance Based on SSRT" of the ASTM, reference [20], the "Plastic Elongation Ratio- RE" is used here for the two environments: air and liquid lead. The value of RE is defined as the ratio of plastic elongation determined for a material in a test environment ε_e (lead) to the corresponding value determined in the control environment ε_c (air).

$$RE = \frac{\varepsilon_e}{\varepsilon_c} \quad (1)$$

Using the values from Table 1, we obtained $RE = 1.119$. An increase of plastic deformation as $\varepsilon_e = 1.119 \cdot \varepsilon_c$ in liquid lead versus air indicates that the influence of the liquid lead environment on pre-fatigued tested samples cannot be neglected compared to those tested in air.

This result is quite interesting, because the effect of liquid lead corrosiveness is not very aggressive upon the micro-cracks, resulting from the pre-fatigue phenomenon. This phenomenon seems to be very complex, as many authors claims in their studies [1] and it needs more dedicated experimental tests and complex interdisciplinary analyses.

2.4 Microstructural Analyses

Microstructural analyses of the sample surfaces before and after mechanical fatigue and also of the fracture surfaces after SSRT tests were performed to determine the pattern of fracture and the possible influences of the mechanical fatigue practiced. Microscopic analyses were carried out with the TESCAN VEGA LMU 2 scanning electron microscope, using both the secondary electron detector (SE - surface topography) and the backscattered electron detector (BSE - lead residue identification). Fig. 8 shows the surface of pre-exposed sample C1 (arbitrarily selected – general representation of features for all samples tested) before and after mechanical cycling, and the surface of another sample (VT1) in the "as-received" condition. After cycling, small cracks can be seen in the oxide layer deposit on the surface of the pre-exposed sample. Without removing this very thin oxide layer, we cannot be sure that these cracks have propagated into the steel substrate. Most likely they only manifested on the brittle oxide layer.

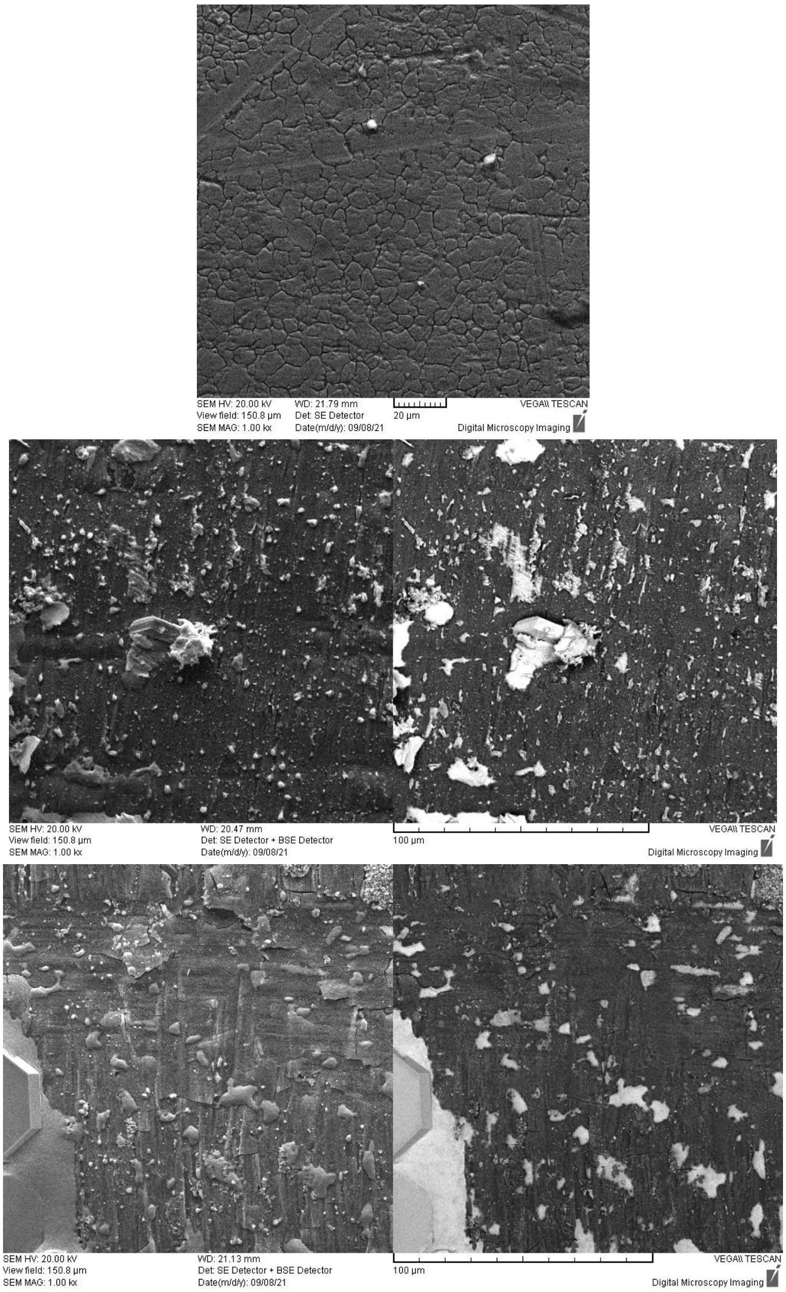

Fig. 8. Surface pictures of sample VT1 in "as-received" condition (TOP) and sample C1 pre-exposed in liquid lead before (MIDDLE) and after (BOTTOM) mechanical cycling.

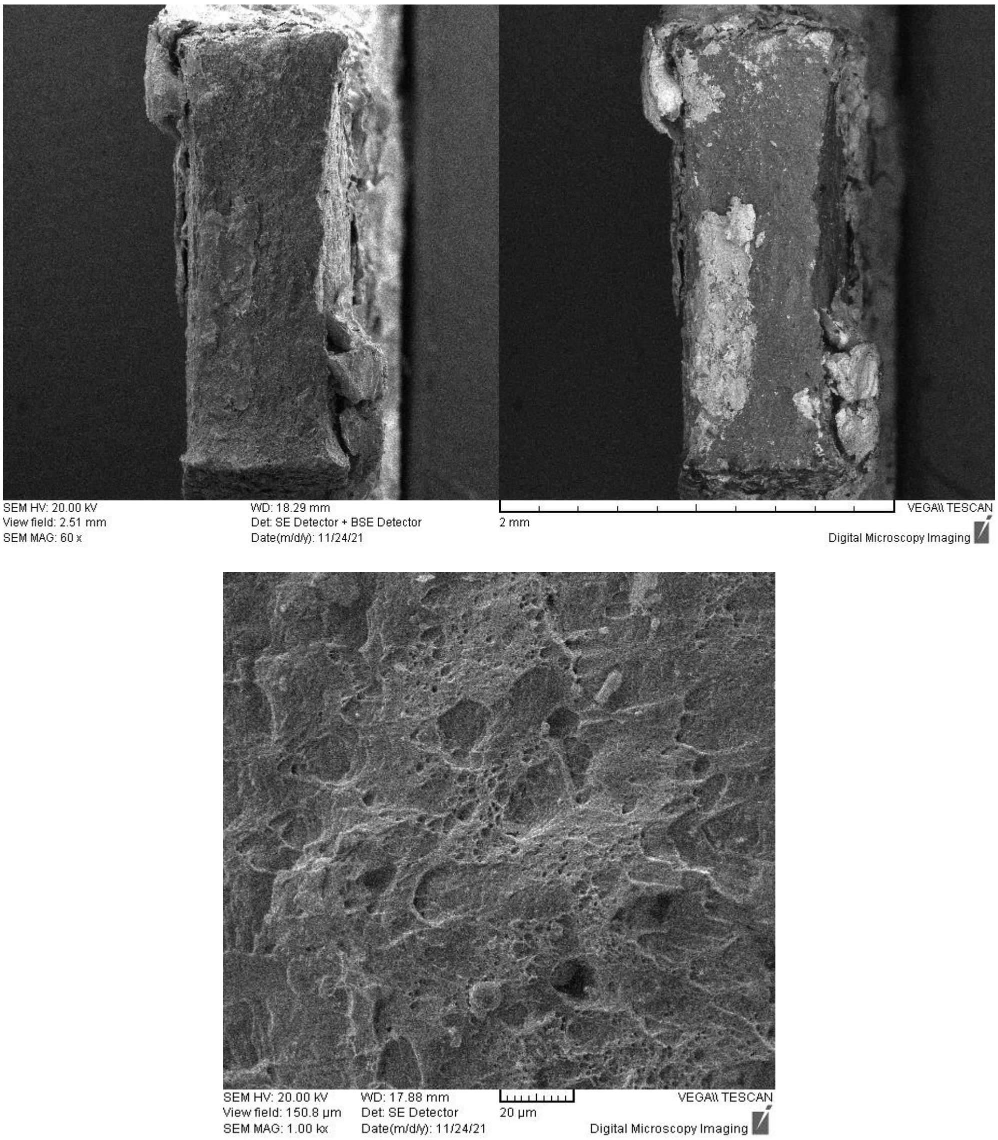

Fig. 9. Pictures of the fracture surface of sample C5, tested in lead at 400°C.

Optical microscopic analyses were also performed to examine the grain size morphology before (Fig. 10) and after (Fig. 11) pre-exposure treatment in liquid lead medium at 550°C for 1000h. To analyze the samples, they were embedded in epoxy resin after which they were sanded and chemically treated (by dissolving 10g oxalic acid in 100ml water) to reveal the grain boundaries. Pictures were taken with an optical microscope under polarized light.

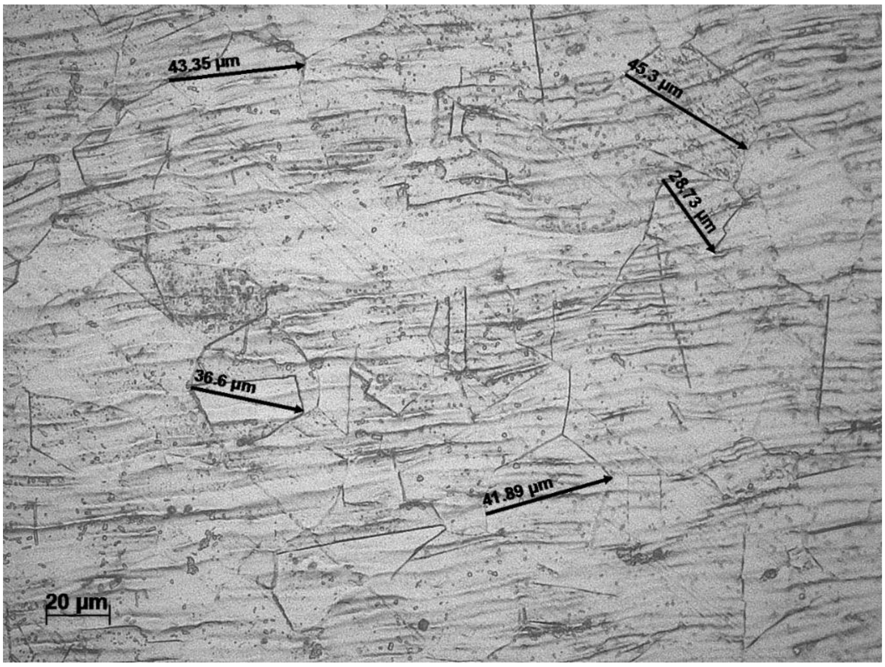

Fig. 10. Grain sizes for the 316L steel sample VT1 not subjected to liquid lead exposure treatment.

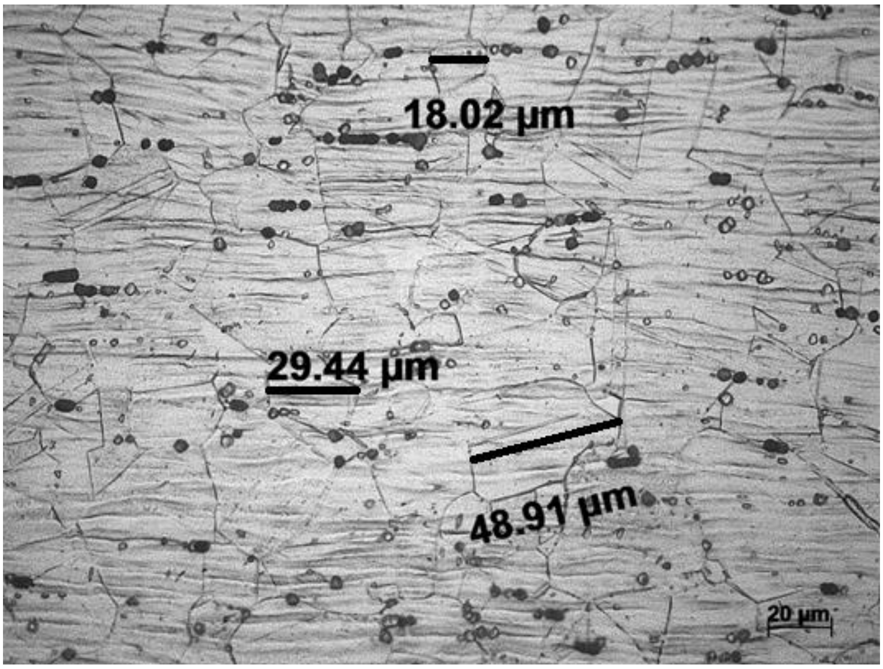

Fig. 11. Grain size of 316L steel sample PTexp after liquid lead exposure treatment at 550°C for 1000h.

The differences between the grain sizes before and after treatment are not significant to explain the slight discrepancy between the tensile curve plots (Fig. 5) of the tests carried out in air at room temperature on the two 316L samples: one exposed in liquid lead (PTexp) and one as-received sample (VT1). One explanation could be given by the existence of internal stresses (residual stresses), which are diminished in the specimens during exposure in liquid lead at 550°C.

3. Conclusions

In this study, mechanical fatigue tests were carried out on samples made of austenitic 316L steel, pre-exposed in liquid lead at 550°C for 1000 hours. Then the specimens were subjected to SSRT (Slow Strain Rate Tensile) tests in liquid lead under temperature conditions of interest to the ALFRED demonstrator.

The conclusions of the present work are:

- Mechanical fatigue has an impact on the mechanical behavior in liquid lead. For instance, among all the studied specimens that were previously subjected to fatigue, the specimen without fatigue has the longest fracture strain in liquid lead.
- The liquid lead environment influences the SSRT behavior, versus air environment, therefore specimens tested in air have a fracture strain smaller by 5% than specimens tested in liquid lead; The "Plastic Elongation Ratio" obtained is RE=1.119, that also indicates that the influence of the liquid lead environment on pre-fatigued tested samples cannot be neglected compared to those tested in air;
- An intriguing conclusion is that the pre-fatigue phenomena causes the liquid lead's corrosiveness to affect the microcracks in a less aggressive manner. Numerous authors state in their scientific investigations that this phenomena appears to be highly complex, necessitating more thorough experimental testing and intricate multidisciplinary analysis.
- From microstructural analyses (SEM) it can be concluded that the fracture, regardless of the test environment, was mixed pattern ductile-brittle, having "cone-cup" aspects with some cleavage paths of the grains observed.
- The differences between the grain morphologies before and after treatment are not significant to explain the slight discrepancy between the tensile curve plots of the tests carried out in air at room temperature on 316L samples: one exposed in liquid lead (PTexp) and one as-received sample (VT1). Perhaps, one explanation could be given by the existence of internal stresses (residual stresses), which are diminished in the specimens during exposure to liquid lead at 550°C.

- Tensile curve graphs of tests conducted in air at room temperature on two 316L samples (one as-received sample (VT1) and one exposed to liquid lead (PTexp) show a small variance that cannot be explained by differences in grain morphologies before and after treatment. Maybe internal stresses, also known as residual stresses, which are reduced in the specimens when they are exposed to liquid lead at 550°C, could provide some insight. This phenomenon could be true also for SSRT test in liquid lead at 400 °C.

Further activities in the research programs which are in progress at RATEN ICN, in the field of Generation IV nuclear reactors, will be focused on investigating the influence of the exposure in liquid lead environment on the mechanical properties of 316L stainless steel specimens by means of the non-conventional "small-punch" test. These tests will contribute to the study of the effect of liquid lead corrosiveness on the candidate structural materials for Generation IV nuclear reactors.

R E F E R E N C E S

[1] OECD-NEA, Handbook on lead-bismuth eutectic alloy and lead properties, materials compatibility, thermal-hydraulics and technologies, Issy-les-Moulineaux, France: OECD-NEA 6195, 2007.

[2] Report to Congress on Advanced Fuel Cycle Initiative: The Future Path for Advanced Spent Fuel Treatment and Transmutation Research (2003), US DOE, Office of Nuclear Energy, Science and Technology.

[3] Integrated Project EUROpean Research Programme for the TRANSmutation of High Level Nuclear Waste in an Accelerator Driven System, EUROTRANS (2004), EC project No. FI6W-CT-2004-516520.

[4] Knebel, J.U., et al. (2005), "Overview on Integrated Project EUROTRANS: EURopean Research Programme for the TRANSmutation of High Level Nuclear Waste in an Accelerator Driven System", 3rd Annual Idaho ADSS Experiments Workshop, Pocatello, ID, 1-2 June 2005.

[5] Abderrahim, H. Aït, et al. (2001), "MYRRHA: A Multipurpose Accelerator Driven System for Research & Development", Nuclear Instruments & Methods in Physics Research, A 463, 487-494.

[6] Abderrahim, H. Aït (2005a), "MYRRHA: A Multipurpose ADS for R&D, Progress Report at End 2004", ICONE 13, Beijing.

[7] Abderrahim, H. Aït, D. de Bruin (2005b), MYRRHA, a Future for Nuclear Research. Pre-design (DRAFT-2) Report, H. Aït Abderrahim, D. de Bruin (Eds.), SCKCEN report, Mol, Belgium.

[8] Hwang, I.S., S.H. Jeong, B.G. Park, W.S. Yang, K.Y. Suh, C.H. Kim (2000), "The Concept of Proliferation-resistant, Environment-friendly, Accident-tolerant, Continual and Economical Reactor (PEACER)", Progress in Nuclear Energy, Vol. 37, No. 1-4, pp. 217-222.

[9] Song, T.Y., et al. (2004), "HYPER Project", Proceedings of OECD/NEA 4th International Workshop on Utilisation and Reliability of High Power Proton Accelerators, Daejeon, Korea, 16-19 May 2004.

[10] Park, W.S., et al. (1996), Development of Nuclear Transmutation Technology, KAERI/RR-1702/96; Proceedings of International Conference on Heavy Liquid Metal Coolants in Nuclear Engineering, Vol. 1, 2, p. 824, 5-9 Oct. 1998, Obninsk, IPPE.
[11] Sasa, T., et al. (2004), "Research and Development on Accelerator-driven Transmutation System at JAERI", Nuclear Engineering and Design, 230, pp. 209-222.
[12] Takahashi, M., et al. (2004), "Design and Experimental Study for Development of Pb-Bi Cooled Direct Contact Boiling Water Small Fast Reactor (PBWFR)", Proc. of ICAPP'04, Pittsburgh, PA, USA, 13-17 June 2004, paper 4058.
[13] Oigawa, H., et al. (2004), "R&D Activities on Accelerator-driven Transmutation System in JAERI", OECD/NEA 8th Information Exchange Meeting on Actinide and Fission Product Partitioning & Transmutation, Las Vegas, Nevada, USA, 9-11 November 2004.
[14] Mukaiyama, T. (1999), OMEGA Programme in Japan and ADS Development at JAERI, IAEA TECDOC--1365, pp. 153-165.
[15] Filin, A.I., et al. (2000), Design Features of BREST Reactors and Experimental Work to Advance the Concept of BREST Reactors, IAEA-TECDOC 1348, pp. 36-47, 23-27 October 2000.
[16] Stepanov, V.S., et al. (1998), SVBR-75: A Reactor Module for Renewal of WWER-440 Decommissioning Reactors – Safety and Economic Aspects, IAEA TECDOC 1056, pp. 165-176, November 1998.
[17] A. Alemberti, et al. (2015), ALFRED and the Lead Technology Research Infrastructure, European Research Reactor Conference (RRFM) 2015 Conference Proceedings.
[18] M.H. Kamdar, "Embrittlement by Liquid Metals", Prog. Mater. Sci., 1973, 15, pp. 289-372.
[19] S. Maloy, Handbook on Lead-bismuth Eutectic Alloy and Lead Properties, Materials Compatibility, Thermal-hydraulics and Technologies, LANL, United States, 2015.
[20] ASTM G 129-00 "Standard Practice for Slow Strain Rate Testing to Evaluate the Susceptibility of Metallic Materials to Environmentally Assisted Cracking", 2000.