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Ingrid Proriol Serre, Ines Ponsot, Jean-Bernard Vogt. Alumina-Forming Austenitic (AFA) steels and aluminium-based coating on 15-15 Ti steel to limit mechanical damage in presence of liquid lead-bismuth eutectic and liquid lead. 6th International Virtual Conference of Engineering Against Failure, Jun 2021, En visio - Grèce, Greece. 10.1051/mateconf/202134902007 . hal-03436219

HAL Id: hal-03436219

<https://hal.univ-lille.fr/hal-03436219v1>

Submitted on 19 Nov 2021

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Alumina-Forming Austenitic (AFA) steels and aluminium-based coating on 15-15 Ti steel to limit mechanical damage in presence of liquid lead-bismuth eutectic and liquid lead

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Abstract. To limit corrosion of the steels in contact with liquid metal (Pb or Pb-Bi), both different solutions based on the presence of alumina at the surface of the steels were selected: 2 Alumina-Forming Austenitic (AFA) steels and the 15-15 Ti steel coated by alumina layer. These technical options to mitigate corrosion by lead or Pb-Bi were investigated in terms of mechanical performances in liquid lead or liquid Pb-Bi. So the liquid metal embrittlement (LME) sensitivity of the different materials selected for their good corrosion resistance was evaluated by tensile tests or small punch tests carried out in presence of liquid metal, and by post-mortem analysis of the cracking and of the fracture surfaces. No LME sensitivity for the tested conditions has been observed for the Al₂O₃ coated 15-15Ti steel and for the two AFA steels (16Ni-14Cr-2.5Al-2.5Mn-1Nb), one without and one with 2% wt.% W and 0.02% wt.% Y in their as received state. But a 650 °C thermal aging promotes modifications of the microstructure specially precipitations and then LME sensitivity of the AFA steels, according the nature of the precipitation.

1 Introduction

The corrosion resistance and the mechanical behaviour assessment of structural alloys (steels) is crucial for the durability and the safety of the Lead cooled Fast Reactor (LFR) and Accelerated Driven Systems (ADS) for which the structural materials are in contact with liquid lead or liquid lead-bismuth eutectic (LBE). Indeed, the presence of liquid lead or LBE promotes for the steels different types of corrosion according to the oxygen content in the liquid metal: 1. at high oxygen content, oxidation of the steel with the PbO formation – 2. at low oxygen content, dissolution of the steel or of a selected element which can promote microstructure evolution [1-5]. The oxygen level in the liquid metal being low to avoid the formation of oxides and plugging, to solve corrosion damage by liquid metal, the presence of a protective layer at the surface of the steels has been proposed because it aims at avoiding

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the contact between the steel and the liquid metal, limiting all steel / liquid metal interactions and therefore reducing the corrosion phenomena. Among the solutions explored over the past 10 years [6], one of the most promising seems to be the presence of a layer of alumina on the surface of steels, because of its stability whatever the oxygen content in the liquid metal [1]. Two types of solution have been proposed: 1. the development of Al_2O_3 coating to protect the steel [7-9], 2. the development of adapted steels containing aluminum to promote the in-situ formation of a stable and protective Al_2O_3 oxide layer [3,10-16]. Note that concerning the coatings, one of the problems could be the damage of the steel in the case of surface deterioration of the coating (cracking, wear, scaling, delamination, dissolution).

Though the essential challenge of these solutions is the corrosion resistance in liquid Pb or/and LBE, the mechanical resistance in liquid metal must be ensured to guarantee the life time of the structure under mechanical loading (due to pure mechanical stresses or due to stresses induced by temperature fluctuations). Indeed, though tough and ductile metallic alloys are selected, they may become brittle when stressed in liquid metal exhibiting thus liquid metal embrittlement (LME), one of the liquid metal assisted mechanical damages. Furthermore, for coated steel, the mechanical resistance of the substrate could be affected by the coating, not only in air but also in presence of a liquid metal.

The presented work concerns the mechanical behaviour in liquid LBE or liquid lead of steels which have been modified for corrosion mitigation in liquid metal. In particular, it aims at evaluating if the corrosion mitigation techniques impact the susceptibility of three steels to LME: an Al_2O_3 coated 15-15Ti steel and two Alumina-Forming Austenitic (AFA) steels. So, the influence of the presence of the liquid metal was investigated by performing monotonic tests (Small Punch Tests and tensile tests) in air and in liquid LBE or/and liquid lead and then by cracking and fracture surfaces analyses by scanning electron microscopy (SEM). Different parameters known to impact the LME sensitivity have been considered: the temperature, the strain rate, a thermal aging.

2 Al_2O_3 coated 15-15Ti steel

15-15Ti austenitic stainless steel in the form of plates obtained by rolling has been covered by a layer of aluminium oxide using the Pulsed Laser Deposition (PLD) technique in the facilities of the Istituto Italiano di Tecnologia (IIT), Milano, Italy [8, 17-18]. The Al_2O_3 coating had an average thickness of 1.5 μm and a roughness parameter R_z inferior to 1 μm .

To evaluate LME sensitivity and the influence of the presence of the coating, tensile tests were performed in air and in lead for the 15-15Ti steel, and in lead for the coated 15-15Ti steel. The used samples are plate specimens with a gauge length of 9.8 mm, for a thickness of 2 mm and a width in the gauge length of 1.5 mm. Since a low oxygen content in liquid lead promotes LME sensitivity of the steels by limiting the in-situ formation of oxide layer at the steels surface [19-20], tests were carried out in liquid lead with a low oxygen content (inferior to 10^{-8} wt.%) using the setup developed by Ye et al. [19] for tests in liquid LBE and adapted for tests in liquid lead. The tests were performed at 400 °C and 500 °C. Furthermore, hardness and strain rate influence LME sensitivity, the tensile specimens were extracted in the rolling direction (noted L) and perpendicularly to it (noted T). Two strain rates were considered: $5 \cdot 10^{-5} \text{ s}^{-1}$ and $5 \cdot 10^{-6} \text{ s}^{-1}$. For each condition, at least two tests were performed.

For all tested conditions, the behaviour of the 15-15Ti steel is ductile (Fig. 1). Contrary to what is expected for an austenitic steel, the studied 15-15Ti steel does not present a strain hardening, what can be explained by the absence of heat treatment after the final cold rolling step of the steel plate. The tensile strength and the yield strength are higher than those reported in the literature for the solution-annealed 15-15Ti steel which presents an appreciable strain hardening [21].

Because the high strength materials are generally more sensitive to LME [22-24], the studied 15-15Ti steel is probably be more sensitive to LME compared to the solution-annealed state. No influence of the presence of lead on the tensile curves obtained at 400 °C and at $5 \cdot 10^{-5} \text{ s}^{-1}$ has been noted for the tested 15-15Ti steel (Fig. 1).

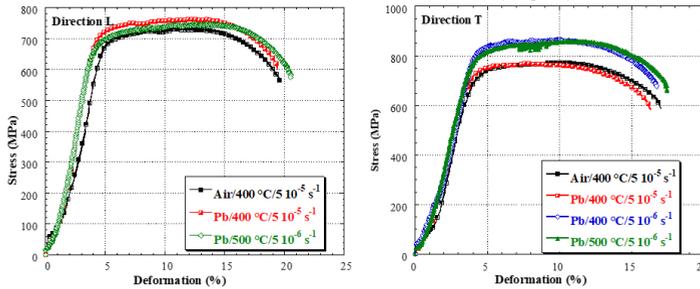


Fig. 1. Engineering stress-strain curves of 15-15Ti steel without coating.

For the direction L, no significant evolution is shown between the curves obtained at the different strain rates and temperatures. But, for the direction perpendicular to the rolling direction (T), in contrast with the absence of temperature effect, the strain rate influences the tensile curves: the stresses are more important with the decrease of the strain rate which is unusual but can be explained by the presence of the TiC precipitates. Finally, as expected, for the direction T, the values of the uniform elongation and the elongation at rupture are lower and, the tensile strength, the yield strength are more important by comparison with the direction L (the rolling direction).

In liquid lead, the behaviour of the coated steel (Fig. 2) is ductile and similar to the behaviour of the steel without coating. But some differences in the stress values are noted for some conditions: direction L at 400 °C and 10^{-5} s^{-1} , direction T at 500 °C and 10^{-6} s^{-1} .

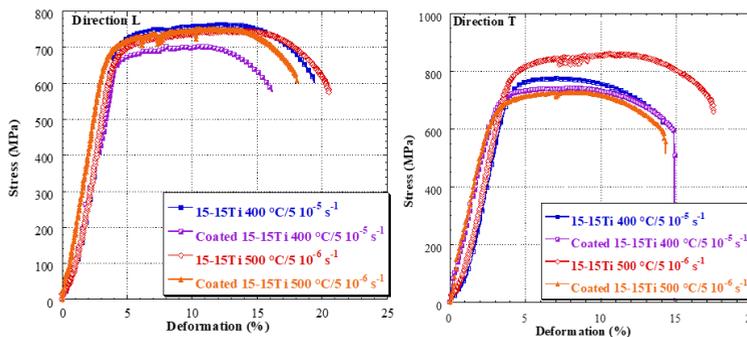


Fig. 2. Engineering stress-strain curves of coated and non-coated 15-15Ti steel loaded in liquid lead.

After test, the fracture surfaces were analysed by SEM. Prior to the SEM examinations, the samples tested in liquid metal were cleaned in a solution containing CH_3COOH , H_2O_2 and $\text{C}_2\text{H}_5\text{OH}$ at a ratio of 1:1:1 to remove the solidified lead. In all cases, with or without coating, a ductile fracture with the presence of dimples has been observed (Fig. 3). We observed neither embrittlement of the 15-15Ti steel near the surface of the sample with Al_2O_3 coating nor cracking of the coating.

The 15-15Ti steel presents a ductile behaviour in air as in liquid lead (with a low oxygen content) in the tested conditions. This result is consistent with that of Hojna et al. [25]. The temperature and the strain rate do not influence significantly the characteristics of the tests curves except for the direction T.

The 15-15Ti steel coated with Al_2O_3 to limit corrosion in lead presents a ductile behaviour with a ductile fracture (Fig. 3). No LME has been observed in the conditions of test. But, in

some cases, the mechanical strength is somewhat lower for the coated steel as compared with the uncoated one but the ductility is the same. The coating process seems therefore to soften the substrate while keeping the same ductility. Modifications of the microstructure of the steel are probably promoted by an increase in temperature of the steel during the Al_2O_3 coating process and by the fact that after cold forming, the steel has not been heat treated and so may not be in a state of stable microstructure.

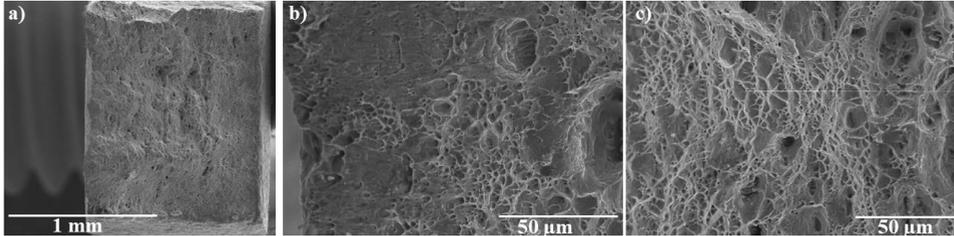


Fig. 3. Fracture surfaces of coated 15-15Ti steel (Direction T - $500\text{ }^\circ\text{C} - 5 \cdot 10^{-6}\text{ s}^{-1}$): a): overview, b): at the edge, c): in the centre.

Thus, if no LME sensitivity is observed, the steel coated with Al_2O_3 does not appear as an optimal option in regards with the mechanical properties which seem in some conditions to be modified not by the presence of the liquid metal or of the Al_2O_3 coating but by the microstructural evolutions generated during the coating elaboration. Optimization of the elaboration process and heat treatments should allow to improve this promising solution.

3 AFA steels

The two studied low-alloyed AFA steels, noted AFA3 and AFA8 were produced by Kanthal, part of Sandvik Group. Solidified heats obtained in a vacuum induction melting furnace were hot-worked and then annealed. The chemical composition of the two steels obtained in the form of bar is given in Table 1. AFA8 heat was produced with additions of W and Y to improve creep strength by solid solution strengthening and by precipitation hardening of Laves phase.

Table 1. Chemical composition of the two studied AFA alloys (wt%).

	Al	Cr	Ni	Si	Mn	C	Nb	Other	Fe
AFA3	2.5	14.1	15.9	0.14	2.5	0.008	0.93	-	Balance
AFA8	2.5	14.0	15.9	0.15	2.5	0.05	0.95	2.0 W, 0.02 Y	Balance

Small punch tests (SPT) in air and in oxygen saturated liquid LBE (44 wt% Pb and 56 wt% Bi) were performed. Indeed, this test has appeared very sensitive to evidence LME [26] and is appropriated to study different conditions (environment, temperature, strain rate, microstructure state). The SPT is based on punching a flat small specimen (disc diameter around 7 mm in our case and thickness equal to 0.5 mm) by a tungsten carbide ball until fracture [26]. For tests in liquid metal, the upper surface of the specimen was in contact with the liquid LBE and was submitted to tensile loading. SPT were carried out in air and in LBE at $350\text{ }^\circ\text{C}$ and $450\text{ }^\circ\text{C}$. The punch was carried out with a controlled cross-head displacement velocity of 0.5 mm/min which corresponds to an average strain rate around $5 \times 10^{-3}\text{ s}^{-1}$. For each condition, at least two tests were performed. The recorded load-displacement curves were corrected by a calibration to take into account the deformation of the set-up. After SPT, the fracture surfaces and cracking were analysed by SEM. Prior to the SEM examinations, the samples tested in liquid LBE were cleaned in a solution containing CH_3COOH , H_2O_2 and

C_2H_5OH at a ratio of 1:1:1. For each condition, two tests were carried out. In the case of large scatter, a third test was performed.

Concerning the SPT carried out for the "as received" steels (noted AFA3-AR and AFA8-AR), the load displacement curves show a typically ductile behaviour in air as in oxygen saturated LBE at 350 °C and at 450 °C (Fig. 4). Furthermore, the main crack is circular without radial cracks often suggesting embrittlement. In addition, fracture surfaces present dimples. Rarely, ductile secondary micro-cracks have been observed at the fracture surfaces for tests in LBE.

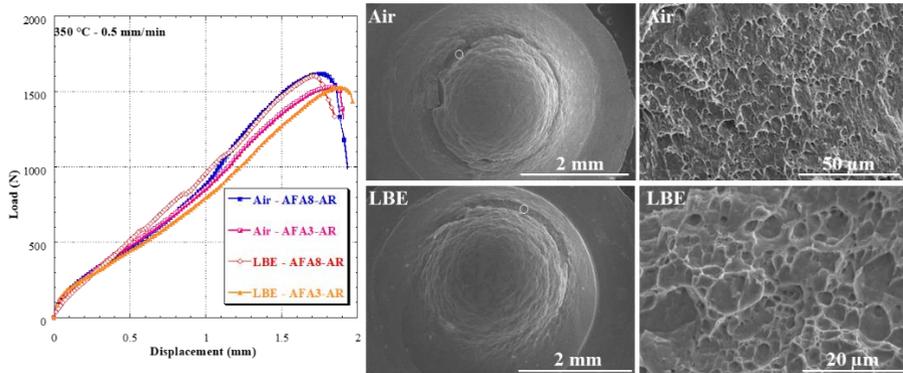


Fig. 4. SPT curves obtained for the two steels (as received state) at 350 °C and 0.5 mm/min and cracking and ductile fracture for samples tested in air and in LBE (AFA3-AR steel).

No significant differences have been observed and noted between tests in air and in LBE (data obtained from the curves, cracking and fracture surface). So, in oxygen saturated LBE at 350 °C and at 450 °C, AFA3 and AFA8 steels in the as-received condition remain ductile without any evidence of LME. As it is known that slower strain rate can increase susceptibility to LME by LBE [19,20], SPT were carried out at 350 °C at a displacement speed of 0.005 mm/min. No effect of the liquid metal has been observed.

Then SPT were performed on steels aged at 650 °C to study whether eventual modifications in the microstructure due to the thermal aging lead or not to a sensitivity to LME by LBE for the two steels. So SPT at 350 °C and 450 °C in air and in LBE at 0.5 mm/min were carried out after an aging of 1008, 2088, 3028 and 5044 hours (steel state respectively noted AFA3-1008/AFA8-1008, AFA3-2088/AFA8-2088, AFA3-3028/AFA8-3028, AFA3-5044/AFA8-5044).

For the AFA3 steel, all the load displacement curves obtained after the different aging durations show a ductile behaviour, confirmed by the observation of ductile fracture and main circular crack (Fig. 5). The absence of significant evolution being observed between results of tests in air and in liquid LBE suggests that the aging does not lead to LME sensitivity of the AFA3 steel. But for some samples tested in LBE, radial cracks accompany the main circular crack and ductile fracture surface with some brittle zones were observed.

The load displacement curves obtained for the AFA8 steel after the different aging durations and for SPT at 350 °C and 450 °C in air and in LBE show a ductile behaviour (Fig. 6) with in some conditions (AFA8-1008 at 450 °C, AFA8-3028 at 350 °C and 450 °C, AFA8-5044 at 450 °C) a significant decrease of the ductility and of the mechanical resistance in presence of LBE. However, for the other cases, due to the scattering of the results, the evolution is not so meaningful. After SPT carried out in air, circular cracking and ductile fracture surfaces have always been observed. But, the circular crack is accompanied sometimes by radial cracks, and some intergranular micro-cracks were observed at the level of the fracture surface (Fig. 6). Concerning the samples tested in LBE, either a main circular crack and a ductile fracture surface or a circular crack with developed radial cracks with a

very large brittle fracture surfaces were observed (Fig. 6). Furthermore, at least half of the AFA8 samples tested in LBE show a brittle fracture surface.

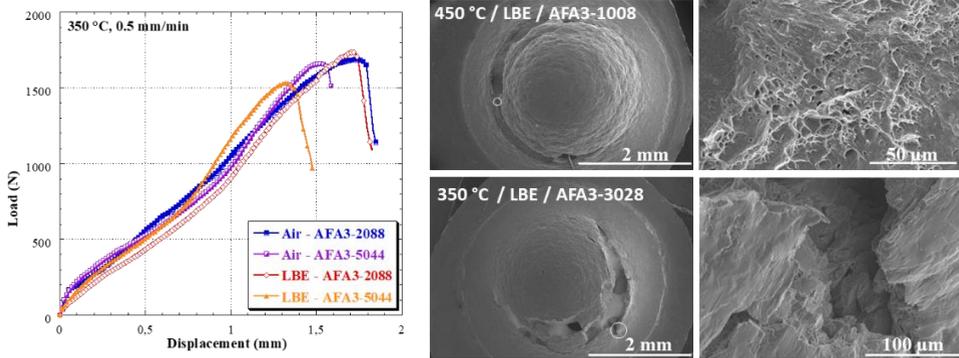


Fig. 5. SPT curves obtained for the AFA3 steel at 350 °C and 0.5 mm/min after two aging durations and, cracking and ductile and brittle fracture surfaces for samples tested in LBE.

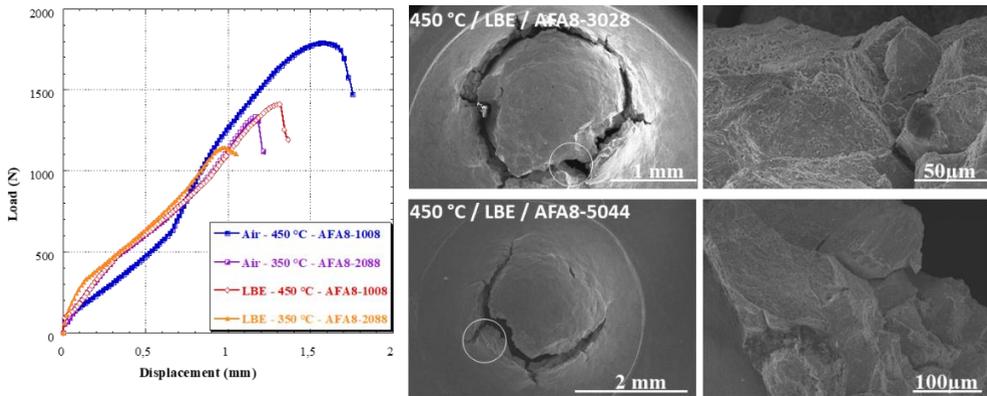


Fig. 6. SPT curves obtained for the AFA8 steel after 2 aging durations (at 0.5 mm/min), and cracking and brittle fracture surfaces for samples tested in LBE.

To summarize, the 650 °C thermal aging does not promote any significant mechanical behaviour evolution for the AFA3 steel tested by SPT in air, but an embrittlement of the AFA8 steel even if it is very localized. So, the aging at 650 °C leads to a modification of mechanical properties of the AFA8 steel without promoting major embrittlement in air and at 350 °C and 450 °C. The macroscopic mechanical behaviour of the AFA3 steel after thermal aging is not affected by the presence of the LBE. In most of conditions, the aging at 650 °C does not promote a LME sensitivity of the AFA3 steel by the liquid LBE. But, some signs of LME have been observed in a minority of samples. Aging AFA8 steel at 650 °C induces sensitivity to liquid LBE embrittlement at 350 °C and at 450 °C. The macroscopic mechanical behaviour of the AFA8 steel is ductile in air as in LBE, but liquid LBE affects the mechanical properties and mechanical behaviour. In presence of LBE, brittle fracture of the AFA8 steel after aging at 650 °C is observed.

To understand the effect of the thermal aging the evolution of the hardness and of the microstructure has been evaluated according to the duration of the 650 °C aging. The main evolution concerns the precipitation and apparition of phases. After 1008 hours, a phase rich in Fe and Cr at grain boundaries and a phase rich in NiAl appear for the AFA3 steel. For the AFA8 steel, we observed a phase rich in NiAl after a 2088 hours thermal aging while a phase rich in Fe and Cr was detected at grain boundaries after a 5044 hours thermal aging. Contrary

to what was observed for the AFA3, the precipitation for the AFA8 steel presents a heterogeneous distribution. Moreover, from a thermal aging of 1008 hours, a significant precipitation of phase rich in W is observed. Concerning the hardness, despite evolutions in grain size and precipitation, few changes are measured for the AFA8 steel according to the thermal aging duration (Table 2). The precipitation of the phase rich in Fe and Cr leads to an increase in the hardness of the AFA3 steel.

Table 2. Hardness (under 1kg) of the two studied AFA steels according to the duration of the 650 °C thermal aging.

	As received	1008 hours	2088 hours	3028 hours	5044 hours
AFA3	167 ± 4	198 ± 7	213 ± 11	221 ± 18	221 ± 13
AFA8	206 ± 6	190 ± 6	183 ± 9	213 ± 16	229 ± 13

Thus, this change in the hardness of the AFA3 steel could explain the local LME sensitivity, as has already been observed and explained for T91 steel with different microstructure states [23-24]. Concerning the AFA8 steel, the LME sensitivity cannot be attributed either to the evolution in the hardness or to the presence of a phase rich in NiAl since LME was observed after a 1008 hours aging. However, precipitates rich in W appear from the first 1008 hours of thermal aging. These precipitates are suspected to be responsible for a large part of the LME sensitivity. The heterogeneity in the distribution of the precipitates rich in W partly explains the dispersion in the results of AFA8 obtained after thermal aging in the presence of LBE.

4 Conclusions

In this work, technical options to mitigate corrosion by lead or LBE were investigated in terms of mechanical performances in liquid lead or liquid LBE. So the LME sensitivity of different materials selected for their good corrosion resistance in lead or LBE was evaluated.

Concerning the 15-15Ti steel protected by a 1.5 µm Al₂O₃ coating, no LME sensitivity for the tested conditions has been observed. But the reliability of the coating for large durations or after an impact/degradation of the surface/coating is questionable. So, as the coating is not self-forming, the corrosion resistance as the absence of LME can not be guaranteed.

The two tested AFA steels (16Ni-14Cr-2.5Al-2.5Mn-1Nb), one without and one with 2% wt.% W and 0.02% wt.% Y, are not sensitive to LME for the tested conditions and in their as received state. But aging the AFA8 steel in vacuum at 650 °C for durations between 1000 hours to 5000 hours promotes modification in the mechanical response and brittle fracture, so LME. This embrittlement seems to be attributed to the precipitation. Optimization of the composition and microstructure is necessary to avoid precipitation which promotes LME of these promising steels. Indeed, AFA steels promote an in-situ forming stable and protective oxide layer from the metallic elements of steel to limit the contact between the liquid metal and the materials.

For their participation to this study, the authors thank Damien Creton, Jocelyn Golek, Yesmine Bayouhdh, Abdessalam Mrabti. Observations and analyses by SEM and DRX were performed respectively at the electron microscope facility and at the DRX platform at Lille University with the support of Chevreul Institute, the European FEDER, and the Région Hauts-de-France (France).

This work has been carried out in the frame of EERA Point Programme of Nuclear Materials, funded by the European Commission HORIZON 2020 Framework Programme under grant agreement No. 755269, the GEMMA (Generation IV Materials Maturity) H2020 program.

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