Overview on Corrosion and Thermal-hydraulic Issues of Liquid Metal Coolants

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The thermo-physical properties of Liquid Metals (LM) with low melting and high boiling temperatures, like the alkali metals with small atomic weight and heavy liquid metals like lead or its alloys, makes them attractive as coolant candidates in advanced nuclear fusion and fission systems. The fast neutron spectrum and the high neutron yield of the spallation reaction enable simple and robust flow structures with high energy densities. Thus, liquid metals are attractive for the development of neutron spallation sources, for fusion blankets, as core coolant of fast reactors and for heavy ion fragmentation experiments. However, the practical use of LM still needs to be demonstrated by experiments and by numerical predictions. With this objective, the KALLA (KArlsruhe Liquid Metal LAboratory) programme has been established as a shared initiative of several institutes merging the individual aspects of fluid and thermal dynamics, material sciences, measurement technique development and classical nuclear engineering. KALLA consists of several stagnant and loop experiments and is representing one of the largest infrastructures in Europe[1] dedicated to liquid metal technologies.

Within this article an overview over the recent nuclear liquid metal technology developments conducted in KALLA is given. After a brief introduction highlighting the individual fields in which KALLA is engaged some new loop systems erected and projected are presented as well as new tested components. Then a generic heat transfer experiment along a rod is shown aimed at qualifying computational codes is described, since it forms the basis for the design of a fuel assembly of a future nuclear reactor. Here, the design of a scaled assembly experiment is shortly given. In the third part a new measurement technique to measure non-intrusively liquid metal surfaces is shown before material aspects in terms of corrosion and corrosion protection by means of alloying are discussed. Finally, the main issues are summarized and an outlook over the future developments planned in KALLA is given.

I. INTRODUCTION

Liquid metals are currently experiencing a renaissance in nuclear engineering since they offer unique thermo-physical and nuclear properties and therefore are facilitating simple and robust structures allowing high power densities in simultaneously small volumes. This bi-functionality is not only of interest in classical nuclear technology like LM-cooled fast breeder reactors (operated e.g. with sodium[2]; lead-bismuth eutectics[3] (LBE) or pure lead[2]) or fusion breeding blankets and divertors[4] but also in new reactor concepts like accelerator driven systems (ADS) aimed at minimizing the radiotoxicity long living fission products. Besides these classical nuclear engineering field dealing with the energy production aspect more recently applications come up, in which high power density charged particle beams (either protons, electrons or ions) are coupled to liquid metal cooled targets. These types of large scaled experimental facilities occupy a large spectrum of physical research areas ranging from material or structure of matter research, health care, instrumentation development etc.. Here, only the international material irradiation facility[5] (IFMIF) or the FAIR-project[6] are named as some of the most prominent examples. Especially the development of the latter named applications with their trend towards more powerful accelerators has been conducive to the whole field of nuclear liquid metal technology development, because the temperatures generated by those beams within the coolant or/and the structure materials are far beyond the limits of conventional systems and very often beyond material sustainable limits.

Liquid metals independent if they belong to the high Z-material class like mercury, lead or its alloys or to the low Z-liquids line represented by the alkali metals like lithium sodium and the corresponding binary alloys require dedicated technological solutions specially adapted to them. This does not only scope all facets of fluid dynamics and heat transfer as turbulence, stability, evaporation/condensation or (shock) wave propagation, but also thermo-chemical aspects of wetting, purification and fluid conditioning. Strongly correlated with the thermo-chemistry are material aspects (corrosion/erosion, embrittlement, fatigue, dissolution attack, etc.), for which dependent on the liquid metal chosen and the expected nuclear damage rate an individual technological solution has to be developed. In order to avoid the degradation of the structural material a fluid conditioning is necessary in almost all cases demanding specific sensing elements and installations within the loop. Also the detection of the local and the global thermal-hydraulic state of the liquid metal system in terms of for instance flow rate, pressure necessitates instruments adapted to the elevated temperatures and the aggressive environment.

In this frame the *KA*rlsruhe *L*iquid metal *LA*boratory (KALLA) program consisting of several stagnant and loop experiments, has been defined. Currently KALLA is one of the most relevant infrastructures in operation within Europe. The interdisciplinary structure of KALLA composed of engineering, thermal-hydraulics and material sciences as well as its technical capabilities make it possible to evaluate thermal-hydraulic parameters in complex geometries, to develop techniques for local and global quantities measurement, to assess the materials compatibility in different conditions and to evaluate basic chemical–physical data as for instance the wetting capability of the liquid metal. In addition material development especially surface alloying and modification by pulsed electron beams to increase the resistance of existing steels against corrosion attack of lead alloys is performed. Beside their compatibility also the mechanical properties under relevant conditions are investigated. The whole experimental program is accompanied by modelling and numerical groups.

II. KALLA LOOP SYSTEMS AND COMPONENTS

The name KALLA embraces all experimental and numerical activities in liquid metal technology, thermal-hydraulics and material research with different loop experiments and different liquids such as lead, lead-bismuth, sodium, gallium, indium-gallium tin and lithium. The individual facilities, which named below, can be divided in two sub-groups; the first one involves stagnant liquids (mainly Pb or $Pb^{45}Bi^{55}$) while the second concerns flowing liquid metals. A more detailed description may be taken from (7). Each KALLA test facility is equipped with the Karlsruhe Oxygen Control System (OCS). The facilities using stagnant fluids are:

- COSTA 1 to 6 (Corrosion test stand for Stagnant liquid lead Alloys)
- KOSIMA 1 to 6 (Karlsruhe Oxygen Sensor In Molten Alloys)
- KOCOS (Kinetics of Oxygen Control Systems)
- OCEAN (Oxygen Controlled Exp. container)

In addition, four KALLA loop experiments are operating, all controlled with an active OCS. These loops are available also to external users, which is supported by the integrated infrastructure initiative VELLA[8]. The liquid metal inventory and the most relevant technical data of each loop are summarised in Table 1. These are:

- THESYS (Technologies for HEavy metal SYStems),
- THEADES (THErmalhydraulics and Ads DESign),
- CORRIDA (CORRosion In Dynamic lead Alloys),
- ALINA (Anlage für LIthium und NAtrium)
- TELEMAT (TEchnology Loop for LEad MATerial Qualification)

Within this article we concentrate mainly on the presentation of the new facilities which are currently set-into operation or will be set into operation in the near future.

The first one is the oil-cooled convertible sodium/lithium operable ALINA loop which is related to windowless targets with free surfaces as well as to small scale generic heat transfer experiments. A plan is shown in Figure 1a. More advanced reactor studies attempt to reach higher operation temperatures and thus to increase the net efficiency. Therefore, temperatures beyond 550°C are under consideration involving a dedicated material qualification program. In this context the high temperature loop TELEMAT (design temp. 750°C, Figure 1b) operated with liquid lead is projected and already partly fabricated. It is 8-shaped consisting of a low temperature part (450°C) and a hot branch coupled by recuperator owing a cold shield. The super-heated is realized by means of radiation heaters operating at ambient pressure. In TELEMAT in a first stage, the corrosion/erosion effects of flowing liquid lead will be investigated for potential cladding materials.

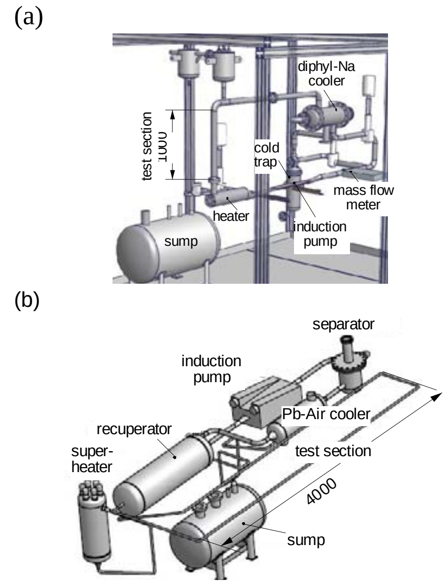

Fig. 1 (a) sodium operated ALINA facility; (b) lead loop TELEMAT. Dimensions are given in millimetres.

TABLE I. Characteristic data of loop experiments in the KALLA laboratory

	THEADES	THESYS	CORRIDA	ALINA	TELEMAT
Liquid	$Pb^{45}Bi^{55}$	$Pb^{45}Bi^{55}$	$Pb^{45}Bi^{55}$	Na,Li	Pb
Pressure head [bar]	5.9	3	2	3.5	2.3
Flow rate [m^3/h]	47	16	9	21	2.1
T_{max} [°C]	400	550	650	400	750°
Inventory [litres]	4000	300	280	150	200
Test ports	4	2(+2)	2	1	2
test sections size [m]	3.85	4.1	1.2	1.0	3.8
El. Power [kW]	1200	250	250	120	110
Operation [h]	3000	2500	18000	100	-
Start-up [y]	2002	2005	2003	2007	2007(8)

One of the crucial components of all LM loop systems is the pump. In many nuclear applications induction pumps are used despite their low efficiency compared to turbo machines or piston pumps because they service several advantages, such as:

- low maintenance costs due to absence of sealings, bearings and moving parts (wings or pistons);
- low degradation rate of the structural material due to low differential velocities between fluid and wall;
- simple replacement without cut of the piping system;
- fine regulation of flow rate by different means;
- change of pump characteristics without change of the mechanical set-up.

Unfortunately, the costs of fabrication especially that of the inductor system is one of the most economical and time consuming issues since it requires a dedicated knowledge of the principle of induction machines[9,10]. A low cost alternative at least for laboratory applications provides an ACHIP-induction pump (Alternating Current Helical Induction Pump) recently developed and qualified in KALLA. The ACHIP consists of the stator of an asynchronous machine, in which concentrically a helically shaped liquid metal duct is positioned, see Figure 2.a. The radial directed rotating magnetic field (B) imposed by the stator current supply induces an axial directed electric current (j) within the liquid metal leading to a Lorentz-force according to jxB. If the helical ducts are electrically coupled e.g. by conducting duct walls the induced currents add up axially and hence a higher pressure head is achieved. In order to minimize the eddy current losses and to increase the magnetic induction a soft iron core is installed rotating with the synchronous velocity of the stator. By changing the supply frequency and the current the pump can be fine regulated. In order to avoid a thermal destruction of the stator an air cooling flowing in the annular gap between stator and the liquid metal duct is installed. This holds especially for weak electrically conducting fluids as e.g. Gallium, GaInSn or PbBi in which due to Joulean heating (Ohmic losses) a significant amount of heat is inductively generated. Via a non-ferromagnetic thermocouple (e.g. a Copper-Constantan pairing) the ventilation can be regulated in order to ensure a safe operation.

Without any major optimizations a reasonable overall efficiency of about 14% was achieved in a test with a sodium-potassium alloy. The Figure 2b shows the experimental realization of an ACHIP pump, while in Figure 2c the corresponding pressure-flow rate diagram is displayed for different supply currents at a fixed feeding frequency of f=30Hz.

III. THERMAL-HYDRAULICS EXEPRIMENTS

One of the crucial issues for a reliable fuel assembly design is a sufficient heat removal capacity at all operation conditions in order to keep the cladding temperature within an acceptable threshold. Fortunately, Lead or its alloys exhibit a high boiling point which limits the energy transfer problem to the consideration of a convective-diffusive heat transfer issue. But, liquid metals show a high thermal conductivity, expressed by a low molecular Prandtl number Pr, which yields to a scale separation of the viscous and the thermal boundary layer. This scale separation does not allow to use turbulence models based on the Reynolds analogy for the calculation of the temperature distribution, because of the different statistical properties of the temperature field compared to the velocity field. Hence, the numerical treatment of the turbulent energy transfer in liquid metals requires an additional turbulence modeling for the turbulent heat fluxes and their dissipation. In order to develop such models simultaneously the mean and fluctuating data of the velocity and temperature are required, which necessitates a generic heat transfer experiment with well defined boundary conditions. In a next step based on the generic heat transfer data a fuel assembly simulator is designed, which is presented in section III.B.

(a)

(b)

(c)

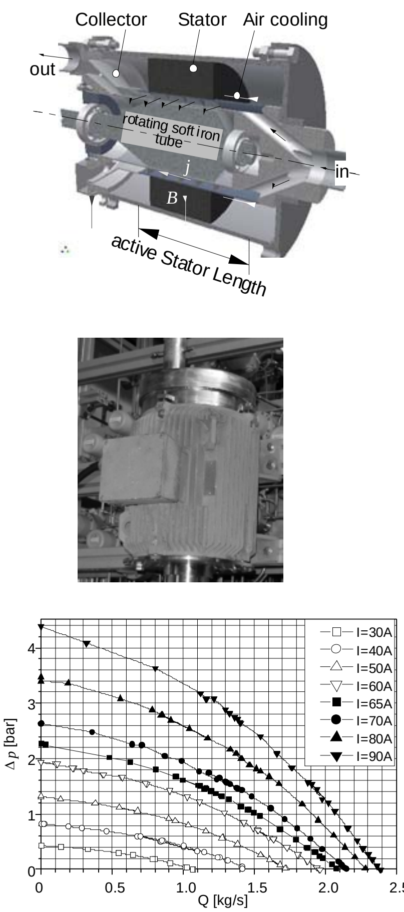

Fig 2: (a) Schematic set-up of a ACHIP pump to circulate LM´s; (b) Photograph of a NaK ACHIP pump installed and (c) corresponding Δp-Q-diagram of the pump shown in (b) for different currents (I) at f=30hz.

III.A. Generic heat transfer experiments

The basic element of a fuel assembly is a single rod. Hence, the detailed knowledge of the thermal transport processes taking place at different flow rates ranging from pure convection via mixed convection to entirely buoyancy driven flows is a major ingredient for the design of a fuel bundle for a nuclear power production or transmutation system.

In order to quantify the validity threshold and to formulate Nusselt number correlations we present here a combined experimental and numerical study of a thermally developing turbulent lead bismuth flow along a uniformely heated rod in a circular tube. This study focuses on one condition typical for an Accelerator Driven System (ADS) or a fast reactor application (Re=2.69·10^5, Pr=0.022, T_{in}=300°C, surface heat flux q''=3.4·10^{5}W/m^2). The data are compared with numerical results. For the later discussion we define Pr, Re, and the Peclet number Pe as follows:

$$Pr = \frac{\nu \rho c_p}{\lambda} \quad ; Re = \frac{u_0 d}{\nu}; \quad Pe = Re \cdot Pr \quad , (1)$$

where ν is the kinematic viscosity, ρ the density, c_p the heat capacity and λ the heat conductivity of the fluid, from (11). The characteristic duct dimension is given by the diameter d and the mean velocity is u_0.

In the experiment a hydraulically developed turbulent $Pb^{45}Bi^{55}$ pipe flow (d=60mm) faces a sharp edged heated rod (d_r=8.2mm) in a concentrical arrangement. In order to minimize the heat losses a thermal insulation with a thickness of 150mm and a heat conductivity of λ_{ins}=0.043 W/(mK) is wrapped around the liquid metal containing ducts. The experiment is installed within the THESYS loop of KALLA. The dimensions of the heated rod, the test section containing the sensing elements are displayed in the Figures. 1. The test section is installed in direction of the gravity vector 2400mm downstream a 90° bend. After the bend a set of flow conditioners consisting of a venturi nozzle, wings and a honeycomb structure is mounted.

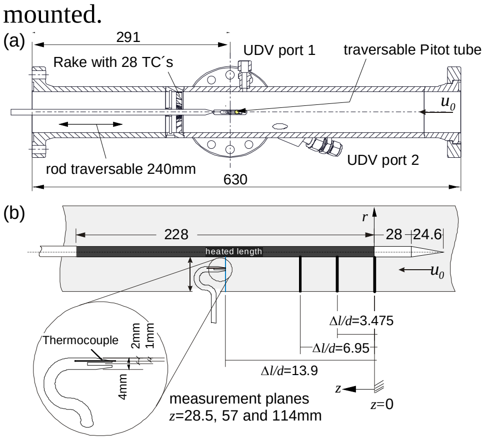

Fig.3: (a) heated rod in the tube with instrumentation; (b) heated rod and coordinate system

The numerical simulations were performed using the STAR-CD code[12]. Here, the flow was considered as highly turbulent and steady. In the calculations axis-symmetry was assumed reducing the problem to a 2D case. To ensure fully developed flow conditions upstream of the heated rod, 2 meters upstream of the heated rod were considered. Further, the following simplifications were been assumed:

Heat losses from the tube to the ambient are neglected.Effects of the side chamber of the Pitot tube are negligible.

The used grid is non-equidistant and structured cells were used. Here, special attention has been paid to the grid generation. Regarding the turbulence models a SST model with a considerably fine resolution (y^+=0.7) was used to get rid of the wall function problem. For the high Reynolds number case (pure forced convective flow) considered in this context, the use of variable density models revealed no significant differences to the constant fluid property version. The simulation was conducted using a first order of hybrid differencing scheme with a constant turbulent Prandtl number (Pr_t) of Pr_t=0.9.

The measurements are conducted at several positions z/d, using a traversable Pitot-tube with two attached NiCr-Ni thermocouples (Tc) and traversing the heated rod. The resolution of the TC´s (ϕ=0.25mm) is ±0.05°K and the flow rate V varies in the range of $\Delta V/V$ of less than ±0.3%;

In Figure 4a the mean measured and calculated radial temperature distributions at z/d_r=22.6 are compared. Both, experimental and numerical values almost coincide indicating that the use of a refined mesh with y^+<1 yields to correct results for a pure forced convective flow. The deviation between both is within the resolution of the TC´s. An impact of buoyancy effects as in[13] was not observed. Larger deviations are found for dimensionless fluid-wall interface temperature along the axis as shown in Figure 4b. Especially, in the middle region of the heated length deviations up to 15% are observed, which have to clarified in further campaigns.

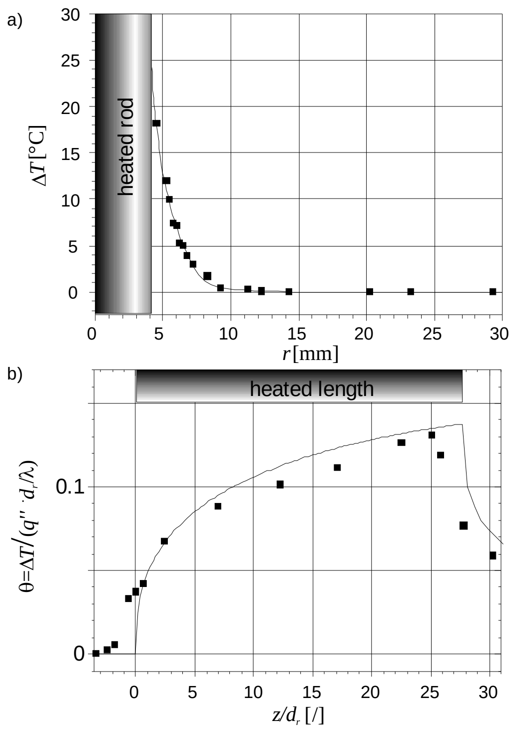

Fig. 4: a.) radial temperature profile at z/d_r=22.6 and b.) dim.-less axial fluid-wall interface temperature. Meas. (■) and calc. (—). Pr=0.022, Re=2.67$\cdot$10^5.

III.B. Prototypical qualification experiments

Based on the single pin/rod experiment a down-scaled fuel bundle experiment is designed. In first step the geometry and the spacer form of the fuel assembly was optimized with respect to a low pressure drop and minimal lateral temperature differences at a given pin/pitch ratio (p/d), power level (q'') and mean flow rate (u_0) using an extended version of the sub-channel analysis code Matra[14].

The set up of the down-scaled test rod bundle is of hexagonal shape and contains 19 electrically heated fuel pin simulators representing a part of the fuel assembly proposed for the PDS-XADS (Preliminary Design Studies of an eXperimental Accelerator-Driven System) that contains 91 fuel pins. The active heated length is 870mm, the power distribution is constant over the active length (q''=100W/cm^2). The schematic overview of the rod bundle and its intended sensor instrumentation is shown in Figure 5.

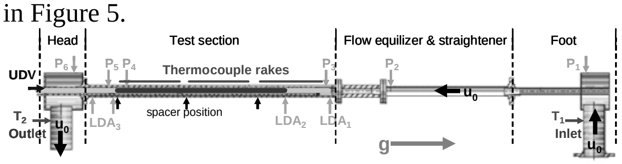

Fig. 5: Complete set-up for the rod bundle experiment inserted into THEADS with the sensor instrumentation. (diagram is rotated 90° counterclockwise).

In a first step the rod bundle will be tested in a water loop at isothermal conditions to determine the pressure drops and the flow distribution in the individual sub-channels and to check the set-up for flow induced vibrations. After that, the heated rod bundle experiment will be carried out at the THEADES loop in KALLA. The entire rod bundle fabricated of stainless steel is divided into 4 dismountable parts. The foot where the incoming fluid at low velocity will be guided into the upstream tube through 6 vertical slits of 70mm height and 6.5mm thickness at fuel bundle conditions, the flow equilizer and the straightener to get a hydraulically full developed flow. Downstream the test section including the 19 fuel pin simulators is attached. The shroud is hexagonally shaped and the pins are focussed by equally positioned spacers formed as proposed in the XADS-design. At the top a head collector is flanged to the test section which guides the fluid side-wards out in order to mount the fuel pin simulators, which are powered electrically. The detailed fabrication techniques partially new invented for this experiment as well as the computational fluid dynamics (CFD) aided design optimization is described in another paper of this conference.

Besides the reactor application nuclear physics experiments require targets with a dedicated thickness of the liquid metal layer in order to produce the secondary particle flux. Due to the high power densities targets owing a so-called window are often not feasible so that the particle beam hits directly the liquid metal film. In general two classes of windowless targets exist, the semi-bounded flows e.g. the IFMIF target[5], in which the liquid metal has only one free surface part facing the beam while the rest is confined with a slip condition to the walls and the unbounded flows, in which a free jet is surrounded by vacuum, the Super-FRS[6].

The challenging task is to design the target consisting of three domains, the flow conditioner, the free surface region and the collector (see e.g. Figure 6a) in such a way, that in the target region the free surface shape and thickness keeps within a certain relatively small threshold in order to ensure a proper functioning. This is problematic because the heat removal from the require flow velocities in the range of 10m/s, where the flow is highly turbulent. An improper nozzle shape induces secondary flows, which yields in the free part of the target to surface oscillations and waves exceeding the acceptable range.

The usual sequence of developing such kinds of targets is first an optimization of the flow conditioner and nozzle by means of CFD. In context of the development of the liquid Lithium operated Super-FRS target a nozzle shaping with a gradual acceleration in x-direction in form of a polynom of 5th order was found to induce the weakest secondary flow at the outlet of the nozzle. CFD also delivered that a fluid acceleration in the nozzle in both directions (x and z) drastically reduces the operational threshold. The final design is depicted in Figure 6b.

The next step is the verification and validation of the CFD predictions in a water experiment. The Figures 6c and 6d shows the measured and the computed surface shape of a water jet a mean velocity of 5m/s. From both graphs it is visible that even at 5m/s the surface tension leads to a contraction of the jet a few centimeter after the nozzle outlet. Qualitatively and quantitative measurements show an acceptable agreement for the water tests. In a next step liquid metal experiments are necessary, since liquid metals exhibits surface tension values about one order of magnitude larger than water. This leads on the one hand to a damping of surface wavers but on the other hand its yields to a scale separation of the statistics of the velocity field compared to the surface area, for which no turbulence models are available in commercial codes. Thus in order to obtain correlation data between velocity field and free surface fluctuations a generic benchmark experiment, the “circular hydraulic jump”, in which both quantities can be measured simultaneously is designed and currently set-up[15]. This experiment is not only aimed to deliver data for an adequate turbulence modeling and verification but also to qualify non-intrusive measurement techniques for totally reflecting surfaces.

IV. MEASUREMENT TECHNIQUES

LM operated systems require measurement technologies especially adapted to them. Due to the corrosity, opaqueness, the specific electric and thermal conductivity of LM´s numerous counter-measures has to be employed in order to transfer conventional fluid mechanical measurement tools to liquid metal flows. The most crucial aspect is likely the elevated temperature range from 200°C to 550°C or even more. The current trends are described in 11.

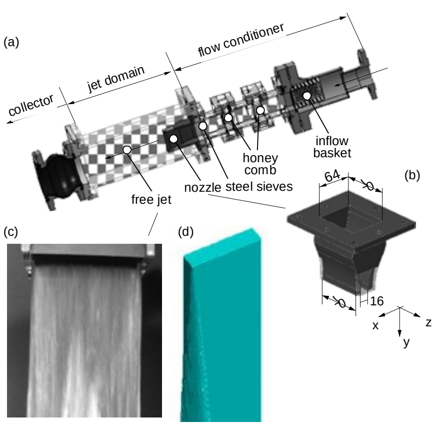

Fig. 6: (a) schematic view on the flow conditioner for the Super-FRS target; (b) CFD aided optimized nozzle shape. (c) and (d) measured and computed surface shape for the water experiment at a mean nozzle exit velocity of 5m/s.

In this context the authors would like to direct the focus to a property unique to LM. They are in the optic sense totally reflecting, which immediately excludes almost all measurement techniques to acquire distances and surface shapes. A non-intrusive highly spatial and temporal resolving technique, however, is necessary to operate liquid metal free surface targets like IFMIF or Super-FRS safe.

For this purpose within KALLA the so-called double layer projection (DLP) technique is currently developed, which is sketched in Figure 7. DLP is based on geometric optics and delivers results with a high spatial and temporal resolution.

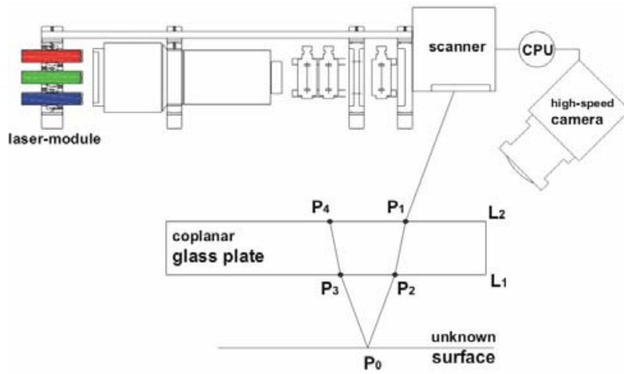

Fig. 7: Schematic of the operation principle of the DLP technique to detect the LM surfaces.

In principle a laser beam is focused by a conventional lens system and guided towards a deflection mirror. From there it is projected onto a co-planar glass plate, in which by diffusive scattering the points P_1 and P_2 are generated. From there the beam proceeds onto the unknown surface, from which it is back-reflected towards the glass plate, in which it generates again by diffusive scattering the points P_3 and P_4. All points P_i are photographed by a high speed camera, see Figure 8a.

In this technique the top and the bottom side of the co-planar glass plate act as two screens, which allows to determine not only the distance of the point P_0 from the glass plate but also its inclination.

The calculation procedure is based on linear algebra; the points P_1 and P_2, which can be evaluated in advance, form a straight g_1, while the points P_3 and P_4 form a straight g_2. The intersection of g1 and g2 in the three room coordinates allows to determine the position P_0 and its inclination. Moreover, since the system is over-determined even an estimate about the error is possible.

However, in practice the laser is not an ideal point source but rather an intensity weighted surface. Due to the transmission path and the reflections the shape and the intensity distribution is distorted, so that specific mathematical algorithms has to be developed to clearly detect the points P_3 and P_4. Also within the glass plate several reflection path occur which require a dedicated exclusion procedure in order to not consider them as the points P_3 and P_4. Finally, the camera observes a picture far away which requires a refractory index correction for P_2 and P_3 and a focal length correction for the individual position where the Points P_i are.

By implementing a scanner instead of a mirror a whole surface area can be scanned with a high speed. An additional use of an array of laser diodes with different allows a color encoding and double checking of the obtained data. Currently the system has a temporal resolution of 25Hz and ±0.28mm, but the goal is 100Hz at accuracies of ±0.15mm, which seems reachable.

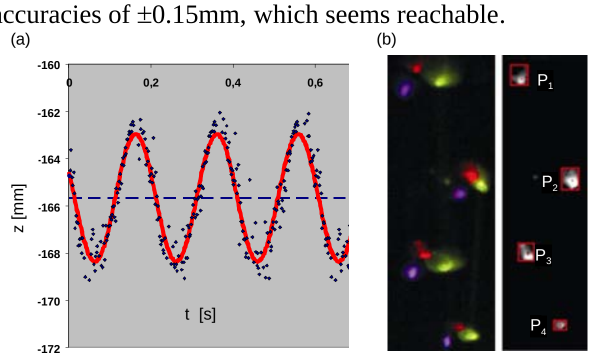

Fig. 8: (a) temporal resolution of the DLP system at a rotating inclined mirror (b) screen shot of the DLP measurement points.

V. MATERIAL COMPATIBILITY

One of the key problems for heavy liquid metal (HLM) especially lead or LBE cooled prospective nuclear and transmutation reactors is the compatibility of structural and cladding materials with the coolant. Unprotected steels are attacked by dissolution of their components in the liquid lead alloys. One measure that is widely being used is dissolution of oxygen in HLM until the oxygen content reaches a level at which oxidation of steel components occurs and protective oxide scales develop on the steel surface [16, 17, 18]. Protection by oxide scale formation requires control of the oxygen concentration. The concentration must be high enough to oxidize iron, the main component of the steels that are considered for application, but should not exceed the value at which oxidation of lead occurs. The concentration range that must be kept to fulfil these conditions is depicted in Figure 9. The $c_{o,s}$ line indicates the upper boarder for the HLM oxygen concentration which extends in the temperature region of interest over 4 orders of magnitude. The same holds for the $c_{o(Fe3O4/FeO)}$ line that indicates the minimal concentration at which iron is oxidized. Since there will be a temperature gradient of about 250°C, the oxygen concentration has to be controlled in a range that stays away from the boarder concentration lines in this temperature range.

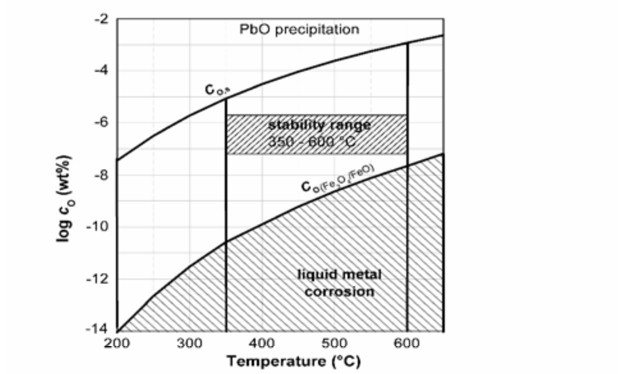

Fig. 9: Range between the solubility limit c_o and the oxygen concentration at which decomposition of iron oxides takes place. The area between the curves indicates the concentrations for which stable conditions are expected.

V.A. Compatibility of steels with liquid HLM

During the last year's researchers in several laboratories examined the suitability of austenitic and ferritic steels as structural and cladding materials in liquid LBE at temperatures relevant for nuclear fission and transmutation reactors [16, 17, 18]. Corrosion tests in static and flowing LBE covered the temperature range from 400 to 650 °C. In most cases the HLM contained around 10^{-6}wt% of oxygen to allow formation of protective oxide scales on the steel surface. Below temperatures of 500°C no difficulties arise with the protection of steel surfaces by formation of an oxide scale. Martensitic steels develop thick multilayer oxide scales growing with the exposure time which probably split off after long times. Austenitic steels have thin, stable, protective spinel scales on the surface. Early experiments on the compatibility of steels and HLM without oxygen showed catastrophic dissolution attack of the HLM onto the unprotected steel [19].

Experimental results of corrosion tests at 500°C in stagnant LBE with 10^{-6}wt% oxygen up to 10000h exposure [20] show clearly the limits of application of steels. The austenitic steel AISI 316 FR (17.3 Cr, 12.1 Ni) in Figure 10, left shows satisfying protection by a thin oxide scale with occasionally thicker oxide nodes like depicted in the upper left specimen cross section with up to 2000h of exposure. However, a dissolution attack starts after 10000h where the oxide scale is penetrated by LBE in some places as shown in the lower picture of Figure 10, left. A different result was obtained for the martensitic steel HCM12a (12 Cr) in Figure 10, right. The initially small oxide scale in the upper right part grows up to a thickness of more than 40µm after 10000h but still protects the surface from dissolution attack. At 550°C surface protection by oxidation of steel components fails because of scale penetration and subsequent dissolution attack on both steels after exposure times below 5000h already [20].

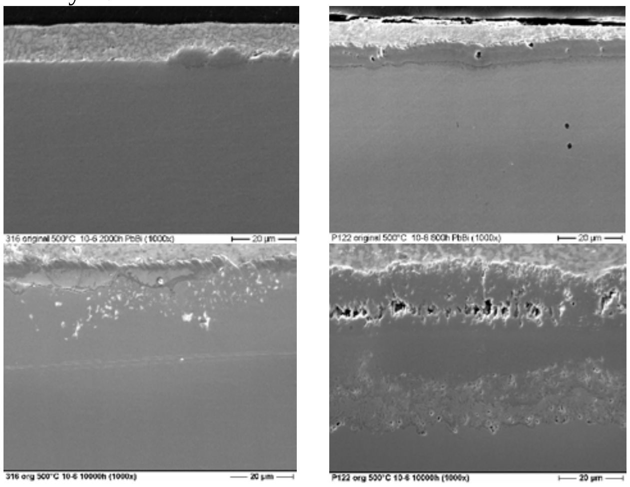

Fig. 10: SEM of steel cross sections through the surface region after exposure to static LBE with 10^{-6} wt% oxygen (20).

Left part: austenitic AISI 316 FR steel after 2000 h (above) and 10000 h (below).

Right part: martensitic P122 steel after 800 h (above) and 10000 h (below).

Steels can be used in HLM with an oxygen concentration of 10^{-6}wt% up to 500°C without any further measures. However, f/m steels form under such conditions already thick oxide scales. On parts of a reactor at which heat removal is a crucial point like claddings or heat exchangers formation of thick oxide scales is not acceptable. Preliminary calculations of the inner clad temperatures [21] show a 10K increase each 10µm of oxide scale considering a thermal conductivity of 1W/mK (which is close to that of magnetite and spinel [22].

Above 500°C the HLM penetrates the oxide scale and starts to attack the steel.

Therefore, only with a coating or surface alloy that ensures the protection against dissolution and oxidation attack austenitic and martensitic steels can be employed in HLM environment above 500°C..

The protective layer has to fulfil following requirements:

- prevention of dissolution attack
- tolerable oxidation rate during oxide scale formation
- long term high temperature stability of the system also under abnormal conditions
- Tolerable influence of the coating and surface alloy on the mechanical properties of the steel
- Durability under irradiation
- Long term mechanical stability of the surface coating and alloy layer
- Industrial feasibility
- Self healing capability of the oxide scale.

V.B. Behavior of steels with modified surface.

FeCrAlY-coatings are well known for their excellent oxidation resistance at high temperatures by forming alumina layers due to selective oxidation[23]. Bulk FeCrAlloy (15%Cr, 4% Al, Fe balance) is already examined by Asher et al [24] in flowing Pb at 700°C and did not show any visible attack also after 13000h of exposure at low oxygen potential. Alumina scales forming on such MCrAlY's are an effective barrier against diffusion of cations as well of anions and prevent, thus, a fast growth of the scale like it is observed with the magnetite and spinel layers on FeCr steel.

Bulk Al containing alloys or thick coatings can not be considered for the envisaged parts (e.g. cladding tubes) to be protected. Only the surface of steels should be enriched with Al. Two methods, alloying an Al layer into the steel surface and coating the surface with an Al-alloy with subsequent GESA treatment, are applied [17, 25].

V.B.a Surface Modification by the GESA process

The GESA process uses pulsed electron beams with a[26] kinetic energy in the range of 50±400 keV, with a beam power density up to 6MW/cm^2 at the target and pulse duration up to 40µs. The energy density absorption of the target is up to 80J/cm^2, which is sufficient to melt metallic materials adiabatically up to a depth of 10-50µm. Due to the high cooling rate in the order of 10^7K/s, very fine grained structures develop during solidification of the molten surface layer. This is a suitable basis for the formation of protective oxide scales with good adhesion[26]. As above described the surface modification process is mainly considered for cladding tubes. Therefore a special GESA the GESA IV (Figure 11) with a cylindrical cathode was designed. This facility allows the treatment of an entire tube of about 30 cm length with a single pulse.

Fig. 11: Picture of the GESA IV facility

At present, coating with an Al-alloy (FeCrAlY) plus GESA treatment, is the favorite process. The coating is applied by low pressure plasma spraying (LPPS). Figure 12 left side shows a cross section of a cladding tube with a ~20µm LPPS FeCrAlY coating[1].

[1] Coating by Sulzer, Wohlen, Switzerland

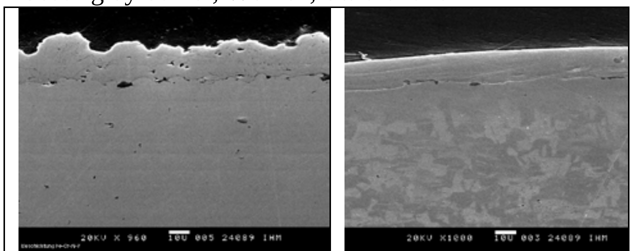

Fig. 12: FeCrAlY coating on austenitic steel 1.4970[1] Left coating after LPPS, right after GESA electron melt treatment (24)

The coating has a rough surface because of the relatively large spray droplets, contains pores and also the adhesion on the bulk material is not perfect. To exclude these inadequacies, the coatings are treated using the GESA melting process. Experiments with GESA show that a GESA treatment smoothes the surface and removes pores. Another important effect of the GESA treatment is that because of the small layer thickness the melting zone reaches beyond the coating substrate interface and, thus causes a mixing of both materials; the coating is "welded" to the bulk material. Figure 12 right side shows such a GESA treated sample [25].

V.B.b Corrosion examinations on surface alloys and coatings

In long time corrosion experiments austenitic AISI 316 FR and HCM12a and martensitic ODS steels with Al alloyed into the surface layer were exposed to LBE with 10^{-6} wt% oxygen. The tests took place over a period of 10000 h at 550 and 650°C without any deterioration of the thin alumina scale that was developed during the initial exposure time [20].

Coating the surface with FeCrAlY leads to the same positive results as observed for surface alloyed steels. The coating behaves like an Al-alloyed surface layer after it is homogenized and the surface smoothed by remelting with GESA electron pulses. Coated T91 steel specimens were exposed 2000h to LBE with 10^{-6} wt% oxygen at 480, 550 and 600°C flowing at 1m/s[27]. There was no dissolution attack, no scale damage and no additional oxide formation observed after the experiments. The result of the surface examination of the specimen exposed to LBE at 600°C is shown in Figure 13, where above the SEM and below the EDX scan is depicted. There is no oxide scale visible, but the EDX scan shows clearly an alumina peak at the surface followed by a Cr peak that seems to be in a metallic state. The EDX shows also the boarder between coating and steel, at which the Al content drops down and some Cr precipitations in the grain boundaries of the coating.

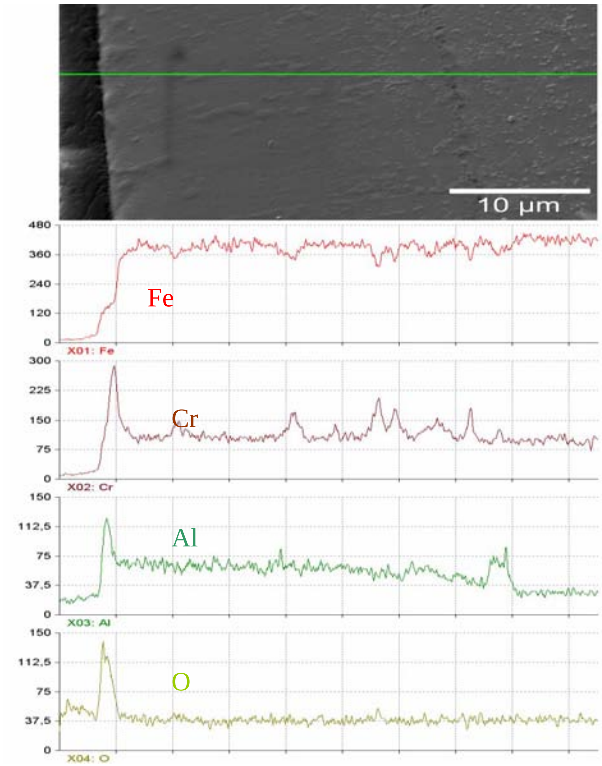

Fig. 13: Surface region of a FeCrAlY coated T91 tube specimen after 2000 h exposure to LBE with 10^{-6} wt% oxygen at 600 °C. Above: SEM of the cross section. Below: EDX elemental scan of the respective region.

Other experiments with FeCrAlY coated steel 1.4970 support the suitability of this coating for high temperature application. No corrosion attack could be observed in 5000 h tests in LBE with 10^{-6} wt% oxygen at 500, 550 and 600 °C (25).

Corrosion experiments with alloyed steels show that 4wt% Al is necessary to achieve selective alumina scale formation. For concentrations above 20wt%, however, the Al activity gets too high and dissolution attack will occur. From experiments performed up to now prevention of dissolution attack, tolerable oxidation rate and long term high temperature stability seems to be fulfilled requirements of the above presented protective layers.

Further going experiments to show the suitability of the surface alloyed layers are described below.

One remarkable experiment was to investigate the enhanced flow corrosion erosion of T91 with and without surface modified layer[27]. A special test section was designed and implemented in the IPPE loop in Obninsk (Figure 14). The velocity evolution was calculated using the Fluidyn code. Samples with and without surface modification were exposed to 1, 1.5 and 3m/s.

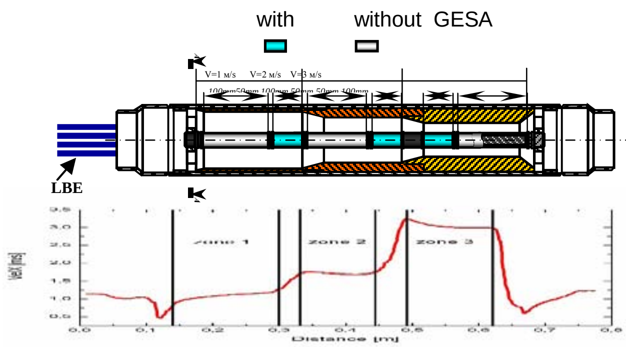

Fig. 14: Set-up and modelling (Fluidyn) of enhanced flow experiment

A magnetite scale is clearly depicted (circle) on top of the 1m/s specimen (Figure 15). At velocities of about 1.5 m/s only small remains are visible at 3m/s the magnetite scale is entirely removed. All three oxide scales are, beside the upper magnetite layer, almost identical in composition and thickness. Therefore, instantaneous removal of the fragile magnetite layer by the flowing LBE is responsible for this behaviour and not a difference in scale growth.

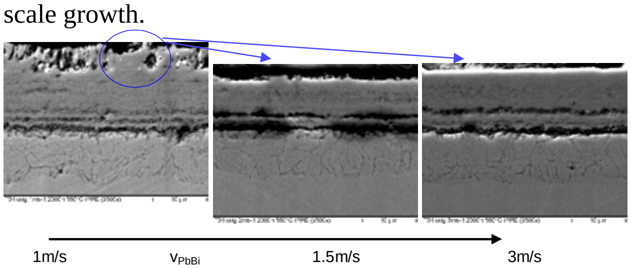

Fig. 15: SEM of T91 steel surface regions exposed 2000 h at 550 °C to flowing LBE with 10^{-6} wt% oxygen. Flow velocities are 1m/s (left), 1.5 m/s (middle) and 3m/s (right)

T91 with surface modified layer do not show any difference in scale growth depending on flow velocity. Thin protective alumina scales like described above cover the steel surface.

One necessary requirement for protective layers is a neglectable influence on mechanical properties. This question about influence of LBE and steel coating was addressed in low cycle fatigue (LCF) tests at 550 °C with 0.5 Hz and elongations $\Delta\varepsilon_t$ between 0.3 – 2.0[28]. The result was that no influence could be observed neither of the LBE with 10^{-6} wt% oxygen nor with a FeCrAlY coating (Figure 16).

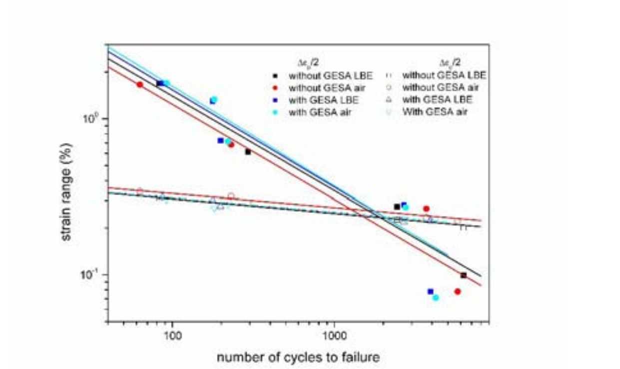

Fig. 16: Fatigue life as a function of strain for T91 with and without surface alloyed layer [28]

T91 cladding tubes with and without surface alloyed FeCrAlY layer are tested under internal pressure at 550 °C in flowing LBE with appropriate oxygen concentration[27]. If an internal pressure of 15 MPa is supplied to the specimen tubes, which causes a strain of 0.7 %, the steel surface is still protected but the magnetite layer observed in Figure 2, above, grows by a factor of 2. Surface alloyed tubes do not show any difference in oxidation at such conditions.

The still open requirement regarding durability under irradiation is addressed in irradiation studies of T91 with and without surface modified layer in the Phenix reactor. Further irradiation experiments also in contact with LBE will be conducted in near future.

An additional topic considered recently is the usability of pump materials (impellers) for lead cooled fast reactors. Screening tests of materials applied in industrial pumps showed the potential of some of the materials. A test stand that is actual taking into operation will allow testing such materials at temperatures up to 600°C in lead at relative velocities of < 20m/s.

Still open questions for a final judgment of the surface alloyed layers are:

- Long term high temperature stability of the system also under abnormal conditions
- Durability under irradiation
- Long term mechanical stability of the surface alloyed layer
- Industrial feasibility

VI. SUMMARY AND OUTLOOK

The KALLA infrastructure organized as an interdisciplinary platform is embedded in different European and national programs dedicated to liquid metal technologies in nuclear fission but also in fusion and generic nuclear physics. In its current state, it is positioned in Europe as one of the larger infrastructures treating all issues related to liquid metals.

This paper gives an overview of the latest scientific and technological achievement obtained in KALLA. The emphasis has been put on the most recent developments in the area of thermal-hydraulics, measurement devices, materials behavior under different temperature, oxygen potentials.

Starting from target investigations, future thermal-hydraulic studies will also concentrate on core components like fuel bundles and spacer problems. They will be extended to study the performance of reactor components like heat exchangers and new advanced pump concepts. In the field of material research and qualification, the focus of the studies will be broadened to investigate more pure liquid lead systems and to intensify the material investigation to higher temperatures. In addition investigation of mechanical properties (e.g. fretting of cladding tubes), erosion resistance of pump materials and resistance to irradiation will be performed. Another issue will be the process development of surface alloying with GESA to industrial level and establishing methodologies for quality assurance of the processed cladding tubes. Regarding the liquid metal adapted instrumentation techniques, more reliable oxygen sensors are being developed. Moreover, an optical technique determining both the free surface shape and distance with a high time and space resolution is developed to investigate the stability of windowless targets appearing in several nuclear applications.

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