Chapter 14

PERSPECTIVES AND R&D PRIORITIES OF HEAVY LIQUID METAL COOLANT TECHNOLOGIES*

14.1 Introduction

HLM technologies are being developed for a number of advanced nuclear systems. These include accelerator-driven transmutation systems to “burn” high-level nuclear waste and HLM-cooled fast reactors. Accelerator-driven systems are presently under investigation in Europe (EUROTRANS), South Korea (HYPER) and Japan (J-Parc). Pb/LBE-cooled fast reactors are one of the six concepts in the Generation IV Nuclear Energy Systems initiative. Different missions can be envisaged, where the more demanding missions include operating above 600°C for hydrogen production, or very long-life cores for non-traditional uses.

The most current knowledge of HLM technologies for applications below 600°C has been described in this handbook. Viewed from the perspective of programmatic applications for HLM technology and materials, there are a number of technological gaps to be filled before a prototypic test/demonstration nuclear system (reactor or ADS) can be designed, constructed and operated based on lead or LBE cooling. There are also a number of scientific issues which remain open, including fundamental physical, chemical and transport properties of HLM and materials, effects of environment, experimental and computational thermal hydraulics, coolant chemistry, measurement techniques and instrumentation. However, with the exception of uncertainties in materials performance over very long service life in cores (20 years or longer) at temperature over the 500-550°C range, no apparent conceptual barriers (i.e. “show-stoppers”) exist for the eventual use of lead and LBE coolants in advanced nuclear applications.

For applications in the higher temperature ranges an extensive R&D programme on materials and coolant technology is needed. But this is not yet foreseen at the international level. A preliminary analysis of existing materials led to the following classification, depending on the upper operating temperatures:

- *Class I.* For temperatures below 600°C, it has been demonstrated that the existing technologies and some code qualified nuclear structural materials (austenitic and ferritic/martensitic steels) possess acceptable performance in short to medium durations and out of pile. These demonstrations need to be extended to longer durations and under irradiation.
- *Class II.* For higher-temperature services envisioned in more advanced system concepts, materials and coolant technology developmental needs are much more extensive and the development much longer term. For reactor outlet temperature up to 650-700°C for higher efficiencies, ODS (oxide dispersion strengthened) steels and/or advanced F/M steels are potential candidates. These materials, categorised as Class II, may be used with an extension of the LBE coolant technology. For this temperature range, it is likely that Pb, rather than LBE, will be used, although the use of LBE is more established.

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- *Class III*. For operating temperature above 750-800°C in systems with more diverse energy products, including hydrogen production, refractory metal and alloys, ceramics and composites are potential candidates (Class III). These systems will require a very different coolant technology, and design, construction and operating methodology. Compatibility is no longer the key obstacle. Other issues, including irradiation stability, fatigue strength, fabrication, joint, costs, etc are challenging. The development of Class III materials are pursued by high temperature reactor and fusion technology development programmes. For this temperature range, Pb will likely be the only choice since LBE and the associated technologies no longer offer any intrinsic or experiential advantages.

14.2 Technology gaps, R&D needs and priorities for HLM systems operating at temperatures below 600°C

In Europe HLM technology is being developed mainly for the transmutation of high level nuclear waste in subcritical systems. The European project EUROTRANS, sponsored by the European Commission, aims to demonstrate the technical feasibility of transmutation of high level nuclear waste using accelerator-driven systems. Within this objective the design of an experimental facility (XT-ADS), should demonstrate the technical feasibilities: 1) to transmute a sizable amount of waste; 2) to safely operate an ADS. In addition, a conceptual design of the European Facility for Industrial Transmutation (EFIT) is also foreseen. The XT-ADS and the EFIT are subcritical reactor systems cooled by liquid lead-bismuth eutectic (LBE) and Pb, respectively. For both systems, a liquid metal neutron spallation target is the reference design basis for the external neutron source as driver.

The current XT-ADS and the EFIT design concepts foresee an outlet temperature for the HLM coolant no higher than 480°C and a maximum temperature of the fuel cladding at about 550°C under normal operating conditions.

Furthermore the European Lead-cooled System (ELSY) project has been outlined for the study in Europe of an about 600 MWe lead-cooled fast reactor. This project will be the European contribution to the LFR of the GenIV initiative and at present it is under evaluation at the European Commission for funding. The HLM technology specifications for ELSY will take advantages from the results coming out of the DEMETRA domain of the EUROTRANS project. However, some specific Pb technology developments have been foreseen as for instance high temperature materials characterisation in Pb and steam generator tube rupture studies.

In the US, HLM technology is developed in two advanced nuclear energy programmes: the Advanced Fuel Cycle Initiative (AFCI) and the GenIV Lead Fast Reactors (LFR). Within AFCI, there is no current system design effort. The basis is from the earlier concepts of accelerator-driven transmutation of waste, which are similar to EU's ADS. Within GenIV LFR, while there are a range of envisioned missions and system concepts, the current focus is on a class of small- to medium-sized reactors with long-life cores (20 years or longer) and no on-site refuelling. The peak cladding temperatures in the design is limited to 650°C while core outlet temperature is approximately 560°C.

In Japan, there are several programmes developing HLM technology. The J-Parc has a transmutation component to design, build and test a LBE-cooled accelerator-driven transmutation system. A LBE spallation target demonstration is currently being pursued. The system conditions are similar to the EUROTRANS ADS concept. Other programmes are developing a range of concepts using HLM in advanced reactors or in intermediate heat exchangers, with conditions similar to or slightly less demanding than the US LFR specifications.

The situation in South Korea is similar, with HYPER (ADS) and PEACER (reactor) programmes developing HLM in collaborations with international partners. In particular, PEACER aims to design a low temperature LBE-cooled transmuter, well within the condition limits of other concepts.

14.2.1 HLM thermal-physical properties

This handbook contains most relevant thermal and physical properties of Pb, Bi and LBE with recommended correlations. For the high temperature range, a set of thermodynamic and transport properties (thermal conductivity, viscosity and surface tension) of LBE has been developed for use in the reactor safety analysis. However, due to a lack of experimental data published in the open literature, the basic properties such as the liquid density, vapour pressure and liquid adiabatic compressibility, were estimated up to the critical point using semi-empirical models based on the extrapolation of low temperature data of LBE or its constituents. A recommendation to produce experimental data in the high temperature range has been given, which would be of use to validate the computed values.

14.2.2 HLM chemical properties

The chemical properties data of solubility and diffusivity of oxygen and some metallic elements (e.g. Fe, Cr, spallation products as for instance Po, etc.) in the liquid metals and some oxides (e.g. iron oxides, chromium oxides, etc.) are of paramount importance for:

- the assessment of the materials corrosion rate;
- the design and engineering of HLM purification systems, for the development of a corrosion protection strategy that is based on protective oxide layers on the structural materials;
- the source term assessment.

A certain effort is needed to produce reliable solubility and diffusivity data.

14.2.3 Materials

The study of material properties changes in radiation environment, and specifically for HLM systems, the combined effects of radiation and corrosion, is a high priority R&D need. For instance the use of protective oxides as barriers against liquid metal corrosion may be compromised by radiation enhanced transport of ions in oxides, resulting in uncertainties for in-core components such as fuel cladding. In addition, there are insufficient modelling tools to analyse the varying test data and extrapolate the results to very long times, and only basic understanding of the triggers and kinetics of corrosion processes with long incubation periods, such as breakaway oxidation at high temperatures.

In addition, nuclear grade materials manufacture, fabrication and qualification are gradually becoming high priority issues as the HLM technology moves out laboratory and into test and demonstration facilities.

In the case of ADS systems developed in Europe, reference structural materials have been selected. These are the T91 martensitic steel for the highly loaded parts (cladding, wrapper, spallation target structure) and the AISI 316L austenitic steel for the vessel and in-vessel components. In addition it has been agreed to characterise Fe-Al based coatings, which are envisaged as an alternative protection method to the oxide layers grow, for the fuel claddings.

For design concepts in other countries, similar materials are selected. For example, the reference materials for LFRs in the US are HT-9 (T91 as a slightly more developmental backup) and 316L. The Russians developed some special alloys with added Si content for HLM cooled reactors, most notably EP823.

For these materials the basic data on compatibility and mechanical property changes in the liquid metals are already available, mostly under out-of-pile conditions. However, for the specific design requirements the following data are needed with high priority:

- long-term corrosion behaviour of the steels and the coatings;
- corrosion tests in LBE with low oxygen concentration in order to verify the low limit of the recommended condition for oxygen control;
- studies on corrosion erosion and friction mechanism;
- development of mass transfer model using reliable solubility and diffusivity data and corrosion data also for non isothermal systems;
- mechanical behaviour of the structural materials and the corrosion protection barriers in a representative temperature-stress field and more specifically:
  - creep;
  - fatigue and creep-fatigue;
  - fracture mechanics;
  - creep and fatigue crack growth.

In addition a very high priority is placed on the assessment of mechanical properties in the liquid metal and under irradiation of the steels and coatings.

These properties need to be measured in the relevant ranges of temperature, neutron fluence, stresses and HLM flow velocity for the different components. At the present the European design group for the ADS system has yet to produce a complete set of specifications to identify the testing ranges. Nevertheless, preliminary designs have allowed the definition of two major irradiation experiments in LBE in the HFR and BR2 reactors. These irradiation experiments will be performed in the temperature range of 350-550°C up to 4.5 dpa. Parallel irradiation experiments on T91 and Fe-Al based coatings in a fast spectrum (Phénix) have also been foreseen. In this case the irradiation temperature is 400-530°C and a dose of about 70 dpa may be reached.

Finally, the need to define standard procedures to perform corrosion and mechanical tests has been clearly expressed. At the European level, the laboratories will work towards the definition of such standard procedures.

After the completion of this materials testing programme, which performs a general assessment of the austenitic and ferritic/martensitic steels in LBE and under irradiation, the next step will be the performance assessment of manufactured materials in well defined shapes, e.g. tubing as fuel cladding.

14.2.4 Technologies

While much progress has been made in understanding the effect of coolant chemistry on materials corrosion, and measurement and control of oxygen in HLM in laboratory and small to medium test facilities, there are significant needs in scaling up to large systems, in particular pool-type and/or natural circulation systems. Oxygen sensors for higher temperatures, deep immersion in liquid metals, and enhanced robustness and reliability in plant-like environment requires continued improvement from the state of art. Oxygen control systems, filtration methods, and oxygen activity maintenance and restoration after drainage, cool-down, freeze and thaw, will need further development and testing. Alternative and/or expanded control ranges, coupled with materials development, are desirable. Most of these are technological issues, and their priorities range from high to medium, depending on specific concepts and programme development paths and time horizons.

In the EUROTRANS the *priorities* have been envisaged for the development of:

- HLM purification systems for the control of aerosols and slags, to be applied in large scale facilities.
- Reliable instrumentation (flow meters, pressure transducers, thermocouples, level sensors, pumps etc.) for the long-term operation of HLM facilities.
- Reliable oxygen control methods and monitoring systems. Different types of methods have been envisaged to set the oxygen content in the liquid metal, these methods are based on liquid/gas or liquid/solid exchange. The efficiency of these systems needs to be assessed in order to make a selection of the most reliable and easiest method to be implemented in a nuclear system.
- Optimisation of on-line oxygen sensors in order to raise their reliability to nuclear standards. Effort is needed in the definition of a calibration strategy and to enhance their long term performance and thermal shock resistance.
- Instrumentation for the in-service inspection and repair (ISIR). These instruments need to be tested and calibrated in a combined LBE and irradiation environment.

14.2.5 Thermal-hydraulics

There are two kinds of open issues in this area. The first is related to the fundamental nature of heavy liquid metals. There are some needs in developing and validating more suitable turbulent model(s) for computational thermal hydraulics, especially for complex geometries and critical components, e.g. high-power spallation target windows in ADS and core configuration.

The second type of issues are technological and mostly related to the nature of the HLM-cooled system design and operation. Using coolant chemistry control and surface protective oxide formation to mitigate steel corrosion have consequences in heat transfer performance, particular for long-term or in abnormal situations, such as build-up of oxides and high level of solid oxide particles. HLM-cooled nuclear reactors usually have open lattice configurations to reduce pumping power needs and enhance passive safety. Flow circulation methods, transients, flow stability and elimination of undesired instability are all important issues to be investigated.

In the frame of the EUROTRANS project several experimental activities have been defined to support the design of the spallation target and the subcritical core.

According to the present design choices, *the priority* has been put on a free surface experiment in water and LBE to support the windowless target design. This activity will be strongly supported by CFD calculations. The CFD group will in turn use the results from the experiments to improve the related physical models, and to evaluate and benchmark the CFD codes.

In the thermal-hydraulics studies related to flow characterisation in the core region of ADS, two experiments have been defined according to design requirements:

- A single fuel pin experiment to evaluate the heat transfer in turbulent conditions across a prototypic cladding under the prototypical ADS conditions. This experiment will be conducted in the TALL facility at KTH, Sweden.
- A fuel rod bundle experiment to analyse the thermal-hydraulics behaviour of fuel assemblies. The plan is to specifically instrument the test section for local measurement of temperature, velocity and integral flow rate. The fuel bundle experiment will be conducted at the THEADES facility of KALLA at FZK, Germany.

At the present, no experiments have been yet been planned to support the safety analysis of severe accident conditions. These experiments need to be planned as soon as the design definition becomes more mature.

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