OVERVIEW OF THE P&T ACTIVITIES AT FORSCHUNGSZENTRUM KARLSRUHE

Joachim U. Knebel
Forschungszentrum Karlsruhe GmbH
Programme Nuclear Safety Research (NUKLEAR)
PO Box 3640, D-76021 Karlsruhe, FR Germany
Tel: +49 (0)7247/82-5510, email: joachim.knebel@psf.fzk.de

Abstract

At Forschungszentrum Karlsruhe, which is one of the largest national research centres in Germany within the Hermann von Helmholtz Association (HGF), the research and development activities on partitioning and transmutation are an important topic of the Programme of Nuclear Safety Research (NUKLEAR).

The activities are embedded in the European Framework programmes, especially in the European Thematic Network ADOPT, which summarises all partitioning and transmutation activities and the objective of which is the development of a European ADS Demonstrator as the long-term perspective.

1. Introduction

Irrespective of issues of acceptance and democratic decisions about the use or the discontinuation of nuclear power, the peaceful utilisation of nuclear power continues to make considerable demands on the safety characteristics of the nuclear facilities operated in the Federal Republic of Germany, and on the treatment and management of radioactive residues. Government responsibility and government policies must ensure that the Federal Republic of Germany will continue to conduct intensive research in the fields of reactor safety and safety of nuclear waste disposal, and participate in international projects, including exchanges of scientific experience. This approach is in conformity, for instance, with the Position Paper by Science and Industry in Germany (Berlin, 2001) and the Green Book by the European Commission (Brussels, 2001).

2. Programme NUKLEAR

2.1 Objectives

The Programme Nuclear Safety Research (NUKLEAR) at Forschungszentrum Karlsruhe (FZK) investigates the scientific aspects of the safety of nuclear reactors and the safety of nuclear waste disposal. The results of this work are applied in the public interest in order to keep and to ensure the safety of the nuclear facilities operated in the Federal Republic of Germany, which presently is at the highest international level. Nuclear power makes up for about one third of the electricity production in the Federal Republic of Germany.

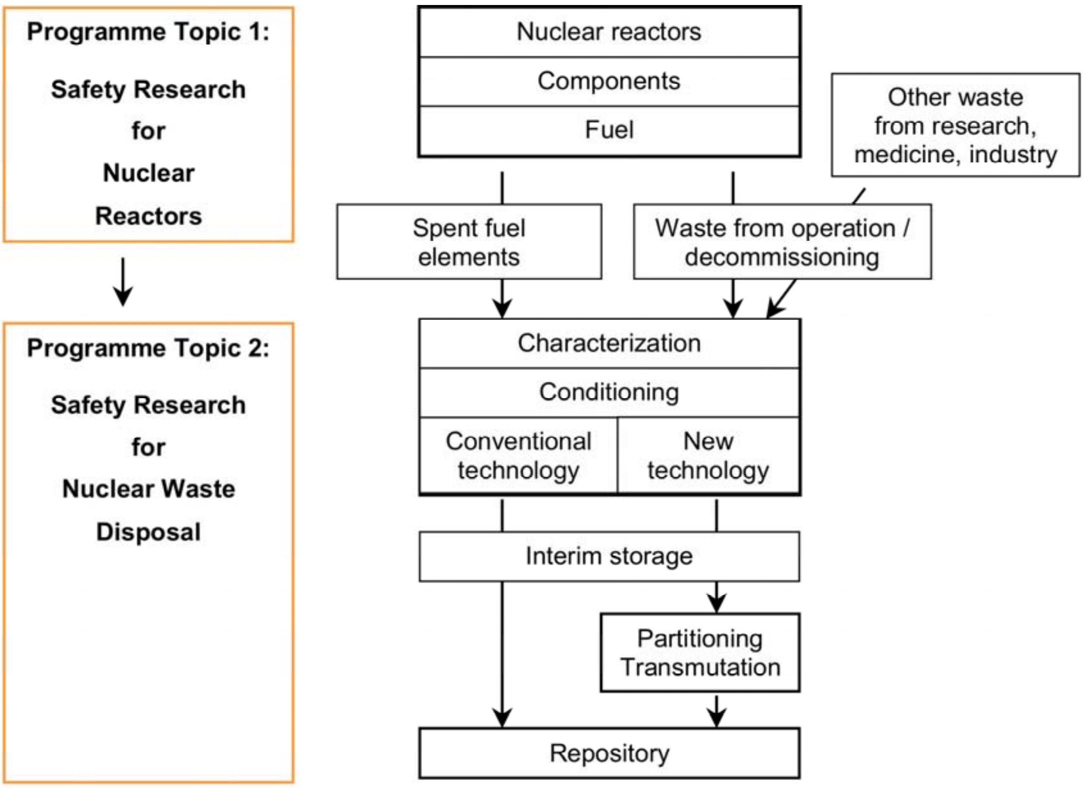
In order to realise this Programme Nuclear Safety Research as long-term provident research, the continuous further development and preservation of the scientific and technical competence and expertise required for the operation of nuclear reactors, the decommissioning of nuclear facilities, and the management of radioactive residues is of outmost importance. This task of maintenance and development of competence is particularly supported by Forschungszentrum Karlsruhe: it is its self-conception to guarantee the availability of skilled nuclear personnel in the long-term, i.e. the whole life-cycle of a nuclear power plant.

Within the Programme NUKLEAR, the following two Programme Topics are conducted (the structure of the Programme and its links to the nuclear fuel cycle is given in Figure 1):

- Safety research for nuclear reactors: Study and evaluation of existing nuclear reactors with respect to design basis accidents, severe accidents, possible radiological consequences, and generic safety characteristics on the basis of advanced R&D work in order to ensure the safe operation of nuclear reactors.

- Safety research for nuclear waste disposal: Characterisation and immobilisation of nuclear wastes, reduction of radiotoxicity (partitioning and transmutation), long-term safety research of nuclear waste disposal and radiation protection in order to ensure the safe and sustainable nuclear waste disposal.

Figure 1. **Schematic diagram of the programme topics of the programme nuclear safety research (NUKLEAR) at FZK and the links to the nuclear fuel cycle**



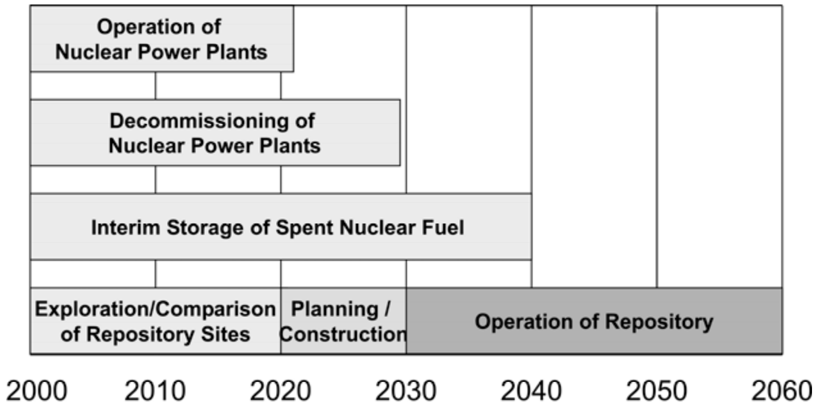
2.2 Long-term strategy

The long-term strategy of the Programme Nuclear Safety Research is formulated under the existing limitations of the present research policy: the political decision to opt-out of nuclear electricity production in Germany until about 2022. If this political decision were to be changed, the strategy of the Programme NUKLEAR will have to be reconsidered.

Looking at a time horizon of about 2060, the following nuclear activities, as is illustrated in Figure 2, have to be performed at a high scientific/technical level and with a highly skilled staff.

The operation of the German nuclear power plants until 2022 has to be accompanied by a high-level research and education programme. In parallel to the operation of the nuclear power plants and their decommissioning until about 2040, the interim storage of the spent fuel, its conditioning and the final safe disposal in a repository has to be guaranteed. These tasks, especially the scientific measures to compare various repository concepts, to choose an adequate formation and to develop/construct/operate (starting in 2030) a final repository, are of typical provident research character and thus in the responsibility of policy. Within the Programme NUKLEAR, the research and development work is focusing on the long-term safety of nuclear waste disposal in Programme Topic 2: this incorporates the option of partitioning and transmutation of highly-radioactive nuclear waste which reduces the radiotoxicity, the heat production and the volume of the highly-radioactive nuclear waste which finally has to be stored in a repository.

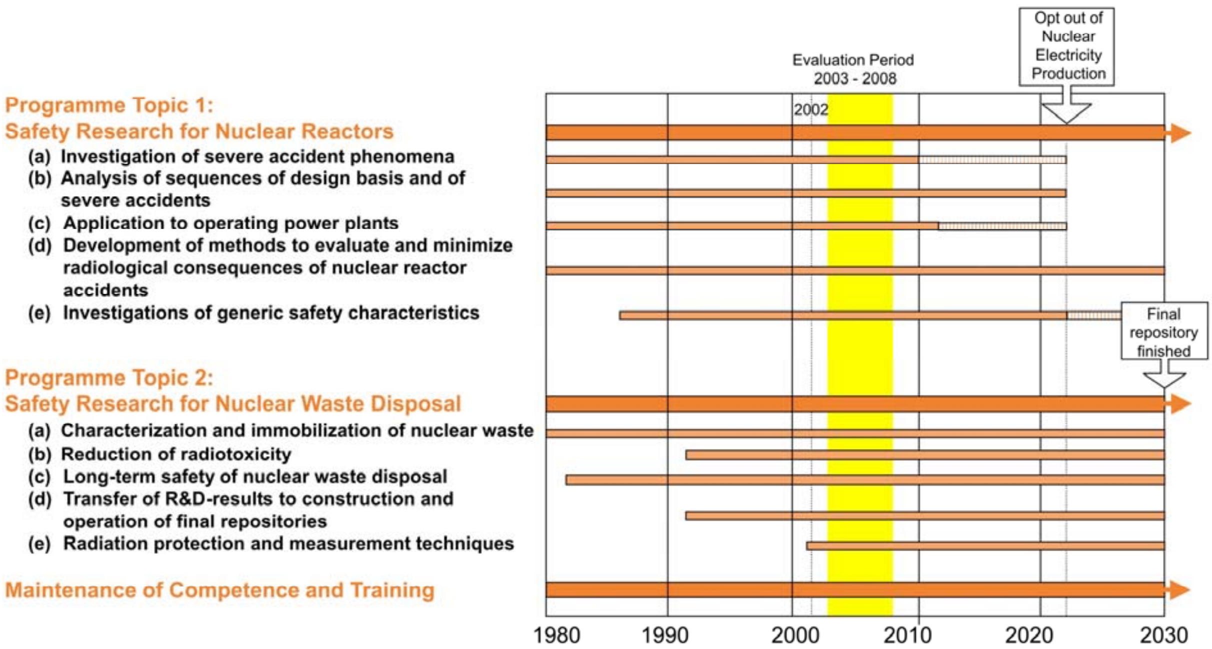
Figure 2. **Required nuclear activities until 2060, under the political decision to opt-out of nuclear electricity production in Germany until about 2022**



2.3 Programme topics

The Gantt chart of the Programme of Nuclear Safety Research and the Programme Topics 1 and 2 are given in Figure 3. The next five-year evaluation period of the Programme is marked in yellow.

Figure 3. **Gantt chart for programme nuclear safety research (NUKLEAR) at Forschungszentrum Karlsruhe (FZK)**



2.3.1 Safety research for nuclear reactors

The starting point of safety research for nuclear reactors is the continued development of theoretical methods and methods of computation as well as the execution of experiments designed to preserve and upgrade the state of knowledge about the safety assessment of existing nuclear reactors. On the one hand, this applies to single phenomena considered in previous risk studies only inadequately or not at all. On the other hand, a consistent mechanistic description of all relevant phenomena in accident sequences is elaborated, and realistic upper limits are indicated for various types of containment loading.

The results of this enhanced R&D work, which are available by late 2008 and 2012, respectively, and which are documented and evaluated by those deadlines, is directly applied in safety analysis and safety evaluations of existing nuclear reactors and in drafting recommendations for action and, if necessary, back fitting measures. With respect to experimental work, a stocktaking exercise between 2007 and 2008 examines the need for further activities after the period of evaluation. The competence for analysing accidents by means of theoretical models must be available right up to the end of the operating lives of German nuclear power plants.

The time up until 2030, i.e. the date when, most probably, a repository will be available, is being spent on upgrading methods of estimating and minimising the radiological consequences of accidents associated with radioactivity releases.

At the same time, basic issues are studied of the generic safety characteristics of nuclear reactors, improved fuels and fuel elements, and scalability so as to provide incentives for further development and improvement of the engineered safeguards features of existing nuclear reactors. This is done by participating in international projects, including exchanges of scientific experience. The work has been planned on a long-term basis until 2022, following the corresponding international projects.

Safety research for nuclear reactors constitutes the basis of the preservation and continuous advancement of competence in the operation of nuclear reactors (according to the opt-out decision, up until 2022) and for the ensuing phase of decommissioning of nuclear reactors.

2.3.2 Safety research for nuclear waste disposal

Safety research for nuclear waste disposal is based on the establishment and evaluation of scenarios resulting in a minimisation and avoidance of radioactive residues, in diminishing the hazard potential of the substances to be emplaced in a repository, and in scientifically based long-term safety research for the final disposal of radioactive residues. This work is planned on a long-term basis approximately until 2030.

Theoretical studies and technical developments are conducted for the characterisation, conditioning, and mainly the immobilisation of nuclear waste so as to transfer it into a state allowing storage over medium- or long-term periods.

In addition to conventional waste treatment, new technologies are developed in accordance with the international state of the art which aims at effectively reducing the long-term hazard potential resulting from the radiotoxicity of high-level residues, especially plutonium, minor actinides, and a number of long-lived fission products. For this purpose, methods and concepts of efficient separation of the minor actinides from high-level waste and the feasibility of transmutation in accelerator-driven sub-critical facilities and in reactors with solid fuel are studied within the framework of the partitioning and transmutation strategy. The safety characteristics of these facilities are of special importance.

The problems of final disposal of high-level wastes and spent fuel elements have not been answered sufficiently world-wide. Within the framework of experimental and theoretical research, the basic principles are elaborated for scientifically-based long-term safety analysis of repositories for high-level waste in deep geologic formations. Transferring the R&D findings to the choice of suitable repository formation and to the construction and operation of a repository are major components of the work. This research work is under the responsibility of the federal government. It is of very high societal priority and, as long-term provident research, designed to extend at least up to 2030. Parts of the work, and the competence acquired in this way, must be continued for the duration of the operation of a repository and upgraded accordingly. This is also true for a future decision about the final sealing of a repository or the retrieval of radioactive residues kept in a repository on the basis of all scientific findings existing internationally at that point in time.

In connection with the dismantlement of existing nuclear reactors, radiation protection is a topic of increasing importance. Under this subject, the capacities that will be released at Forschungszentrum Karlsruhe in the sector of decommissioning of nuclear facilities until about 2008 will be used in order to utilise existing experience for new dismantling concepts and for the development of new methods of residue management and clearance measurement.

2.3.3 National and international schemes of co-operation

The Programme Nuclear Safety Research is part of the national and international research community and closely connected with European and international institutions. On a national level, the Forschungszentrum Karlsruhe is member of the Kompetenzverbund Kerntechnik (Alliance for Competence in Nuclear Technology), of which the Research Centre Jülich (FZJ), the Research Centre Rossendorf (FZR) and the Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) and associated universities are members.

At the present time, no comparable R&D work is being conducted nationally in industry or with the operators, as those parties limit themselves to maintaining the present state of the art. As a consequence, the Forschungszentrum Karlsruhe as institution without any industrial interests takes the job, on behalf of government policy, to examine current findings in the light of the state of the art for applicability to the plants operated in Germany, and to conduct appropriate research projects.

The R&D work described under Programme Topics 1 and 2 are integral parts of the 5th EURATOM Framework Programme under the Nuclear Energy main topic. Forschungszentrum Karlsruhe is co-ordinator of several ongoing EU projects on reactor safety, assessment of accident consequences, partitioning and transmutation, and final storage. As competent partner, it actively contributes to the definition and contents of the 6th Framework Programme. Three important projects, in the preparation of which Forschungszentrum Karlsruhe is involved, are an “Integrated Project on Maintaining Safety Barriers in Severe Accidents”, a “Network of Excellence for Actinide Sciences” and an “Integrated Project on Partitioning & Transmutation,” the latter being closely associated with several integrated projects dealing with dedicated technical aspects of partitioning and transmutation. These projects help to promote and consolidate the contributions by young scientists and engineers.

2.4 P&T activities

In addition to conventional waste treatment, the new technology of partitioning and transmutation is taken up in accordance with the international state of the art, which serves to the effective reduction of radiotoxicity of the long-term hazard potential resulting from the high-level residues. According to the American and the European Roadmaps on Transmutation Systems this research work is scheduled on a time horizon of about 20 years before operation of a transmutation demonstrator or of about 50 years before operation of an industrial transmutation system. For this purpose and embedded in the framework of the partitioning and transmutation strategy of the European Union, the following tasks are worked upon at FZK.

- Performance of scenario studies for the transmutation capabilities of various types of burner reactors, together with the calculation of the fluxes of nuclear materials between the different systems and reactors in the nuclear fuel cycle, applying codes such as KORIGEN and SOLEQ.
- Development and study of methods and concepts of efficient separation of the minor actinides from high-level waste, and application of hollow fibre module extraction units for actinide-lanthanide separation using the highly effective n-Pr-BTP extractant.
- Investigation of the feasibility of transmutation in critical reactors and in accelerator-driven sub-critical systems. Here, the issues of system design, fuel development, neutron-physics of transmutation and blanket layout (applying codes such as KAPROS/KARBUS, and CITATION), safety characteristics of system components and the system as a whole (applying codes such as SIMMER IV and SAS4A) are of special importance. Systems with different fuels (solid or liquid), different coolants (heavy liquid metal or gas) and different energy spectra are considered. Special attention is given to the implementation of inherent and passive safety features.
- Performance of technological studies on heavy liquid metals concentrating on Pb-Bi and Pb technology (handling and operation, measurement techniques, thermalhydraulic bench-marks), on material selection and corrosion protection, and on thermalhydraulic experiments (beam window, fuel element, spallation target) including numerical modelling.
- Participation in international projects such as MEGAPIE and TRADE which aim at performing prototypic experiments on a spallation target of 1 MW of proton beam power and on a low power sub-critical system controlled by an accelerator, respectively.

3. Some results on lead-bismuth technologies

Here, some specific results in the field of heavy liquid metal technology are presented, which arise from the work performed in the KArlsruhe Lead LAboratory KALLA of Forschungszentrum Karlsruhe, and which mark a considerable step forward in understanding technological, corrosion and thermal-hydraulic phenomena in liquid lead-bismuth.

3.1 Oxygen control system (OCS)

The compatibility of structural materials with liquid Pb or Pb-Bi is a critical issue for loop systems containing such metal melts. It is known that unprotected steels are heavily corroded by dissolution of components in the metal melt. The experiments of Russian laboratories clearly show that oxide layers on steel prevent the dissolution process provided that the chemical potential of the dissolved oxygen in the metal melt is kept on a level at which the oxide layer on the steel is stable. It is obvious that this oxygen potential must be higher than that for oxidation of iron, the main steel component, and lower than that of PbO. The formation of PbO would lead to plugging of the cold regions of the loop system. In terms of the Gibbs free energies this condition is given by

$$2\Delta G^{o}_{PbO} > RT\ln p_{O_2} > 0,5\ \Delta G^{o}_{Fe_3O_4} \tag{1}$$

where P_{O2} gives the oxygen partial pressure in the gas atmosphere in equilibrium with the metal melt. The required conditions can clearly be shown by drawing an Ellingham diagram that contains the lines for constant oxygen partial pressures and constant H_2/H_2O ratios as a function of temperature. The latter will be used to control the oxygen partial pressure as follows:

$$p_{O_2} = \frac{p^2_{H_2O}}{p^2_{H_2}} \exp\frac{2\Delta G^0_{H_2O}}{RT} \tag{2}$$

The diagram in Figure 4 demonstrates in which region the stable conditions exist and how they can be established. The ordinate shows the chemical potential of oxygen, the abscissa the temperature. The important region with protective oxide scale formation is the one between the lines of the oxygen potential for PbO and Fe_3O_4 in the temperature regime of 250°C to 650°C.

In order to calculate the oxygen concentration c_0 in equilibrium with the oxygen partial pressure P_{O2}, the activity coefficient γ_0 has to be known. The activity coefficient can be calculated from the relation for the oxygen activity in lead a_O:

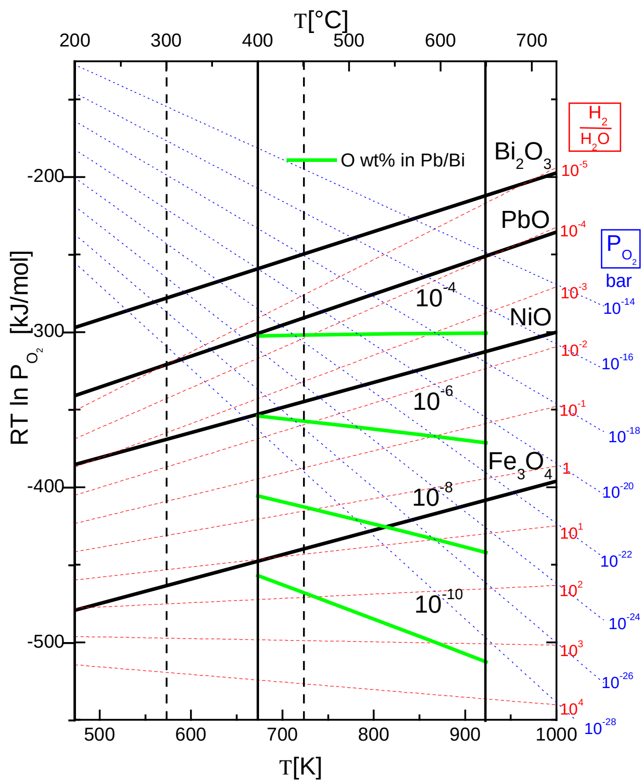
$$a_o = \gamma_o c_o = \frac{c_o}{c_{o,s}} = \left(\frac{p_{o_2}}{p_{o_2,s}}\right)^{0.5} \tag{3}$$

assuming the saturation solubility of oxygen, $C_{O,S}$ and the corresponding oxygen partial pressure $p_{O_2,s}$, as the standard state. It corresponds to the oxygen potential line for PbO. Here the $c_{O,S}$ data for Pb-Bi are used according to:

$$\log c_{O,S}[wt\%] = 1.22 - \frac{3403}{T} \tag{4}$$

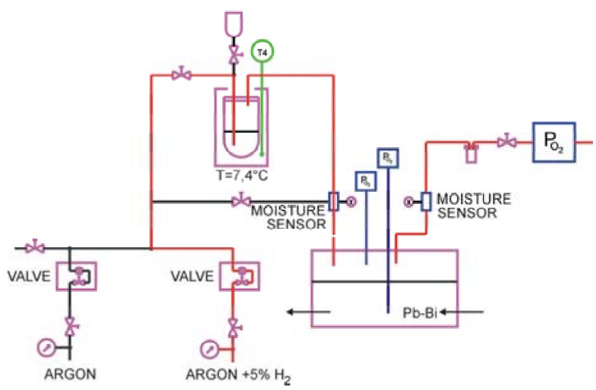
Values at temperatures below 400°C are not well known but may be approximated by extrapolation of the concentration lines. However, this procedure has to be confirmed by additional data for low temperatures. Since a loop works with constant oxygen concentrations, one has to take care that the concentration line does not touch the PbO and Fe_3O_4 oxygen potential lines, which would be a violation of the relation in equation (1).

Figure 4. **Ellingham diagram containing oxides of steel components and Bi, Pb and lines of constant oxygen concentrations in Pb-Bi**



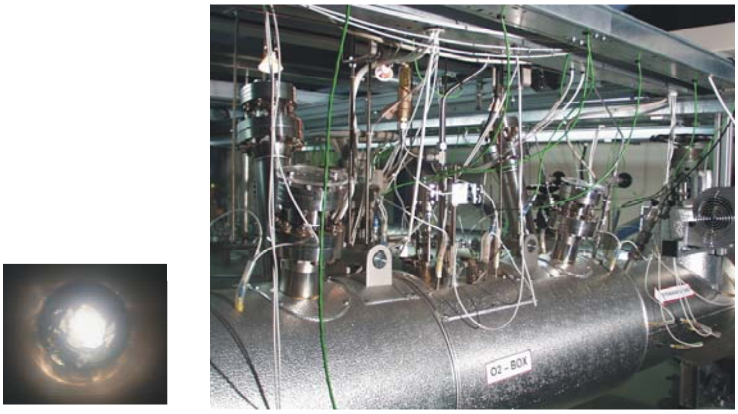
A scheme of the *in situ* Karlsruhe Oxygen Control System (OCS) is given in Figure 5.

Figure 5. **Scheme of the *in situ* Karlsruhe Oxygen Control System (OCS)**



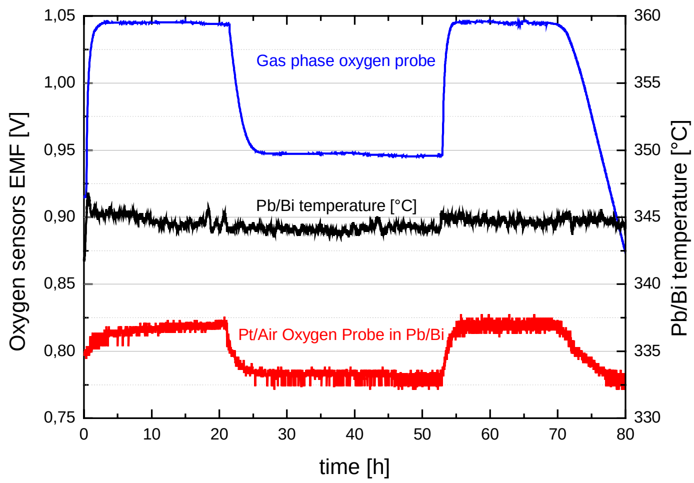
Ar and Ar/5%H_2 are mixed by 2 flow controllers to obtain the required hydrogen concentration in the cover gas. The gas passes then through a moisturiser, the water vapour pressure of which is controlled by a precision thermostat. Before it enters the gas plenum above the flowing liquid melt in the test loop, it passes a moisture sensor. The outcoming gas is checked again by a moisture sensor and an oxygen partial pressure detector. On this principle the Karlsruhe Oxygen Control System was developed and built to control the oxygen concentration in the three large loops of KALLA at FZK. In the Technology Loop THESYS the oxygen control of about 100 l of Pb-Bi inventory is accomplished under flow conditions. Figure 6 shows the gas plenum above the flowing liquid melt, which is equipped with sensors, thermocouples, gas feed pipes, sampling equipment and view windows. The flow direction of the lead-bismuth is from right to left. The cylindrical box is half way filled by the flowing liquid metal. The surface that is exposed to the control gas stream is about 0.14 m^2.

Figure 6. **The Karlsruhe Oxygen Control System operating on the technology loop THESYS of KALLA. Bottom left: looking onto the flowing Pb-Bi through a view window**



The time scale in which the equilibration of the oxygen activity between the gas phase and the liquid metal is reached in THESYS is demonstrated in Figure 7.

Figure 7. **Equilibration of oxygen activities in the gas phase and in the liquid Pb/Bi**



The oxygen activity in the liquid melt, represented by the EMF signal of the Pt/air oxygen meter in the lower curve, follows quite closely that of the oxygen partial pressure change in the gas phase. This latter one is depicted by the signal of the gas oxygen meter in the upper curve. It takes only about 5 h to establish the new oxygen activity in Pb/Bi that corresponds to the new H_2/H_2O ratio of 0.25. The reverse process, i.e. the reduction of the activity to the initial value at H_2/H_2O ratio of 2.5 took only about half of the time. The difference can be explained by considering the hydrogen that gets dissolved in the liquid metal.

In any case the experiment shows that the response of the oxygen activity in the liquid metal loop to changes in the gas phase is fast enough in order to allow an oxygen control via the gas phase in large loops. The Karlsruhe Oxygen Control System (OCS) is a ready-to-use *in situ* device which can be adapted to any lead-bismuth loop.

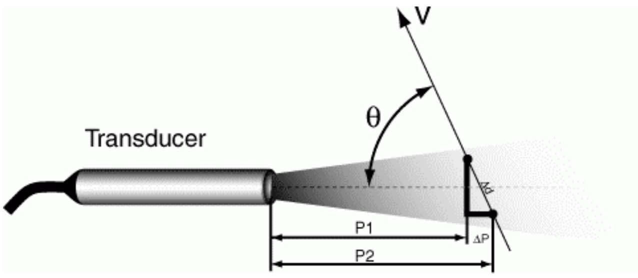
3.2 Ultrasound Doppler Velocimetry (UDV)

In order to validate physical models and to assess the performance of CFD codes which are applied for heavy liquid metal cooled transmutation systems, benchmark experiments are needed that provide a broad database. Mean and fluctuating velocity and temperature data are of prior importance.

During the last years the Ultrasound Doppler Velocimetry (UDV) technique was developed as a very powerful tool to measure the velocity field in liquid metal flow. The ability of ultrasound to travel through opaque media makes this non-intrusive technique interesting for use in liquid metal flows. However, the use in such environments is connected to some specific problems: the temperature range for ultrasonic transducers is normally restricted to temperatures up to 200°C and it has to be guaranteed, that the ultrasonic energy can be coupled into the fluid. The coupling depends strongly on the wetting conditions.

The UDV is able to deliver full velocity profiles in real time by evaluating the echoes of ultrasonic pulses which are sent into the fluid. In contrast to an LDA system, the velocity component of the flow in direction of the ultrasonic beam is obtained, which forms an angle with the main flow. The velocity information is derived from the shift in position of scatterers between two pulses. By sampling the incoming echoes at the same time relative to the emission of a pulse, this shift can be measured. Figure 8 shows the situation for one particle travelling through the ultrasonic field.

Figure 8. **Situation for one scatterer in the ultrasonic beam**



By analysing the time delay between the emission of one pulse and the echo of a particle the distance of this particle from the transducer can be calculated by,

$$p = \frac{c \cdot T_d}{2} \quad , \tag{5}$$

where c is the sound velocity of the ultrasonic wave in the liquid. If the particle is now moving with the flow, its velocity V can be calculated out of the variation in its distance from the transducer.

$$(P_2 - P_1) = V \cdot T_{PRF} \cdot \cos\Theta = \frac{c}{2}(T_2 - T_1) \tag{6}$$

Here TPRF, the Pulse Repetition Frequency, is the time between two successive pulses. The very short time difference T_2-T_1, mostly lower than a microsecond, is measured via the phase shift of the received echoes

$$\delta = 2\pi \cdot f_e (T_2 - T_1) \ , \tag{7}$$

where f_e is the emitting frequency. So the velocity V can be written as

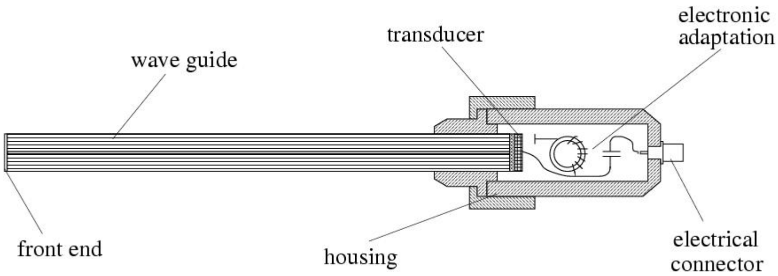
$$V = \frac{c \cdot \delta}{2 \cdot f_e \cdot T_{PRF} \cdot \cos\Theta} = \frac{c \cdot f_d}{2 \cdot f_e \cdot \cos\Theta} \ . \tag{8}$$

The last equation looks exactly like the Doppler equation, where this method has its name from. But the phenomenon used to derive velocity information is different. In reality scatterers are randomly distributed in the ultrasonic beam and a randomly combined echo signal is received. Out of the correlation of these signals the velocity information in different distances from the transducer is extracted.

Having the knowledge of the technique described above, a problem concerning application becomes obvious: the flow has to be seeded with scatterers, which produce the desired echoes. In case of liquid metals one has to rely on impurities which have to be equally distributed in the flow.

In the Technology Loop THESYS of the KArlsruhe Lead LAboratory KALLA a UDV application was tested successfully. To overcome the mentioned problems of high temperature, coupling and wetting, an integrated ultrasonic sensor with acoustic wave guide has been developed within a collaboration of Forschungszentrum Rossendorf (Dresden, Germany) and University Nishni-Novgorod (Nishni-Novgorod, Russia). Figure 9 gives a schematic view of the sensor which has a diameter of 5 mm and a total length of about 25 mm.

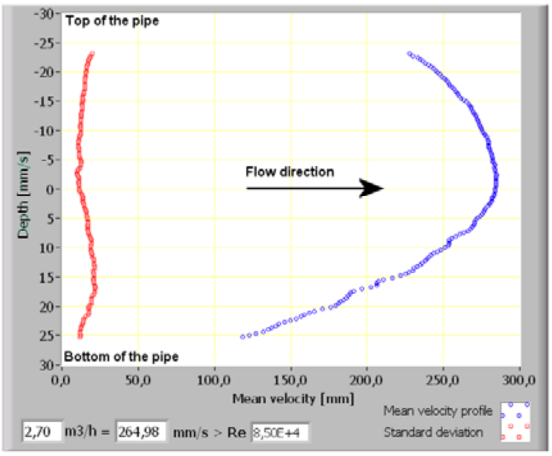
Figure 9. **Schematic view of the integrated Ultrasonic Doppler Velocimetry (UDV) sensor**



The front end of the sensor is in direct contact with the liquid metal and chemically treated to guarantee wetting. Protected by a housing, the transducer itself is located outside of the loop, cooled by air. This design allows as for measurements in fluids with temperatures up to 600°C. The integrated sensor was installed in a measuring port of the Technology Loop with an inclination of 45° with respect to the flow. Figure 10 shows the measurement device (Signal-Processing DOP 2000) and the wave guide installed in the test section (on the left).

Figure 10. **UDV test section in the technology loop of KALLA**

Figure 11. **Mean velocity profile at 300°C (blue) and fluctuation intensity (red)**



In order to check the performance of the integrated sensor and the measurement device, steady-state and transient velocity profiles of a pipe flow where measured at temperatures between 180°C and 300°C for various pump power levels (flow rates). Figure 11 shows the mean velocity profile over the pipe diameter at a temperature of 300°C. Stable velocity signals could be received for about 3 days. After that period, wetting problems occurred again, so further development in the treatment of the front end of the sensor will be necessary, to guarantee a long term wetting. In addition, the question of flow seedings in the liquid metal is not yet satisfactorily answered. The observed echo signals are showing a homogenous distribution of scatterers within the fluid, but at the present stage it is not clear, what kind of particles are responsible for this behaviour. However, especially for turbulence measurements the knowledge about the size and the density of the scatterers is essential for the interpretation of the measured data.

3.3 Enhancement of corrosion resistivity

At the KALLA Laboratory of FZK dedicated experimental equipment for screening tests in stagnant liquid lead alloys, test facility COSTA, and for material corrosion in flowing lead alloys in a loop, Corrosion Loop CORRIDA, have been installed. While the COSTA tests are running since 1998 with a large number of results that help to understand the principles of liquid metal corrosion attack, the CORRIDA loop will start operation by the end of 2002. Meanwhile Russian Pb-Bi loops (IPPE, Obninsk and PROMETEY, St. Petersburg) have been employed to obtain first results on the steel behaviour in flowing lead alloys.

Materials tested in the experiments are the martensitic steels OPTIFER IV, Manet II and the austenitic steels AISI 316, 1.4970. Here, loop tests performed at IPPE and PROMETEY with 1.4970 steel, which is very similar to the AISI 316 steel and which was developed for fast breeder reactors at FZK, are referred to. The specimens were tubes of 8.6 mm outer diameter. A surface layer of 10 to 30 µm is alloyed with aluminium (Al) in one part of the specimens. The surface of the other part is just kept in the as fabricated state. Aluminium alloying was achieved by wrapping an Al-foil around the tube and heating it at 1 040°C for about 0.5 h.

The effect of liquid Pb-Bi with 10^{-6wt}% of oxygen on the original 1.4970 steel specimen after 4000 h of exposure in the Pb–Bi loop is shown in Figure 12: no attack on the surface can be seen at the SEM cross section after exposure to 420°C. The EDX analysis indicates that the steel composition is not changed in the vicinity of the surface. This points out that no dissolution took place. A thin oxide scale protects the material from dissolution attack. During exposure to 550°C the specimen develops thick oxide scales consisting of a magnetite and a spinel zone underneath. The thin oxide layer observed after exposure to 42°C is maintained on some other parts of the surface. Although Pb-Bi has penetrated through the magnetite oxide scale into the spinel zone, no real dissolution attack occurred because no steel components are leached into the liquid metal bath. The Pb-Bi seems to be solidified by dissolution of some iron, chromium and nickel. This arrangement looks, therefore, relatively stable. Deep liquid metal penetration and massive ablation of material by erosion is observed at 600°C on the specimen in the lower part of Figure 12. Bright inclusions consist of a Pb-Bi alloy enriched in Bi. The penetration zone is depleted in Ni down to 1wt%. In this case the penetrating Pb-Bi is not halted by freezing in a saturated state like at 550°C. It dissolved Ni and transported it into the liquid Pb alloy bath proceeding by this process to a depth of >100 µm.

A completely different behaviour is observed for the surface alloyed steel specimens in the loop tests. The specimens show good corrosion resistance at all temperatures. This is demonstrated by the SEM cross sections and the overview picture for the example of the 1.4970 specimen after 4 000 h of exposure in Figure 13. The FeAl layer on top of the specimens is maintained with the original thickness. No dissolution attack or liquid metal penetration is visible even at 600°C. It can be concluded, since also after 4 000 h of exposure no sign of attack is to be observed, that surface alloying with Al can be an appropriate measure to avoid corrosion attack in liquid Pb-Bi.

In conclusion it can be stated that loop tests at 420 to 600°C with 1.4970 steel in flowing Pb-Bi containing 10^{-6}wt% of oxygen reveal bad performance of the steel at temperatures above 550°C because of deep liquid metal penetration and leaching of Ni into the metal bath. No such problems appear with the steel after surface alloying with aluminium. It does not show any signs of liquid metal attack. Therefore, surface alloying of aluminium seems to be the choice for preventing liquid metal attack at temperatures above 550°C.

The next step is to investigate the aluminium treated steels under mechanic stress and irradiation conditions.

Figure 12 **Original 1.4970 steel after exposure to flowing Pb-Bi containing 10^{-6}wt% of oxygen**

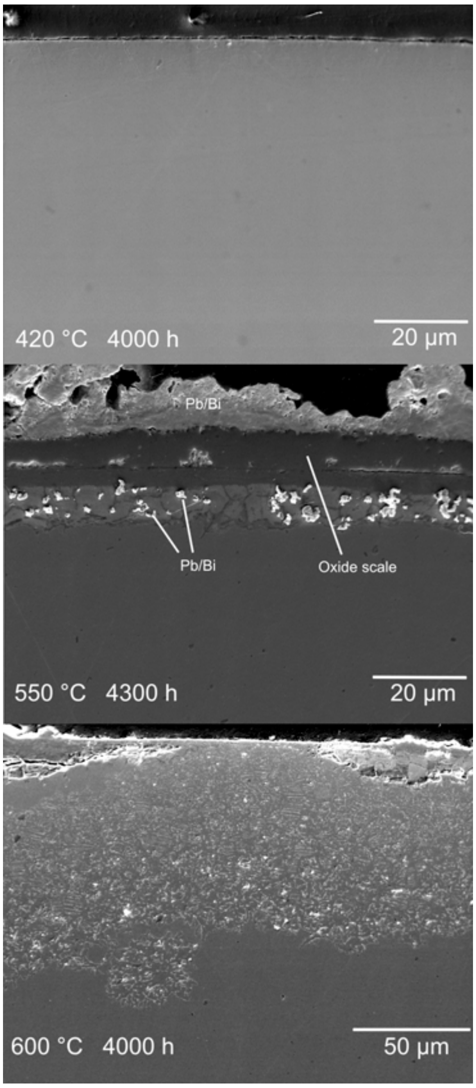
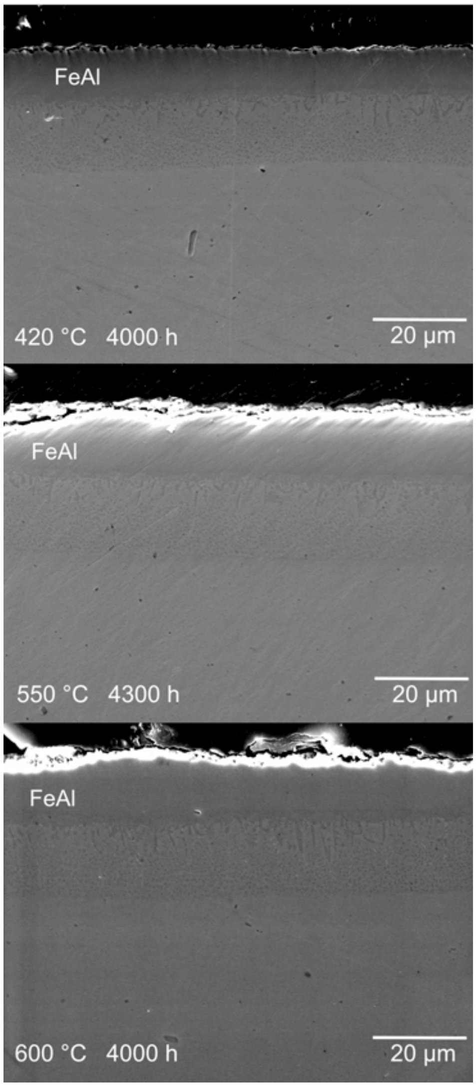


Figure 13. **Al-alloyed 1.4970 steel after exposure to flowing Pb-Bi containing 10-6wt% of oxygen**

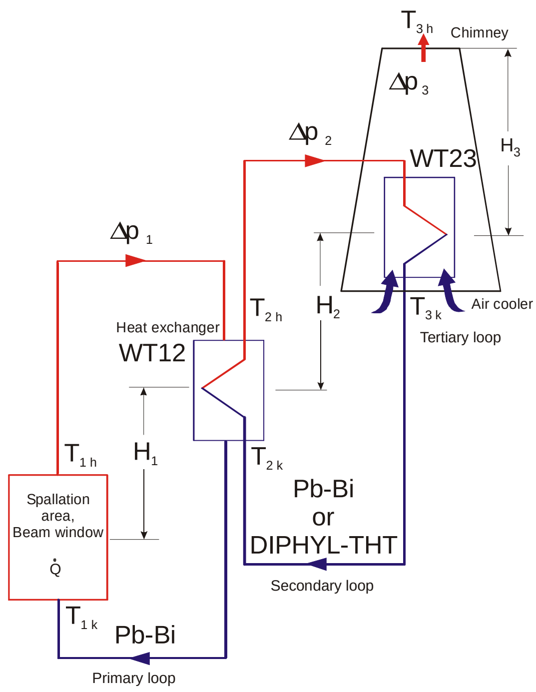


3.4 Spallation target design

A first application of the technological aspects of oxygen control, measurement techniques and structure material protection against corrosion is an Accelerator Driven System (ADS), which consists of an accelerator, a spallation target module and a subcritical blanket. The target module is a technologically new component, the technical feasibility of which has to be assessed and experimentally proved first. With most designs of high power spallation target modules, the spallation material and the coolant is liquid lead or lead-bismuth. Besides the design work within the ISTC 559 Project and the MEGAPIE Initiative, which are both designing liquid Pb-Bi spallation targets of *1 MW* of proton beam power, little experience is existing. Here, the overall geometrical design of a spallation target of *4 MW* of proton beam power together with the heat removal chain, which could become relevant for a European Experimental ADS Demonstrator Facility as is currently being designed within the PDS-XADS Project of the Euratom 5th European Framework Programme is described.

In the following the investigated coupled loop system of the spallation target will be described and calculated for three different cases. The technical feasibility will be checked. The heat removal chain of a closed spallation target consists of three coupled loops as is sketched in Figure 14. The primary loop (index i=1) is the spallation target, the spallation area/beam window being the heat source $\dot{Q}$ and the heat exchanger WT12 being the heat sink. In the secondary loop (index i=2) the heat sink is the air cooler WT23, which removes the heat via an open tertiary loop and a stack (index i=3) to the environment. The relevant parameters such as temperatures T_i, pressure drops Δp_i and driving heights H_i are indicated in Figure 14.

Figure 14. **Sketch of the spallation target with heat removal chain**



The main components are:

- the spallation module itself, consisting of the spallation area within the funnel neck and the beam window, the riser and the downcomer, Figure 15;
- the primary-secondary heat exchanger, being a straight tube heat exchanger working in counter-current mode, the primary side fluid flows in the pipes, Figure 15;
- the secondary-tertiary heat exchanger, being an circular air cooler with inlet and outlet annular plenums, the secondary side fluid flows in the pipes, and a stack, Figure 16.

Here, three different cases are discussed, with Case A being the reference case. It is envisaged to design the heat transfer solely by natural circulation.

Case A: Fluid in primary loop: lead-bismuth (Pb-Bi) at high temperature level

$T_{1h} = 500°C$ (hot riser), $T_{1k} = 350°C$ (cold downcomer)
Fluid in secondary loop: lead-bismuth (Pb-Bi)

For the two other cases the temperature level of the primary loop is lowered by *50 K*. For Case B the fluid in the secondary loop is lead-bismuth, for Case C the fluid is an organic fluid (Diphyl THT of Bayer AG). The lowering of the temperature level is favourable in order to reduce both the high thermal loads on the beam window and the increasing corrosion attack to structure and beam window materials. Corrosion is a serious problem above about *400°C* unless precautions such as an oxygen control system and some surface treatment (thermal restructuring of and aluminium alloying into the top surface layer) is applied. The use of an organic fluid in the secondary loop is cheap and technically simple to handle, however, boiling of the organic oil has to be omitted in order not to deteriorate the heat transfer characteristics of the oil.

Case B: Fluid in primary loop: lead-bismuth (Pb-Bi) at low temperature level

$T_{1h} = 450°C$ (hot riser), $T_{1k} = 300°C$ (cold downcomer)
Fluid in secondary loop: lead-bismuth (Pb-Bi)

Case C: Fluid in primary loop: lead-bismuth (Pb-Bi) at low temperature level

$T_{1h} = 450°C$ (hot riser), $T_{1k} = 300°C$ (cold downcomer)
Fluid in secondary loop: organic oil Diphyl THT

Figure 15. **Spallation target module with primary-secondary heat exchanger WT1**

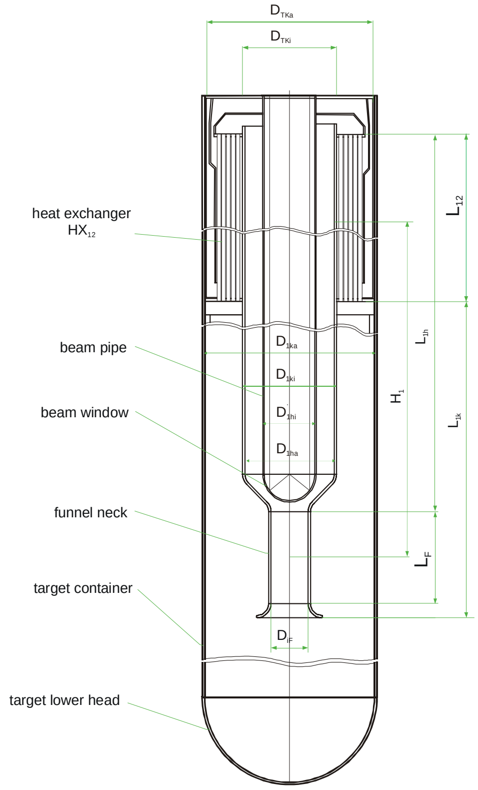
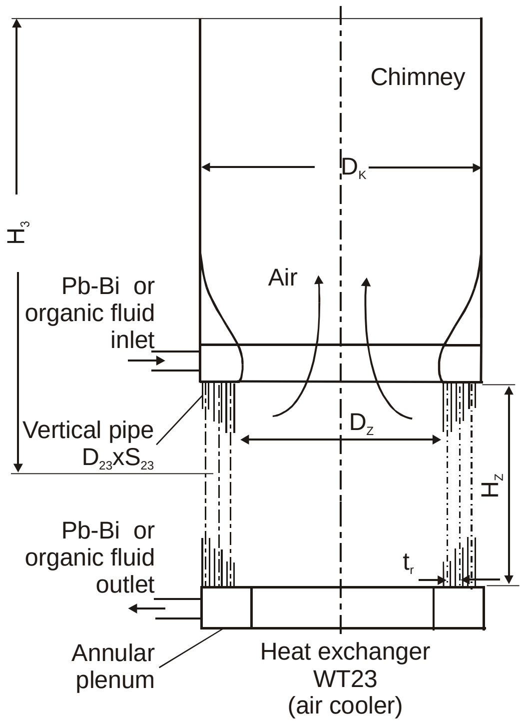


Figure 16. **Secondary-tertiary heat exchanger WT23 with stack**



In all three cases, the lead-bismuth has a maximum heat-up span of *150 K* in order to prevent liquid metal corrosion effects due to leaching out of steel components in the hot regions and deposition in the cold regions of the loops. The air inlet temperature in the air cooler is set to $T_{3k} = 30°C$. When changing the fluid of the secondary loop, the dimensions of the heat exchangers WT12 and WT23 were both kept the same. This is important in order to keep the costs for a future integral experiment as low as possible.

Supposing a spallation heat of $\dot{Q} = 4\ MW$, which has to be removed from the spallation target to the environment, the results for the main parameters such as temperatures, driving heights, pressure drops, flow rate, and length of the heat exchangers, are given in Table 1.

Lowering the temperature level of the primary loop and thus the driving temperature difference between primary and secondary side of the heat exchanger, results in a longer primary-secondary heat exchanger. The effective length L_{12} for Case B is increased by a factor of *1.52* compared to Case A. This increases the total flow resistance, so that the required natural circulation driving heights of the primary and secondary loop and Case B are increased slightly by a factor of *1.07* and *1.08* respectively.

In order to set the same heat transfer coefficient on the primary side for Case C as was calculated for Case B, the Diphyl THT flow rate in the secondary loop has to be significantly increased by a factor of *11.5* in comparison to the flow rate required for Pb-Bi. Then, the temperature increase of the Diphyl THT results in *10 K* only. A comparison between Case B and Case C shows that the length of the heat exchanger pipes has to be increased only by *25%* and *4%* for the primary-secondary and the secondary-tertiary heat exchanger respectively.

The required natural circulation driving height for the primary loop which result in *6.02 m*, *6.44 m* and *6.76 m* for the three cases, are realistic for an ADS application. For Case C, however, a natural circulation heat removal in the secondary loop is no longer possible ($H_2 = 744\ m$). The pressure drop of $4.7 \cdot 10^4\ Pa$ and the flow rate of $713\ m^3/h$ can only be provided by a pump.

The stack has a height of $H_3 = 4.78\ m$ for Cases A and B and of *5.64 m* for Case C, which is reasonable when thinking of a technical realisation.

Besides the integral layout of the spallation target, the local cooling of the beam window has to be investigated in detail using three-dimensional CFD codes. The calculated integral primary side flow rate of $63\ m^3/h$ for all three cases is sufficient to allow a cooling of the beam window which has its hottest spot in the stagnation point (supposing a parabolic radial power profile of the proton beam). The stagnation point is located at the lowest position of the beam window.

Tables 2 and 3 give the heat transfer characteristics of the heat exchangers WT12 and WT23 respectively. When substituting the Pb-Bi in the secondary loop by the organic oil Diphyl THT, the flow rate has to be increased by a factor of *11.4* in order to achieve the same order of magnitude heat transfer coefficient on the secondary side of heat exchanger WT12. The velocity of the primary side Pb-Bi in the heat exchanger pipes is below *0.5 m/s* so that erosion effects are of no importance. The flow both on the primary and on the secondary side of the heat exchanger is highly turbulent. The heat transfer coefficient on the Diphyl THT side of the heat exchanger WT12 and Case C results only in $2\,370\ W/(m^2\ K)$ which is lower than the values of $2680\ W/(m^2\ K)$ and $2640\ W/(m^2\ K)$ for Cases A and B respectively. This is despite the fact that the Nusselt number on the Diphyl THT side is larger than the Nusselt number on the Pb-Bi side by a factor of *92*. Here, the effect of the thermal conductivity is obvious which is two orders of magnitude lower for Diphyl THT in comparison to Pb-Bi.

With heat exchanger WT23 the over-all heat transfer coefficient results to about $71\ W/(m^2\ K)$ for all three cases. This is due to the comparatively low heat transfer on the air side, having a heat transfer coefficient of about $65\ W/(m^2\ K)$ for all three cases, which is limiting the overall performance of the heat exchanger.

Summarising, the heat removal from a *4 MW* spallation target for an Experimental ADS Demonstrator Facility, as is foreseen within the European Project PDS-XADS, is technically feasible under steady-state natural circulation conditions, if lead-bismuth is used as fluid in the primary and in the secondary loop. If the secondary loop is operated with the organic oil Diphyl THT a pump is needed due to the required high flow rate and thus high flow resistance.

Future investigations have to be directed towards the transient behaviour of the coupled loop system and further local three-dimensional CFD calculations for the cooling of the beam window. Finally, the Integral Experiment K4T, simulating the complete heat removal chain given in Figure 1, will be performed at KALLA in order to validate the theoretical findings and model assumptions.

Table 1. **Results for the main parameters**

Symbol	**Unit**	**Case A** Fluid 1: Pb-Bi $T_{1h} = 500°C$ Fluid 2: Pb-Bi	**Case B** Fluid 1: Pb-Bi $T_{1h} = 450°C$ Fluid 2: Pb-Bi	**Case C** Fluid 1: Pb-Bi $T_{1h} = 450°C$ Fluid 2: Diphyl THT
$\dot{Q}$	*MW*	*4.0*	*4.0*	*4.0*
T_{1k}, T_{1h}	*°C*	*350, 500*	*300, 450*	*300, 450*
T_{2k}, T_{2h}	*°C*	*200, 350*	*200, 350*	*263, 273*
T_{3k}, T_{3h}	*°C*	*30, 150*	*30, 150*	*30, 140*
H_1	*m*	*6.02*	*6.44*	*6.76*
H_2	*m*	*2.61*	*2.80*	*744.5*
H_3	*m*	*4.78*	*4.78*	*5.64*
Δp_1	*Pa*	*10 623*	*11 379*	*11 941*
Δp_2	*Pa*	*4 860*	*5 226*	*47 472*
Δp_3	*Pa*	*15*	*15*	*17*
$\dot{V}_1$	m^3/h	*63.4*	*63.0*	*63.0*
$\dot{V}_2$	m^3/h	*62.3*	*62.3*	*713.4*
$\dot{V}_3$	m^3/h	*103 200*	*103 200*	*112 600*
L_{12}	*m*	*0.615*	*0.936*	*1.17*
L_{23}	*m*	*4.88*	*4.88*	*5.07*

Q	spallation heat	*V*	flow rate
T	temperatur	L_{12}, L_{23}	length of heat exchanger pipes
H	driving height	index *1, 2, 3*	primary, secondary, tertiary
Δp	total pressure drop	index *k, h*	downcomer, riser

Table 2. **Heat transfer characteristics of the heat exchangers WT12. Re Reynolds number, Nu Nusselt number, α heat transfer coefficient**

Symbol	Unit	Case A	Case B	Case C
Primary side: fluid 1 flows in heat exchanger pipes.				
Re_{12i}	-	34 020	31 250	31 250
Nu_{12i}	-	6.694	6.766	6.766
α_{12i}	$W/(m^2K)$	8 489	8 242	8 242
Secondary side: fluid 2 flows in free volume between heat exchanger pipes.				
Re_{12a}	-	18 700	18 700	70 320
Nu_{12a}	-	6.375	6.375	584.7
α_{12a}	$W/(m^2K)$	6 265	6 265	4 735
Over-all heat transfer coefficient:				
K_{12}	$W/(m^2K)$	2 682	2 644	2 367

Table 3. **Heat transfer characteristics of the heat exchangers WT23**

Symbol	Unit	Case A	Case B	Case C
Secondary side: fluid 2 flows in heat exchanger pipes.				
Re_{23i}	-	6 414	6 414	24 120
Nu_{23i}	-	5.297	5.297	217.1
α_{23i}	$W/(m^2K)$	6 280	6 280	2 121
Tertiary side: fluid 3 flows perpendicular to pipes.				
Re_{23a}	-	1 874	1 874	1 989
Nu_{23a}	-	43.75	43.75	45.23
α_{23a}	$W/(m^2K)$	64.15	64.15	65.56
Over-all heat transfer coefficient:				
k_{23}	$W/(m^2K)$	71.22	71.22	70.92

4. Summary

The research and development activities on partitioning and transmutation at Forschungszentrum Karlsruhe are embedded in and co-ordinated by the Programme of Nuclear Safety Research (NUKLEAR).

The paper gives an overview on the long-term perspectives of the nuclear activities at Forschungszentrum Karlsruhe which are sub-divided in safety research for nuclear reactors and safety research for nuclear waste disposal.

The work on partitioning and transmutation is mainly concentrating on scenario studies, separation processes, feasibility and design studies on transmutation systems and accelerator driven systems (neutron-physics, thermal-hydraulics, materials, safety), technology studies on heavy liquid metal technologies, and participation in international projects such as MEGAPIE and TRADE.

The paper gives some results on technological, corrosion and thermal-hydraulic phenomena in liquid lead-bismuth, which were achieved in the Karlsruhe Lead Laboratory KALLA. In KALLA, an *in situ* oxygen control system (OCS) which is an indispensable prerequisite to, first, condition the liquid lead-bismuth used as coolant and spallation material, and second, to prevent liquid metal corrosion. It could be shown that the corrosion resistance of steels at temperatures of up to 600°C can be dramatically increased if aluminium is alloyed into the top layer of the steels which is in contact with the liquid metal. In the area of instrumentation an ultrasonic Doppler velocimetry was successfully applied in co-operation with Forschungszentrum Rossendorf (FZR) to turbulent Pb-Bi pipe flow at temperatures of 400°C.

These technological achievements will allow Forschungszentrum Karlsruhe to operate a spallation target module – being 1:1 scaled in power and height – and to investigate its performance under steady state/transient and normal operation/decay heat removal conditions, including the performance of the complete heat removal chain. With the Integral Experiment K4T, in which the spallation heat is produced by specially designed high performance heaters, the geometry proposed by the PDS-XADS Project within the 5th European Framework Programme can be investigated.

Acknowledgements

This work is performed under the basic funding of Forschungszentrum Karlsruhe and partly funded by the European Commission through the Projects TECLA and MEGAPIE-TEST of the Euratom 5th Framework Programme.

The author is grateful to the help and support of his colleagues Georg Müller (IHM), Cord-Henrich Lefhalm and Hans-Joachim Neitzel (IKET) from Forschungszentrum Karlsruhe who produced most of the academic results presented here.