3 Safety Research for Nuclear Reactors (NUSAFE): Transmutation -Liquid Metal Technology-

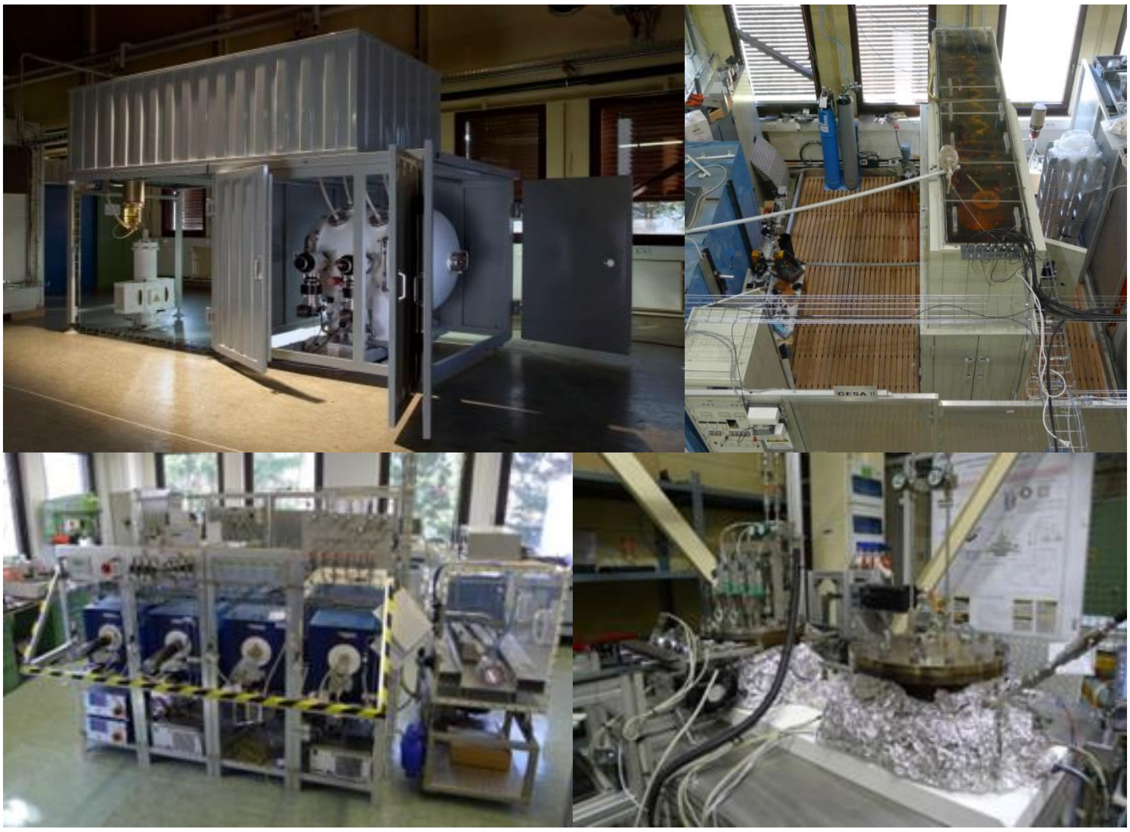
Contact: Prof. Georg Müller

Long-living high-level radioactive waste from existing nuclear power reactors should be transmuted in short-living radio nuclides using fast neutrons provided by a spallation target in an accelerator driven subcritical system or by a fast nuclear reactor. The objective is to reduce the final disposal time of high-level radioactive waste (plutonium, minor actinides) from some 10^6 years down to about 1000 years. Lead (Pb) and lead-bismuth (PbBi) are foreseen as spallation-target and coolant of such devices.

The aim of the institute’s contribution is the development of advanced corrosion mitigation processes especially for parts under high loads like fuel claddings or pump materials in contact with liquid Pb or PbBi. Pulsed large area electron beams (GESA) are used to modify the surface of steels such that they fulfill the requirements of their surrounding environment. Corrosion test stands for exposure of specimens under relevant conditions are developed and operated. Conditioning the Pb-alloy with regard to its oxygen concentration and the transport of oxygen in PbBi are additional aspects of the work.

All tasks are embedded in European and international projects and cooperations like e.g., MaTISSE, MYRTE and GEMMA.

The most relevant results obtained in the reporting period are briefly presented:



3.1 Material development and advanced corrosion mititgation for heavy liquid metal-cooled nuclear systems

Contact: Dr. Alfons Weisenburger

3.1.1 Simulation study of cylindrical GESA

In order to modify tubular specimens at their surface and improve specific material properties, the pulsed electron accelerator GESA in cylindrical geometry with radial converging electron beam is investigated. The previous facility GESA IV was revised and a new design with a 1 m long cathode was developed at the Efremov Institute in St. Petersburg, Russia. Simulations of the electron beam characteristics performed at IHM showed two different operation regimes of the new facility: stable operation with homogeneous beam profile for low cathode-grid voltages and unstable operation with the formation of a virtual cathode between grid and anode for large cathode-grid voltages. Recently, the new accelerator was put into full operation. Although the cathode and grid voltages were chosen to obtain stable operation, first experiments showed an inhomogeneous beam profile and an enhanced current to the grid. These results triggered an advanced simulation study of the operation of the coaxial triode system, investigating the effect of electrons that pass by the anode and circulate inside the accelerator. The number of electrons missing the anode on their first approach depends on the azimuthal velocity distribution at emission, which is determined by the temperature of the cathode plasma. Because the electrons that pass by the anode circulate until striking the control grid, their residence time in the free space of the accelerator and thus their effect on the operation strongly depends on the grid transparency. It was found that circulating electrons shift the limit of the stable operation regime to much lower beam currents. Additionally, the beam profile becomes inhomogeneous and the electron current to the grid increases.

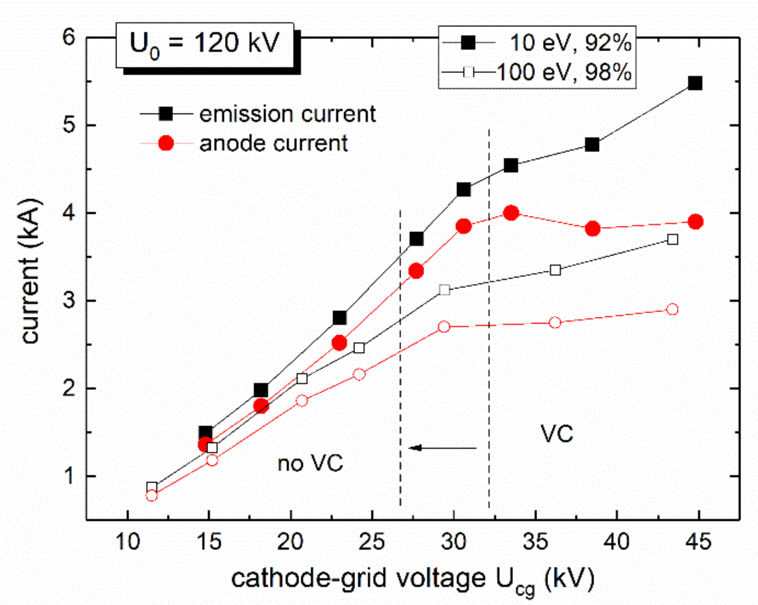


Fig. 3.1.1: Simulated emission and anode currents as function of cathode-grid potential without (closed symbols) and with (open symbols) the influence of circulating electrons.

3.1.2 MATISSE (Materials´ Innovations for Safe and Sustainable nuclear in Europe)

The MATISSE project aimed for the development of GENIV materials. The IHM was involved in three tasks; the development and testing (erosion/corrosion test in CORELLA) of MAXPHASE materials, the improvement of existing ODS by GESA and testing of ODS in Pb alloys under accidental conditions and the development of advanced corrosion barriers based on FeCrAl compositions. The project ended in 2017 and the IHM contributed to one deliverable concerning MAXPHASE materials, and were responsible for two other ones on ODS testing and corrosion barriers develoment.

All the MAXPHASES did not show any erosion/corrosion attack from the CORELLA tests (500°C 1000h) in contrary to the 316 steel reference samples. The best behviour of the tested MAXPHASE showed the $(Nb_{0.85},Zr_{0.15})_4AlC_3$ alloy were absolutely no signs of erosion corrosion could be observed.

The ODS related work concerned their behavior in liquid lead under transient conditions and off-normal conditions like high temperatures as well as under low oxygen conditions. ODS materials used are samples from 9Cr and 12Cr ODS plates in two conditions; as received or with a pulsed electron beam surface treatment (GESA). The latter was applied to remove impurities in the near surface region that are actually discussed as source for local corrosion attack. Additionally, some 14Cr ODS steel sample and specimens with Al alloying into the surface were tested.

After longer exposure time (5000h) at higher temperature (650 and 700°C) both steels 9 and 12Cr ODS with and without GESA treated surface showed dissolution attack. Al alloying in the surface by GESA had a clear positive effect. After 5000h at 650°C no dissolution attack and no thick oxide layer was found.

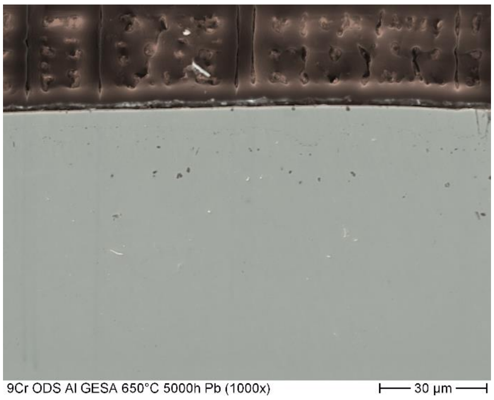


Fig. 3.1.2: SEM cross section of Al-surface alloyed 9Cr-ODS after exposure to Pb at 650°C for 5000h. No dissolution attack due to formation of thin Al-rich oxide scale.

The rapid cooling after the GESA treatment results in a columnar structure that seems to favour diffusion along grain boundary, which can deteriorate the corrosion behaviour. A positive effect by GESA was found on the 14Cr ODS steel at 650°C and 10^{-6}wt%O in the liquid Pb where local inner diffusion was prevented. The temperature transient test 550°C-750°C (24h) -550°C showed no influence on the corrosion behaviour. After an increase to 1000°C, the thick oxide layers spalled off. The thinner oxide layer on the GESA samples seems to be a bit more stable. Results are described in the MatISSE deliverable D4.3.2

The focus of the advanced corrosion barrier development was on the development and optimization of FeCrAl alloys. Avoiding the formation of Cr-carbides by controlling the C content and the ratio between RE like Ti and Zr e.g. and C is one of the key issues for the formation of protective Al-rich scales. 10wt% Cr and 4 wt% Al, given that the RE and C concentrations are strictly controlled and optimized are the values to be respected. These results show that it is feasible to design ductile alumina-forming FeCrAl alloys as construction materials in corrosive environments at temperatures as low as 450°C. For Fe-Cr-Al-based alloys and modified surface layers exposed to molten lead with 10^{-6} wt.% oxygen in the 400-600°C temperature range an experimental criterion was defined concerning the aluminium and chromium content necessary to form a highly protective Al_2O_3 layer. It was found that higher Cr content leads to alumina formation at lower Al concentration. Outside this alumina stability domain, a concentration of 4 wt.% Al is sufficient, in synergy with 16 wt.% Cr content, to reduce drastically the growth rate of $Fe(Cr,Al)_2O_4$ sub-layer with spinel structure, on Fe-Cr-Al alloys exposed to oxygen-containing molten lead. Results are described in the MatISSE deliverable D5.2.1

Collaboration: SCK-CEN, ENEA, KTH, SANDIVK, CEA, …

Funding: EU-Project and NUSAFE

3.1.3 MYRTE (Multi-Purpose Hybrid Research Reactor for High Tech Application)

The IHM is performing fretting tests of wire wrapped fuel cladding samples designed for MYRRHA reactor. Therefore, the existing fretting test facility FRETHME was adapted to the MYHRRA relevant geometry and simultaneously completely overhauled. Fretting tests are conducted at 400°C hot liquid PbBi with a target oxygen content of 10^{-7} wt%. The amplitudes of the relative movement varied between 5 and 300 µm and the loads between 5 and 75 N. All tests are done at a frequency of 10 Hz with a duration between 100 h and 500 h and in cross orientation (contact point) of wire and tube, because previous experiments revealed that only this orientation provides trustworthy results.

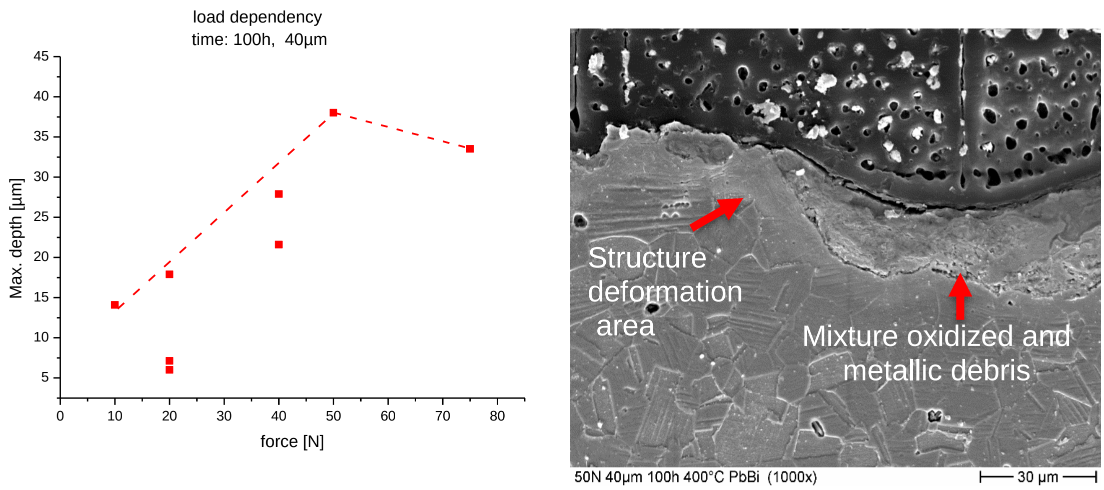


Fig. 3.1.3: Max. fretting depth as function of contact load (left) and etched SEM cross section of fretting area (right).

The maximum fretting depth is increasing with an increasing load up to 50N and decreases by a further increase of the laod. This phenomena can be explained with a required threshold pressure for the debris retention in the fretting area that acts as a kinf of protection layer. Fig. 3.1.3 shows a cross section though a fretting area. The debris is a mixture of oxidized and metallic debris, whereas the amount of metallic debris is increasing to the transition to the bulk material.

Funding: EU Project and NUSAFE

3.1.4 GEMMA

Based on the state of art in alumina-forming austenitic steels, 13 model alloys with Al, Cr, Fe and Ni are designed and prepared. The addition Al (2wt.%~ 5wt.%) aims to form alumina rich oxide scale at elevated temperature; Cr (12wt.%~ 16wt.%) plays a role of "third element effect" and Ni (20wt.%~ 32wt.%) is added to stabilize the austenitic structure. In addition, phase compositions of these models at 550°C and 600°C are also calculated by Thermo-Calc in order to make sure the austenite dominated structures at the test temperature range.

Compatibility with liquid Pb

Only 6 AFA survived corrosion test at 600°C for 1000h in Pb. These surviving model AFA, formed alumina scales combined with chromia on top and contained a minimum amount of 2% Al and 16% Cr. As an example the cross section of a surviving AFA alloy is depicted in Fig. 3.1.4.

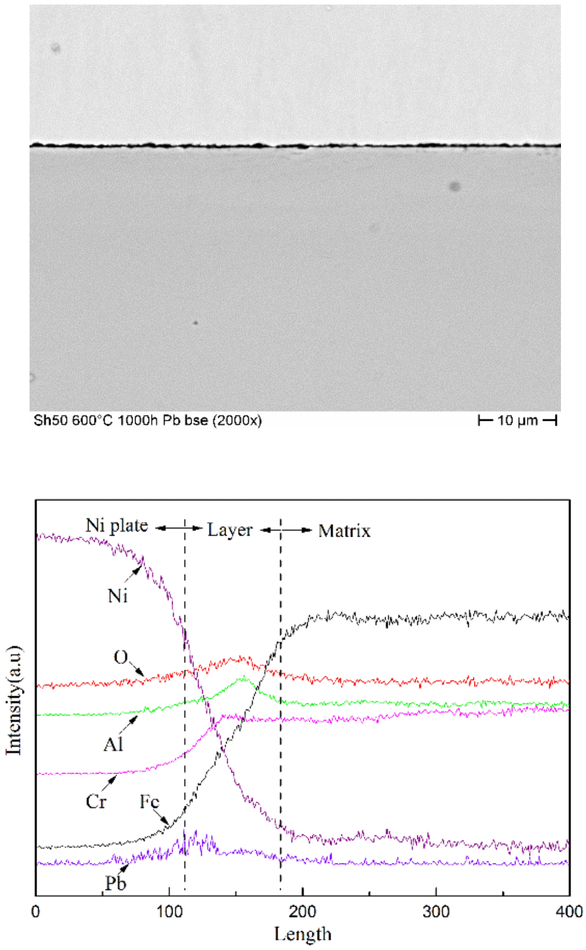


Fig. 3.1.4: SEM cross section (above) and elemental depth profile (below) of AFA exposed to Pb at 600°C for 1000h showing the formation of a protective Al-oxide scale.

Collaboration: IIT, ENEA, KTH, SANDIVK, SCK-CEN,

Funding: EU Project and NUSAFE

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Journal Publications

Fetzer, R.; An, W.; Weisenburger, A.; Müller, G. A. (2017). Pulsed electron beam facility GESA-SOFIE for in-situ characterization of cathode plasma dynamics. *Vacuum*, vol. 145, 179-185.

Fetzer, R.; An, W.; Weisenburger, A.; Mueller, G. (2017). Different operation regimes of cylindrical triode-type electron accelerator studied by PIC code simulations. *Laser and particle beams*, vol. 35 (1), 33-41.