

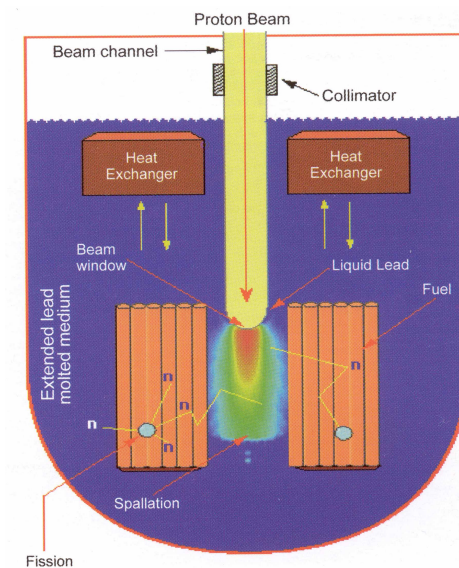
Chapter 1

INTRODUCTION*

Liquid metals have been studied since the early development of fission energy as reactor core coolants for fast reactors, fusion energy blanket applications and, more recently, for accelerator-driven systems (ADS) proposed for high-level radioactive waste transmutation. Moreover, heavy liquid metals are being proposed as target materials for high power neutron spallation sources.

Accelerator-driven systems (ADS) are nuclear fission reactors with a subcritical core, i.e. $k_{\text{eff}} < 1$. Therefore to operate ADS an external neutron source is needed for a stationary behaviour of the core. A possible external neutron source is provided by a proton accelerator and a spallation target (a heavy liquid metal is often considered). The protons hitting the heavy liquid metal generate neutrons which sustain the chain reaction in the sub-critical core. In Figure 1.1 a schematic view of an accelerator-driven system is provided.

Figure 1.1. Schematic diagram of an ADS [A European Roadmap]

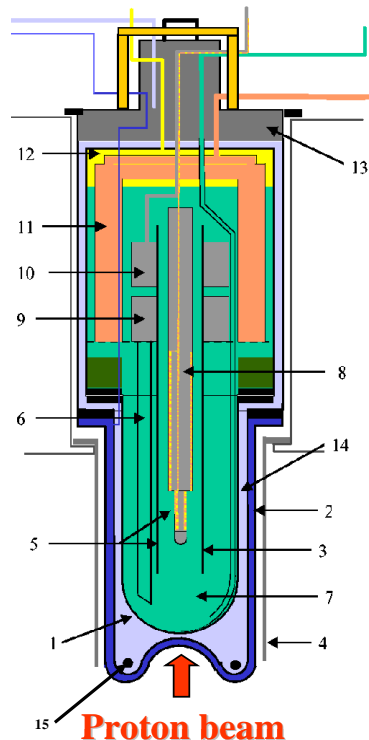


Neutron spallation targets are also being developed to provide a neutron source for other applications. For example, the MEGAPIE spallation neutron target (a schematic view of the MEGAPIE target is shown in Figure 1.2), which will be tested at the SINQ facility of the Paul Scherrer Institut in Switzerland, has been designed and constructed in the frame of ADS development. Its objective is to demonstrate the operability of such a liquid metal target while providing a neutron source for the typical applications at SINQ, i.e. material investigation with neutrons.

* Chapter lead: C. Fazio. For additional contributors, please see the List of Contributors included at the end of this work.

Figure 1.2. Schematic diagram of the MEGAPIE target
[Proceedings of the 4th MEGAPIE Technical Review Meeting]

1 – T91 window, 2 – lower target enclosure (AlMg₃), 3 – main flow guide tube, 4 – moderator, 5 – heater, 6 – bypass flow guide tube, 7 – LBE, 8 – central rod, 9 – bypass pump, 10 – main pump, 11 – heat exchanger, 12 – expansion volume, 13 – shielding, 14 – insulation gas (Ar), 15 – LBE leak detector

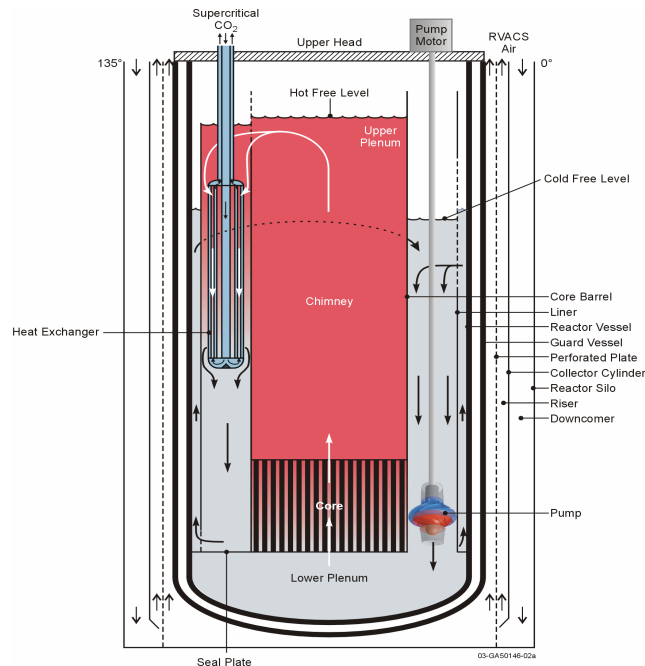


Fast reactors are fission reactors where the neutron spectrum in the core is close to the fission neutron spectrum, since the neutrons are not thermalised as in a conventional light-water-cooled reactor. The fast reactor coolant is appropriately chosen in order to provide an effective heat transfer, without a significant thermalisation of the neutron spectrum. In order to achieve this goal, liquid metals (Na or Pb,Pb/Bi) or gas can be (or have been) used. In Figure 1.3 a schematic view of a Pb-cooled fast reactor is given.

Heavy liquid metals (HLM) such as lead (Pb) or lead-bismuth eutectic (LBE) were proposed and investigated as coolants for fast reactors as early as in the 1950s (e.g. in the USA). Sodium became the preferred choice in the sixties, due to a higher power density achievable with this coolant, which resulted in lower doubling times, an important objective at that time [IAEA TECDOC 1289]. However, LBE was chosen as the coolant for a number of alpha class submarine reactors in the former Soviet Union, which led to very extensive research and development of the coolant technology and materials, with particular emphasis on the chemistry control of the liquid metal to avoid plugging due to slag formation and to enhance corrosion resistance of the steels specifically developed for such services.

More recently, there has been renewed interest in Russia in lead and LBE coolants for civilian fast reactors [Kirillov, 1998, 2000, 2003]. The lead-cooled BREST (Russian acronym for Pb-cooled fast reactor) [Filin, 2000] concept developed since the early 1990s is the most widely known, with the LBE-cooled SVBR (Russian acronym for lead-bismuth fast reactor) concept [Stepanov, 1998] competing for attention. Their features and the associated technologies inspired several projects in the

Figure 1.3. Schematic of Pb-cooled fast reactor [Hejzlar, 2004]



emerging field of ADS, and in particular lead cooling was associated, in the mid-1990s, with the proposal for an energy amplifier project together with LBE as a spallation target coolant and material. Subsequent development of ADS in the USA, Europe, Japan and the Republic of Korea has adopted a heavy liquid metal (most often LBE) as the coolant for the subcritical core and as coolant and material for the spallation target which provides the external neutron source.

At the Korea Atomic Energy Research Institute (KAERI) and Seoul National University (SNU) in the Republic of Korea, both ADS and LFR systems are under the development in order to explore proliferation-resistant and safe transmutation technology. KAERI has been developing ADS since 1997. KAERI's ADS, the Hybrid Power Extraction Reactor (HYPER) is designed to transmute TRU and some fission products such as ^{129}I and ^{99}Tc . HYPER uses Pb-Bi as both the coolant and target material. At SNU, a Pb-Bi-cooled transmutation reactor, the Proliferation-resistant, Environment-friendly, Accident-tolerant, Continual and Economical Reactor (PEACER) has been developed since 1998.

At SCK•CEN, Belgium, since 1997 studies in the field of lead-bismuth eutectic (LBE) technology have been related to the Multi-purpose Hybrid Research Reactor for High-tech Applications (MYRRHA) project and are aimed at the development of a research reactor driven by an accelerator, where LBE is used as spallation target and coolant.

In Japan, both ADS and LFR systems using LBE are under the development. At the Japan Atomic Energy Research Institute (JAERI)¹ an ADS with the thermal power of 800 MW has been designed, where 250 kg of minor actinides and some long-lived fission products (LLFP) can be transmuted annually. R&D has been conducted on ADS using LBE as a spallation target and a coolant, and research using J-PARC is also planned. The LFR systems using LBE as a coolant have been studied both at Tokyo Institute of Technology (TIT) and the Japan Nuclear Cycle Development Institute (JNC)¹ separately. One of the LFR systems studied at TIT is designated as the Pb-Bi-cooled Direct Contact Water Fast Reactor (PBWFR).

¹ Now JAEA (Japan Atomic Energy Agency).

In summary, at present a number of experimental programmes are ongoing world-wide for the transmutation of nuclear waste and the development of HLM cooled fast reactors. These include:

- The USA Advanced Fuel Cycle Initiative [Report to Congress, 2003];
- The European Commission four-year (04/2005-04/2009) Integrated Project EUROpean Research Programme for the TRANSmutation of High Level Nuclear Waste in an Accelerator Driven System, IP-EUROTRANS [Integrated Project, 2004], [Knebel, 2005]. In addition in Europe there are several programmes ongoing at national level, as for instance in France the GEDEON, now GEDEPEON (*Gestion de Déchets Radioactives par des Options Nouvelle*) programme, and the MYRRHA project at SCK•CEN in Belgium. MYRRHA is being developed as a multi-purpose neutron source for R&D applications on the basis of an ADS [Abderrahim, 2001, 2005a, 2005b].
- The South Korean programmes of HYPER (ADS) and PEACER (reactor) [Park, 1996], [Hwang, 2000], [Song, 2004].
- The Japanese programme in the framework of ADS development and LFR development [Mukaiyama, 1999], [Oigawa, 2004], [Sasa, 2004], [Takahashi, 2004].
- The Russian programme for the BREST [Filin, 2000] and SVBR [Stepanov, 1998] reactors.

Finally, in the framework of the Generation-IV Nuclear Energy Systems initiative, a class of Pb/LBE-cooled fast reactors (LFRs) has been chosen as one of six system concepts for further development. A host of new missions have been proposed for LFRs made possible by the properties of Pb/LBE, including hydrogen production, nuclear waste transmutation, and small modular reactors with long-life cores for supplying electricity and heat in remote areas and/or developing economies. In this context a multiyear project at the Idaho National Laboratory and the Massachusetts Institute of Technology investigated medium power lead alloy cooled systems with the aim of producing low cost energy and, at the same time, burning actinides [Todreas, 2004].

In the area of the fusion technology programme the eutectic alloy Pb-17Li is largely studied as breeder and as coolant. A wide range of activities have been conducted in order to characterise materials and develop appropriate technologies [Kleykamp, 2002].

The selection criteria for the use of liquid metals as heat-transfer media in a nuclear environment include the following:

- Neutronics, related to the fast spectrum necessary for breeding, fuel conversion and actinide transmutation in the next generation fast reactors and ADS concepts. In this case the coolant should have:
 - small (fast) capture cross-section (for small parasitic loss of neutrons);
 - high scattering cross-section (for small leakage of neutrons from the core);
 - small energy loss per collision (for small spectrum softening (moderating) effect);
 - high boiling temperature (for prevention of reactivity effects from boiling related coolant voiding).
- Materials:
 - acceptable corrosion and mechanical degradation of structural and containment materials, and lifetime of equipment;
 - high stability of the liquid metal (e.g. limited chemical reactions with secondary coolants and air or formation of spallation products, etc.).

- Thermal-hydraulics:
 - moderate power requirement for circulating the liquid metal;
 - high heat transfer coefficient and small size of heat exchanger.
- Safety:
 - controllable chemical and radioactive hazards;
 - simple and reliable safety measures and systems.
- Economics.

Based on these factors and on the inspection of Table 1.1, it can be concluded that heavy liquid metals are well suited for fast reactor cores (see for example [Todreas, 2004]). Indeed, the use of heavy liquid metals (e.g. Pb/LBE) allow the achievement of a harder neutron spectrum, which results in better neutron economy (essential e.g. for burning actinides). Some other favourable features of using LBE in nuclear systems are based on its high boiling temperature and low melting temperature. The high boiling temperature is an important safety feature, essentially eliminating the pressurisation and boiling concerns while enhancing the inherent safety of reactor cores. Higher allowable operating temperatures also improve efficiency and feasibility of other energy products. The relatively low melting point eases use at low temperatures with reduced risk of uncontrolled freezing. High density and wider range of possible operating temperature offer increased design space for passive safety. A comprehensive comparative assessment of thermo-physical and thermo-hydraulics characteristics of lead, lead-bismuth eutectic alloy and sodium is also given in the IAEA TECDOC 1289.

Table 1.1. Basic characteristics of reactor coolants

Table taken from [Todreas, 2004]

Coolant	Atomic mass (g/mol)	Relative moderating power	Neutron absorption cross-section (1 MeV) (mbarn)	Neutron scattering cross-sections (barn)	Melting point (°C)	Boiling point (°C)	Chemical reactivity (with air and water)
Pb	207	1	6.001	6.4	327	1737	Inert
LBE	208	0.82	1.492	6.9	125	1670	Inert
Na	23	1.80	0.230	3.2	98	883	Highly reactive
H ₂ O	18	421	0.1056	3.5	0	100	Inert
D ₂ O	20	49	0.0002115	2.6	0	100	Inert
He	2	0.27	0.007953	3.7	–	-269	Inert

Other potentially favourable features of HLM are: lower reactivity associated with hypothetical voiding of the coolant; better shielding against gamma rays and energetic neutrons; high solubility of the actinides in the coolant, which could help to minimise the potential for re-criticality events upon core melting, and no energetic reaction with air and water, thereby eliminating the possibility of fires. One drawback associated with the use of liquid metal coolants, is the potential complexity of in-service inspection and repair.

With respect to spallation neutron sources, there is a general consensus that above 1 MW of beam power, solid targets are hardly feasible from a heat removal point of view. Therefore, liquid metals targets are the best choice (see e.g. [Bauer, 2001]), among the liquid metals lead-alloy-based liquid metal targets are to be preferred if high operating temperatures are required. Properties that make heavy liquid metals ideal as spallation materials for neutron sources are listed in Table 1.2.

Table 1.2. Some relevant properties of possible liquid metal target candidate materials*Table taken from [Bauer, 2001]*

Coolant	Composition (at.%)	Density at 20°C (g/cm ³)	Density liquid (g/cm ³)	Linear coefficient of thermal expansion 10 ⁻⁵ /K (solid)	Linear coefficient of thermal expansion 10 ⁻⁵ /K (400°C)	Volume change upon solidification (%)	Specific heat (J/gK)	Thermal neutron absorption (barn)
Pb	Elem.	11.35	10.7	2.91	4	3.32	0.14	0.17
Bi	Elem.	9.75	10.07	1.75	–	-3.35	0.15	0.004
Pb-Mg eutectic	Pb 97.5% Mg 2.5%		10.6	–	–	0	0.15	0.17
Pb-Bi eutectic	Pb 45% Bi 55%	10.5	10.5			0	0.15	0.11
Hg	Elem.		13.55		6.1		0.12	389

The emerging worldwide interests in the applications of HLM coolants have led to many R&D activities in the fields of materials, thermal-hydraulics, physical chemistry, etc. It is becoming increasingly clear and urgent that a HLM handbook is needed for designers of HLM systems and for researchers in this field. Such a handbook should be a comprehensive compilation of all relevant properties, material test results, primary monitoring and control techniques, and instrumentation. Just as important, it should discuss the state of the art in research methodology and R&D resources (test facilities), and suggest a commonly accepted reporting and analysis protocol for systematic advancement of the scientific understanding and technological applications of HLM.

Several liquid metal handbooks dating back to the 1950s with data available at that time have been issued. However, this data was limited due to restrictions associated with strategic national programmes. Although it has been reported that the Russians had a manual or database for designers, this is not publicly available. The US Advanced Accelerator Application (AAA) programme included in its materials handbook a brief chapter on this topic. However, none of these can fulfil the demanding needs of today's vibrant and diverse international research community.

In this context, the OECD Nuclear Energy Agency (NEA), in the framework of the former Working Party on Partitioning and Transmutation (WPPT), now Working Party on Fuel Cycle (WPFC), launched the HLMC handbook project. The original scope to cover the relatively more mature LBE coolant technology and materials has been expanded to include Pb for higher temperature and high-performance next-generation nuclear systems. The higher availability of basic property data for Pb can serve as a reference, and in some cases, serve as proxy for relatively scarce LBE property data. Conversely, the higher availability of LBE test data and facilities can benefit R&D for Pb. It is also envisioned that this handbook will be an evolving and working document of the continued R&D efforts around the world in the next several years, with increasing utility for designers.

The structure of this handbook is as follows: four chapters are dedicated to HLM properties; the next four chapters cover the materials and testing issues; and the subsequent two chapters summarise the key aspects of the thermal-hydraulics and system technologies. In the last three chapters, other issues such as existing test facilities, safety guidelines and open issues and perspectives are presented.

HLM properties are reported in Chapters 2-5. Chapter 2 compiles the thermo-physical and electrical properties of the LBE and Pb (e.g. density, molar volume, isobaric heat capacity, viscosity, thermal and electrical conductivity, etc.) reported in the open literature. In some cases, significant discrepancies exist among the different sources, and recommendations based on the “best fit” of data are offered.

Chapter 3 addresses the thermodynamic relations, transport properties and chemistry of HLM, such as the solubility and diffusivity of oxygen and metallic elements in the liquid metal. In Chapter 4 the chemistry control and monitoring systems are reported. The main chemistry issue is the monitoring and adjustment of the oxygen level in HLMS for the mitigation of corrosion and coolant contamination problems. For this purpose the development, calibration and performance of electrochemical oxygen sensors and oxygen control systems are extensively described.

Chapter 5 deals with the properties of irradiated LBE and Pb. For this topic, very little data is available and most of them have been produced in the framework of the international MEGAPIE initiative and CERN experiments.

Materials issues are covered in the Chapters 6-9. The compatibility of ferritic/martensitic and austenitic steels with the liquid metals are given in terms of corrosion (Chapter 6) and effects on the mechanical properties in stagnant and flowing liquid metals (Chapter 7). While substantial amount of corrosion test results are available from many sources, most results pertain to relatively short durations (up to a few thousand hours). Although several key qualitative conclusions can be drawn, the wide ranging test conditions and materials render it very difficult at the present to derive a consistent set of correlations for design use, especially in long-term applications. It is also noted that the data on the mechanical property changes is fairly scarce.

In Chapter 8 a collection of data is given representing the combined effect of proton irradiation and HLM on the properties of structural materials. These data have been produced principally at the Paul Scherrer Institute (Switzerland) and in the framework of the MEGAPIE initiative, with contribution from Los Alamos National Laboratory (USA). More data on irradiation effects on compatibility of structural materials with Pb and LBE in the neutron field will be available at the end of the next five-year period, after the completion of the experiments described in this chapter.

Chapter 9 is dedicated to corrosion-protection methods. In particular, two types of methods are under development and testing – the *in situ* growth and control of a “self-healing” protective oxide layer on the steel surface, and the deposition of a Fe/Al-based surface coating. Other types of coatings, such as in-situ formation of carbides and nitrides via addition of inhibitors, have been tested but not as extensively as the previous one mentioned.

Chapters 10 and 11 address the thermal-hydraulics behaviour and instrumentations needed for scientific, technological and operational purposes. As far as the thermal-hydraulics quantities, it has been seen that the available set of data is still not sufficient for a complete validation of computational fluid dynamics (CFD) codes and for development of reliable and realistic physical models.

A compilation of the existing OECD experimental facilities with their main parameters and key objectives is given in Chapter 12. Chapter 13 briefly reviews the effects of HLM containing Pb and Bi on human and environmental health and safety, and outlines the safety guidelines for the use of HLMS. Finally in Chapter 14 the open issues and the strategic outlook for R&D are summarised.

REFERENCES

- A European Roadmap for Developing Accelerator Driven Systems (ADS) for Nuclear Incineration* (2001), The European Technical Working Group, ISBN 88-8286-008-6, April 2001.
- Abderrahim, H. Aït, *et al.* (2001), “MYRRHA: A Multipurpose Accelerator Driven System for Research & Development”, *Nuclear Instruments & Methods in Physics Research*, A 463, 487-494.
- Abderrahim, H. Aït (2005a), “MYRRHA: A Multipurpose ADS for R&D, Progress Report at End 2004”, *ICONE 13*, Beijing.
- Abderrahim, H. Aït, D. de Bruin (2005b), *MYRRHA, a Future for Nuclear Research. Pre-design (DRAFT-2) Report*, H. Aït Abderrahim, D. de Bruin (Eds.), SCK•CEN report, Mol, Belgium.
- Bauer, G.S. (2001), “Physics and Technology of Spallation Neutron Sources”, *Nuclear Instruments and Methods in Physics Research*, A 463, 505-543.
- Filin, A.I., *et al.* (2000), *Design Features of BREST Reactors and Experimental Work to Advance the Concept of BREST Reactors*, IAEA-TECDOC 1348, pp. 36-47, 23-27 October 2000.
- Handbook on Thermo-physical Properties of Materials for Nuclear Engineering* (2005), pp. 180 (in Russian).
- Hejzlar Pavel, Philip E. Macdonald, Neil E. Todreas (2004), “Design Strategy and Constraints for Medium-power Lead-alloy-cooled Actinide Burners”, *Nuclear Technology*, Vol. 147, Sept. 2004, pp. 321-343.
- Hwang, I.S., S.H. Jeong, B.G. Park, W.S. Yang, K.Y. Suh, C.H. Kim (2000), “The Concept of Proliferation-resistant, Environment-friendly, Accident-tolerant, Continual and Economical Reactor (PEACER)”, *Progress in Nuclear Energy*, Vol. 37, No. 1-4, pp. 217-222.
- IAEA TECDOC 1289 (2002), *Comparative Assessment of Thermophysical and Thermohydraulic Characteristics of Lead, Lead-bismuth and Sodium Coolants for Fast Reactors*, June 2002.
- Integrated Project EUROpean Research Programme for the TRANsmutation of High Level Nuclear Waste in an Accelerator Driven System, EUROTRANS* (2004), EC project No. FI6W-CT-2004-516520.
- Kirillov, P.L. (1998), *Thermophysical Properties of Lead, Bismuth and Their Eutectic Alloy*, FEI-0286, pp. 28 (in Russian).
- Kirillov, P.L., N. Deniskina (2000), *Thermophysical Properties of Liquid Metal Coolants*, the tables and correlations, FEI-0291, pp. 41 (in Russian).
- Kirillov, P.L. (Ed.) (2003), *Knowledge Preservation Initiative – Generalization of Data on Hydrodynamics and Heat/Mass Transfer Processes in Liquid Metal Cooled Reactors*, prepared at the IPPE, pp. 660, 200 tables, 400 figures, more than 1000 references (in Russian and English).

Kleykamp, H., J. Linke, G.E. Lucas, B.N. Singh (2002), *Proceedings of the 10th International Conference on Fusion Reactor Materials (ICFRM-10)*, *Journal of Nuclear Materials*, Vols. 307-311.

Knebel, J.U., *et al.* (2005), “Overview on Integrated Project EUROTRANS: EUROpean Research Programme for the TRANSmutation of High Level Nuclear Waste in an Accelerator Driven System”, *3rd Annual Idaho ADSS Experiments Workshop*, Pocatello, ID, 1-2 June 2005.

Mukaiyama, T. (1999), *OMEGA Programme in Japan and ADS Development at JAERI*, IAEA TECDOC--1365, pp. 153-165.

Oigawa, H., *et al.* (2004), “R&D Activities on Accelerator-driven Transmutation System in JAERI”, *OECD/NEA 8th Information Exchange Meeting on Actinide and Fission Product Partitioning & Transmutation*, Las Vegas, Nevada, USA, 9-11 November 2004.

Park, W.S., *et al.* (1996), *Development of Nuclear Transmutation Technology*, KAERI/RR-1702/96; *Proceedings of International Conference on Heavy Liquid Metal Coolants in Nuclear Engineering*, Vol. 1, 2, p. 824, 5-9 Oct. 1998, Obninsk, IPPE.

Proceedings of the 4th MEGAPIE Technical Review Meeting (2003), C. Fazio, J.U. Knebel, F. Gröschel (Eds.), Paris, France, 18-19 March 2003, Report Forschungszentrum Karlsruhe, FZKA 6876, December 2003.

Report to Congress on Advanced Fuel Cycle Initiative: The Future Path for Advanced Spent Fuel Treatment and Transmutation Research (2003), US DOE, Office of Nuclear Energy, Science and Technology.

Sasa, T., *et al.* (2004), “Research and Development on Accelerator-driven Transmutation System at JAERI”, *Nuclear Engineering and Design*, 230, pp. 209-222.

Song, T.Y., *et al.* (2004), “HYPER Project”, *Proceedings of OECD/NEA 4th International Workshop on Utilisation and Reliability of High Power Proton Accelerators*, Daejeon, Korea, 16-19 May 2004.

Stepanov, V.S., *et al.* (1998), *SVBR-75: A Reactor Module for Renewal of WWER-440 Decommissioning Reactors – Safety and Economic Aspects*, IAEA TECDOC 1056, pp. 165-176, November 1998.

Takahashi, M., *et al.* (2004), “Design and Experimental Study for Development of Pb-Bi Cooled Direct Contact Boiling Water Small Fast Reactor (PBWFR)”, *Proc. of ICAPP'04*, Pittsburgh, PA, USA, 13-17 June 2004, paper 4058.

Todreas, N.E., P.E. MacDonald, J. Buongiorno, E.P. Loewen (2004), “Medium-power Lead Alloy Reactors: Missions for this Reactor Technology”, *Nuclear Technology*, Vol. 147, No. 3, 305 (Sept. 2004).