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FOREWORD

All prototype, demonstration and commercial liquid metal cooled fast reactors (LMFRs) have used liquid sodium as a coolant. Sodium cooled systems, operating at low pressure, are characterised by very large thermal margins relative to the coolant boiling temperature and a very low structural material corrosion rate. In spite of the negligible thermal energy stored in the liquid sodium available for release in case of leakage, there is some safety concern because of its chemical reactivity with respect to air and water. Lead, lead-bismuth or other alloys of lead, appear to eliminate these concerns because the chemical reactivity of these coolants with respect to air and water is very low. Some experts believe that conceptually, these systems could be attractive if high corrosion activity inherent in lead, long term materials compatibility and other problems will be resolved. Extensive research and development work is required to meet this goal.

Preliminary studies on lead-bismuth and lead cooled reactors and ADS (accelerator driven systems) have been initiated in France, Japan, the United States of America, Italy, and other countries. Considerable experience has been gained in the Russian Fedaration in the course of development and operation of reactors cooled with lead-bismuth eutectic, in particular, propulsion reactors. Studies on lead cooled fast reactors are also under way in this country.

The need to exchange information on alternative fast reactor coolants was a major consideration in the recommendation by the Technical Working Group on Fast Reactors (TWGFRs) to collect, review and document the information on lead and lead-bismuth alloy coolants: technology, thermohydraulics, physical and chemical properties, as well as to make an assessment and comparison with respective sodium characteristics. This report considering these issues has been prepared in response to the recommendation from TWGFR.

The report has been reviewed by G. Heusener (Forschungszentrum Karlsruhe, Germany), Y. Nishi (Central Research Institute of Electric Power Industry, Japan), and D. Wade (Argonne National Laboratory, USA). A. Rineiskii and A. Stanculescu of the IAEA’s Division of Nuclear Power were the IAEA officers responsible for the publication.

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1. INTRODUCTION

In the current structure of nuclear power, light water reactors (LWRs) are predominant over a small number of heavy water reactors (HWRs), and even smaller number of fast breeder reactors (FBRs). However, an increase of FBR share can be predicted for the future, taking into account their unique properties. First of all, there is the capability of nuclear fuel breeding by involving ^{238}U into the fuel cycle. Secondly, there is the fast reactor's flexibility permitting its use as plutonium incinerators and minor actinides transmutation. Thus, unless new sources of energy are found, the development of nuclear power will be necessarily based on fast breeder reactors.

For the next few decades, saving of uranium resources by nuclear fuel breeding will not be of primary importance for countries with a significant nuclear power sector. This is caused by a number of factors, namely: (a) accumulation of plutonium as a result of reprocessing of spent fuel of the operating NPPs; (b) release of considerable amount of plutonium and enriched uranium owing to disarmament; (c) decrease of uranium consumption in the military industry; (d) slowing down of the rate of nuclear power development; and (e) ready availability of fossil fuels (natural gas and oil).

The development of fast reactors has been delayed in countries characterised by advanced market economics, because of relatively slow primary energy consumption growth and availability of fossil fuel resources. On the other hand, in countries with more rapid energy consumption growth and limited indigenous uranium ore resources, fast reactor development is continuing. In due course, when it is economically necessary, fast reactors will almost certainly make the major contribution to the world's energy supplies. There will undoubtedly be further improvement of the technology to achieve the highest standard of safety, non-proliferation, environmental protection and economics.

By now, several developed and advanced developing countries have passed the stage of demonstrating technical feasibility, safety and reliability of FBRs. Starting from the mid 1970s, advanced FBR designs have been developed in a number of countries, these designs meeting the most strict safety requirements and assuring their competitiveness with LWRs in terms of electricity generation cost. These are primarily the demonstration European fast reactor EFR, the commercial fast reactor BN-800 in the Russian Federation, the demonstration fast breeder reactor DFBR in Japan and the prototype fast breeder reactor PFBR in India (rated power of 1500 MW(e), 800 MW(e), 600 MW(e) and 500 MW(e), respectively).

There is a widespread opinion that the existing water, gas and liquid metal cooled reactor systems can meet present safety standards quite adequately, and are expected to do so for the foreseeable future. Nevertheless it is essential to continue the search for greater safety because: (1) no opportunity to improve technology should be ignored; (2) safety standards may be raised by the regulatory authorities; (3) nuclear power plants suitable for countries or regions with less developed infrastructure will be needed; and (4) more cost-effective ways of meeting existing safety standards may be found.

Now, because of the delay in the fast reactors commercial introduction, there is an opportunity to investigate alternative technical solutions. The objective is to obtain complete knowledge of their characteristics allowing to make, when the time has come, the best choice for the reactor design for concrete realization. Reactor designs using gas, steam, lead and lead-bismuth alloy coolants are considered as possible alternatives.

The appropriate choice of the primary coolant is of great significance for achieving high FBR performances. This determines also the main design approaches of FBR and, to a great extent, the technical and economical characteristics of the nuclear power plant.

Among all liquid metal coolants, it is sodium that has gained the widest acceptance. This coolant has been chosen for liquid metal cooled fast reactor (LMFR) because of its good thermal and physical properties. Sodium offers the possibility to achieve high specific power densities in the core and thus for a short doubling time. This was the world-wide strategic line of fast reactor development in the 1960s, and is continued in some countries presently. For this very reason, when considering various liquid metal coolants for FBR in the 50-ies, sodium was given preference, although initially, in the former USSR, lead-bismuth coolant was also considered.

Large experience has been already gained with sodium cooled fast reactor operation. The use of sodium as a coolant poses fire danger in case of its leakage and interaction with air or water. Operating experience testifies the possibility of coping with the mentioned problem, but the quest for excellence calls for future improvement in LMFRs technology.

In the earlier phases of breeder reactor development, especially in the 1950s and 1960s, high pressure gases, such as helium,CO_2 or superheated steam were studied. Between 1960 and 1970, H_2 O-steam cooled and D_2 O-steam cooled fast reactor concepts were studied in the USA and the former FRG. Helium cooled fast reactor concepts have been pursued as an alternative coolant concept in Europe and the USA. Some fuel development for a CO_2 cooled fast breeder has been continued on a small scale in the UK. Lead-bismuth alloy as a coolant was studied in the former USSR for propulsion and land based reactors.

However, the choice of liquid sodium as a coolant and principal design features of fast reactors were mainly determined in the 1960s, as already mentioned, by the requirement of high power densities in the reactor core (about 500 kW(th)/l for MOX fuel), and the need of a weakly moderating material with good heat transfer properties. The important fact was also that sodium is practically non-corrosive to stainless steal.

Presently, the availability of great amounts of fissile plutonium produced by thermal reactors and that released from now-redundant nuclear weapons in the USA, the Russian Federation, the European Union, and in some other countries with significant nuclear power sector and relatively slow primary energy consumption growth, has eliminated the initial requirements of short doubling time and high breeding ratio.

The changing of the strategic line, delaying of fast reactors commercial introduction and some drawbacks of sodium technology related in some demonstration LMFRs have given the rise to the question to reverse the trend of the last 20 years, which focused fast reactor R&D on one option — sodium cooled fast reactors. Some experts believe that a more generic exploratory research on the different options, including “revisited” ones, should be open again as in the early stage of fast reactor development.

Not only new innovative ideas as, for example, lead or lead-bismuth cooled fast reactors are being studied in Member States now, but almost all old ones mentioned above that have also a considerable innovative character: this is the cases of gas-cooled high temperature and superheated supercritical steam-cooled fast reactors. This is due to an increase of the gas turbines thermal efficiency from 35% to 50% within the last 10 years and to work on supercritical water/steam high performance LWR with thermal efficiency of 44% and high power density.

Considerable experience has been gained in the Russian Federation with lead-bismuth (PbBi) eutectic alloy application as reactor coolant. Since Bi is sufficiently rare and expensive metal, and also it is a source of volatile α-active ^{210}Po, the proposal to use lead as a coolant in power fast reactors is now under consideration in several countries. Lead based alloys are currently being considered for hybrid systems (accelerator driven fast reactors) in which the coolant could double as the spallation source for driving the core.

Techniques to counter the heavy metal coolant disadvantages are being developed, but in spite of this work and the apparent disadvantages of sodium, the consensus in favour of sodium remains strong. This is demonstrated by fact that "before lead-cooled fast reactor BREST-300 is built, MINATOM will first build a sodium-cooled LMFR BN–800"(E. Adamov, NW, 23 September 1999). Moreover, in the last few years sodium has been chosen in both China and the Republic of Korea for the respective fast reactor development project. This is a significant endorsement for sodium as a fast reactor coolant.

However, given that there is now this tendency for countries to have their own viewpoint and their own preferred option: evolutionary sodium-cooled LMFR models or an innovative one with new coolant (gas, steam, other than sodium liquid metal coolants), it is essential to clarify as far as possible the scientific issues related to the different innovative options and to exchange information on advances in development of traditional and innovative fast reactors.

That is why this report is devoted to the comparative assessment of general characteristics of a standard fast reactor coolant (sodium) and innovative ones, such as lead and lead-bismuth alloy.

2. TECHNICAL BACKGROUND

2.1. SODIUM- COOLED FAST REACTORS

LMFRs(liquid metal cooled reactor) have been under development for more than 50 years. Twenty LMFRs have been constructed and operated. Five prototype and demonstration LMFRs (BN-350/Kazakstan, Phenix/France, Prototype Fast Reactor/UK, BN-600/Russian Federation, Super Phenix/France) with electrical output ranging from 250 to 1200 MW(e) and large scale (400 MW(th)) experimental fast flux test reactor FFTF/USA have gained nearly 110 reactor-years. In total, LMFRs have gained nearly 310 reactor-years of operation.

In many cases the overall experience with fast reactors has been extremely good, the reactors themselves and, more frequently, particular components, showing good performance well in excess of design expectations. They also have been shown to have very attractive safety characteristics, resulting to a large extent from being low pressure system with large thermal inertia and negative power and temperature reactivity coefficients [2.1, 2.2].

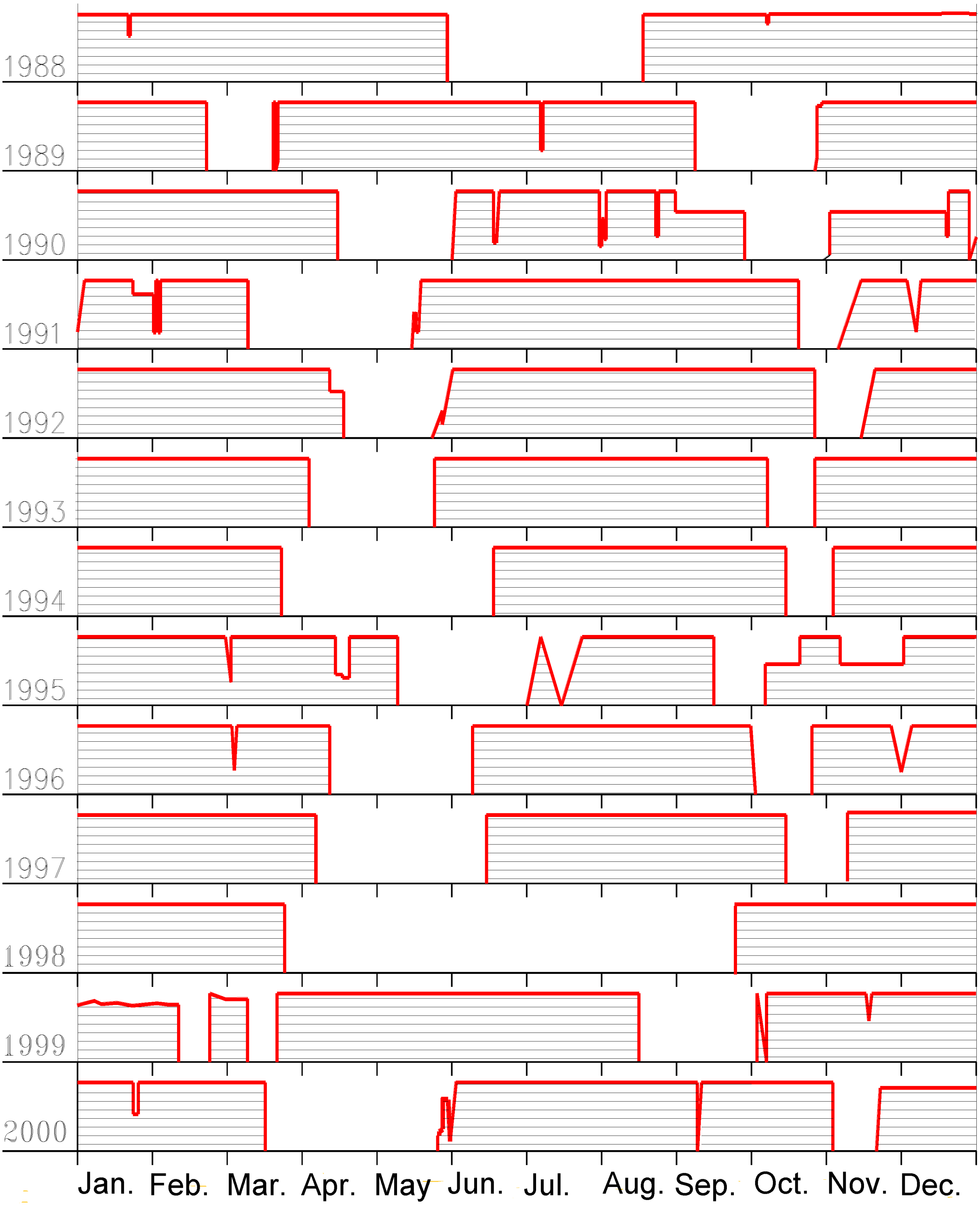

Fig.2.1. BN-600 NPP operating histogram [Source: OKBM, MINATOM, Russia] (reactor is being shut down two times per year for refuelling).

Significant technology development program for LMFRs is proceeding in several countries, namely in France, India, Japan and the Russian Federation. Activities are continued in a number of other countries at lower levels.

Commercial introduction of fast breeder reactors in France has been postponed; however, alternative LMFR application is being developed, namely transmutation of long-lived nuclear waste and utilization of plutonium. Continued operation of Phenix at 350 MW(th) is related to these requirements. One of the objectives of expanding the lifetime of the Phenix reactor is to perform the necessary irradiation experiments in support of the project identified as "Concept of Intensive Plutonium Reduction in Advanced LMFR.

In France, Super Phenix (SPX) plant (rated power of 1200 MW(e)) was successfully commissioned in January 1986 and operated, but not without difficulty. The most long outages of SPX reactor were caused not only by and not because of using sodium as a coolant. Sodium bears relation neither to the fall of the turbine hall roof caused by snow load with extensive damage to the steam plant, nor to the vibration of water/steam piping system resulted from the water hammer. There were also some other incidents in the BOP. As regards sodium systems, a crack appeared in the wall of the spent fuel storage vessel in March 1987. Although there was no sodium leak outside and its ignition (leak into the cavity under the guard vessel), this and second incident (an air leak into an auxiliary circuit causing pollution of the sodium) in 1990 had a large social response because in that time anti-nuclear sentiments raised by Chernobyl in some social groups and governmental structures were too strong. With downturns in energy demand and political changes in 1998 the French Government finally confirmed to discontinue it operation.

In Japan, fast reactor development program symbolises national nuclear fuel recycling program, as it is stated in the national long-term plan [2.3]. The experimental fast reactor Joyo has shown excellent performance for more than 20 years. The prototype reactor Monju (280 MW(e)) was stopped temporarily due to the leak in the non-radioactive secondary circuit in1995. The design of demonstration fast reactor (DFBR-600 MW(e)) is in progress [2.4, 2.5].

Russian experience gained on operation of experimental and prototype fast reactors (BR-10, BOR-60, BN-350 and BN-600) is rather good. Stable operation of the demonstration reactor

BN-600 with nominal power output of 600 MW(e) for 20 years (Fig. 2.1) with an average load factor of ~70%, as well as construction of the largest fast reactor SPX in France, are milestones in the implementation of LMFR technology.

Current efforts with regard to LMFRs in the Russian Federation are directed towards improving safety margins and economics. While these efforts will take some time, an immediate use is foreseen for fast reactors for energy production, as well as Pu and minor actinide utilization. In Russia, detailed design of commercial fast reactor BN-800 was completed, and license was issued for its construction on Yuzno-Uralskya and Beloyarskaya NPP sites.

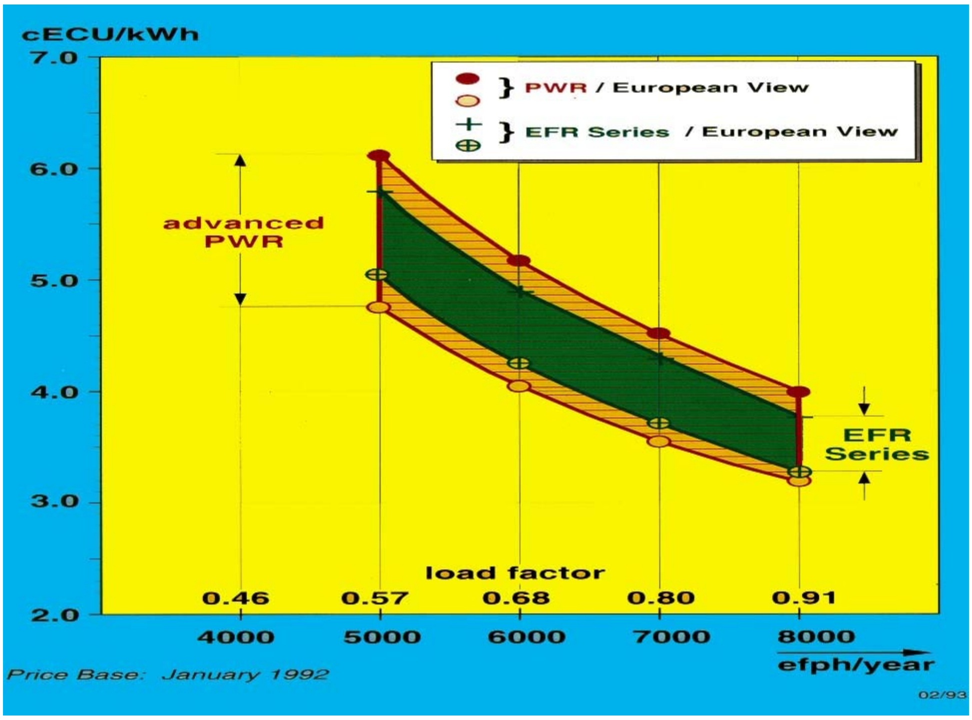

Fig 2.2. Generating cost comparison EFR vs. advanced PWR [Sourse :EFR Associates].

In Western Europe the European fast reactor (EFR) design has been completed. This synthesises the extensive experience from France, Germany and the United Kingdom of large pool-type oxide-fuelled reactors. One of the outstanding achievements of the EFR programme has been to make firm and reliable cost estimates. Construction of a reactor to the EFR design may not be possible in the near future, but a well- validated way forward to commercial utilisation of fast reactors has been established. This way is generally consistent with other studies, and indicate that the goal of competitive fast rectors may be within reach (Fig. 2.2).

India is now in a position to confidently embark on the construction of 500 MW(e) prototype LMFR in this decade. Development of technology for manufacture of key components and of low doubling time fuels and structural materials capable of sustaining high neutron fluence has already been initiated and work is going on satisfactorily. The central facility in the unit is the fast test breeder reactor (FBTR) which is operational from 1985. FBTR is a loop type sodium cooled fast reactor of 40 MW(th) capacity equipped with a steam generator and a turbogenerator to generate 13 MW of electricity. Presently per capita electrical energy consumption in India is about 420 kW•h per annum which is roughly one fifth of world average. Therefore to reach a level of world average itself would require the electricity generation capacity to be raised from 100 GW(e) at present to about 500 GW(e) at least considering the population increase also. Coal and hydro are the sources of electricity now in India and hydro may have reached saturation for all practical purposes because of remoteness of untapped major source and environmental issues. The resource potential of coal is estimated to be about 200 billion tonnes (bt) and at a capacity of 500 GW(e), it may not sustain for more than 70 years.

The only major alternative for India is nuclear energy. However, the potential of natural uranium resources, estimated to be around 50,000 tonnes is negligible [about 1 bt of coal equivalent (btce)] if utilised in an once-through cycle and the capacity also will be limited to about 10 GW(e). If the same U along with the Pu generation in PHWR is invested in FBR the resource potential enhances to 180 btce and the capacity also can be increased to 250 GW(e). FBRs thus form the second stage of the nuclear power of country. FBRs in India will be deployed on U-Pu cycle for rapid growth of nuclear power capacity and generate enough nuclear fuel simultaneously for deployment of Th-U cycle in the third stage the program. Th is abundantly available in India and the resource level of 320,00 tonnes is estimated to be equivalent of 1000 btce [2.5].

It should be noted that in other South and East Asia countries with few indigenous fossil fuel and little uranium ore reserves there is the same situation concerning effective nuclear fuel breeding by LMFR. Republic of Korea's LMFR program consists of development, design and construction of a prototype reactor of 150–350 MW(e) power. The first fully-proven reactor is planned to be in operation by 2025. In China, experimental fast reactor CEFR-25 is planned to become critical in 2005.

The IFR project at the Argonne National Laboratory(USA) has been based on the use of a ternary alloy U-Pu-Zr fuel for its core loading. The IFR concept was integrated by General Electric into a full plant design of an ALMR of 300 MW(e). The plutonium is not separated from the higher radioactive actinides; these are recycled together in the reactor and never leave the reactor site. Advantages of IFR system and metallic fuel are in areas of: (1) fuel performance; (2) passive safety; (3) waste management potential;(4) economics; (5) minimum burn-up swing (BR_{core} ~1, if needs); and (6) proliferation resistance. All IFR designs are based on full actinide recycling using a pyrochemical processing and new fuel fabricating plant collocated with the reactor complex. In fact, by using an U-Pu-10%Zr alloy and ferritic-martensitic HT9 cladding and duct, a burn-up of about 20% has been achieved in the USA EBR-2. All irradiation results in the EBR-2 and FFTF have demonstrated reliable performance of metallic fuel and the potential to achieve high burn-up in prototypical fuel elements cooling by liquid sodium. These ideas is borrowed for some other reactor concepts.

The USA/GE advanced liquid metal cooled reactor (ALMR) was designed to provide high reliability for the key safety functions, including reactor shutdown, heat removal, and containment. These functions can be achieved by passive means (thermal expansion, temperature effects on neutron absorption, natural circulation of the sodium coolant, and natural air circulation).

The significant experience of MOX-fuel elements development for fast reactors has been gained during many years in many countries (Table 2.1).

An important progress has been made since the prototype FR’s were put into operation. Findings of recent work have led to improvement in LMFR designs. For example, an advanced design of the system: main vessel — safety vessel — vault was developed for EFR (Fig. 2.3a). The main vessel is completely surrounded by a leak-tight safety vessel anchored to the surface of the concrete vault. A layer of metallic insulation covers the inside surface of the safety vessel, which reduces both the heat losses and the thermal cycle skin fatigue. A layer of sodium resistant concrete is provided between the safety vessel and the structural concrete of the vault. The structural concrete is kept cool by the vault cooling system. In combination with the solid roof design, it has been possible, firstly, to reach “deterministic” exclusion of radioactive sodium leakage and fire (explosion) and, secondly, to provide a further enlargement of safety margins of the LMFR. These and other advanced designs were implemented simultaneously with substantial specific weights of steel in t/kWe and investment cost reduction in comparison with Superphenix (figs. 2.2, 2.3b).

TABLE 2.1. MOX FUEL BURN-UP [2.6–2.9]

Fuel producer	Reactor	Burn-up,% h. A.
France	Phenix	16.9
Japan	JOYO	13
USA	FFTF	15 (24.5 max)
UK	PFR	23.5
Russia	BOR-60 (vibro-compacted fuel)	33
	BOR-60	24–29
	BN-600	11.8 max

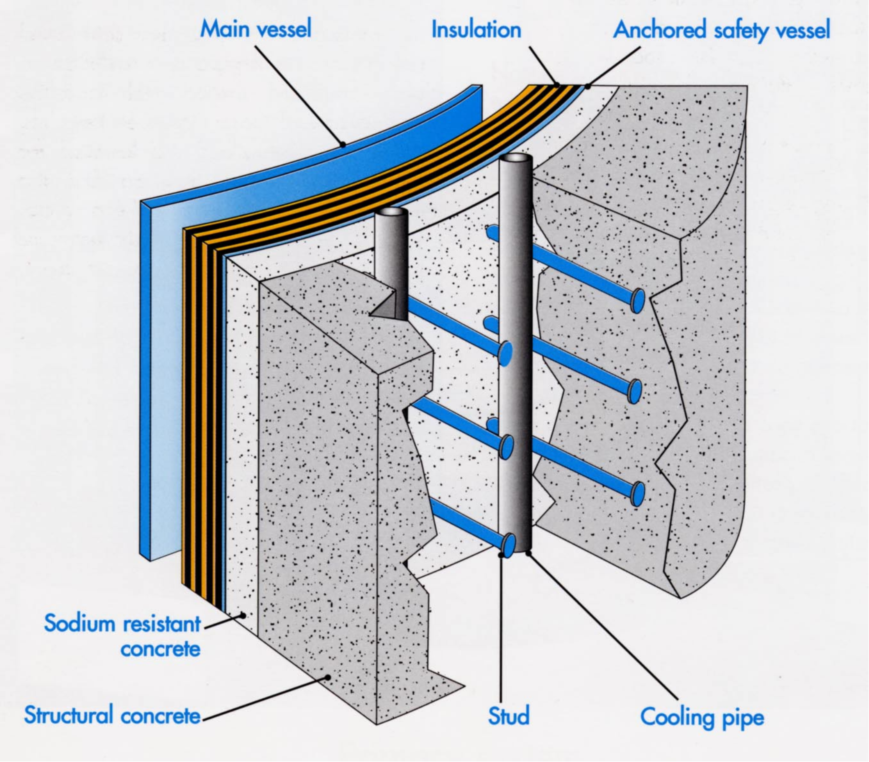

Fig. 2.3a. EFR: Safety vessel and vault [Source: EFR Associates].

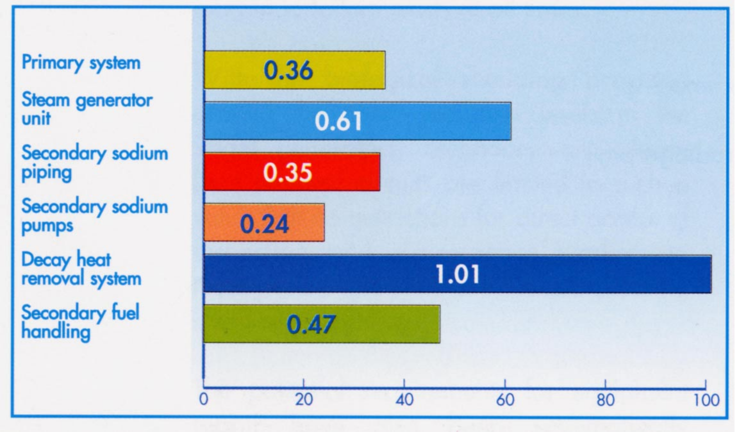

Fig. 2.3.b. EFR vs Superphenix: comparison of the specific steel weight in t/kWe [Source: EFR Associates].

The current view is that the technologies of sodium coolant and mixed oxide fuels are largely mastered, and large prototypes and demonstration LMFRs have been built and have clearly demonstrated that a LMFR is capable of sustained reliable contribution to an electricity supply system. Fast reactors will contribute not only to power production, but also to waste management and the control of nuclear materials. The world-wide investment already made in the development and demonstration of LMFR technology exceeds US $50 billions.

Since the remaining cost to solve the outstanding technical and scientific issues to make this technology commercially available is relatively modest, the completion of development makes economic sense. With regard to LMFR design approaches the accumulated knowledge on materials, thermohydraulics and mechanical science indicates, that a substantial decrease in investment costs together with better assurance that safety margins are effectively maintained, might be provided by an accurate analysis of actual transients, as well as by an optimum reactor and auxiliary systems design [2.10–2.18].

Perhaps there is only one disadvantage inherent in the LMFR coolant-liquid sodium, namely that it interacts chemically with air and water/steam. Therefore, providing integrity of the sodium circuits is the most important requirement to observe on the LMFR design, construction and operation tests.

The solution of the problem of the reliable elimination of coolant leaks is determined by the application of experimentally and analytically proved design, structural materials, manufacture and installation, as well as quality control of all stages of LMFR components manufacture. Technological procedures and approaches, as well as quality criteria, should strictly correspond to the related regulatory documents.

Efficient criteria and rules and effective LMFR technology, as experience has shown, can be established by design, construction and comprehensive testing of three or four LMFR plants. This stage has been reached in some countries. For example, in Russia an experimental reactor BR-10, an experimental 15 MW(e) NPP BOR-60, a prototype NPP BN-350 (presently in Kazakhstan) and a semi-commercial NPP BN-600 have been operating respectively for 40, 30, 25 and 20 years, providing invaluable information on FR technology. That is why the BN-600 plant has been running successfully for 20 years with an overall lifetime load factor of ~70%. This success was achieved because in the design and construction of the plant, manufacture of the equipment and operation of the plant past errors were not repeated and good design solutions were incorporated.

The comprehensive operational experience with LMFRs BN-350, Phenix, PFR, BN-600, Super Phenix and Monju has shown that, if plant components have been designed and manufactured without errors and representative specimens or models have been tested prior to installation, reliable operation can be ensured during the whole operational life.

The very low corrosion activity of sodium, the near atmospheric operating pressure, the use of ductile structural materials, and the reliable heat removal by a coolant having no phase change, imply that there should be nothing to provoke loss of the sodium system integrity in a LMFR.

2.2. LEAD AND LEAD-BISMUTH COOLED REACTOR STUDIES

Studies of lead-bismuth and lead-cooled fast reactors are being carried out in the Russian Federation (RF) organizations: Institute of Physics and Power Engineering (IPPE) and EDO

GIDROPRESS, in which a great deal of experience has been accumulated in the course of the development and operation of submarine reactors cooled with lead-bismuth eutectic. However, bismuth is expensive and the resources are limited. It is possible that its use must be confined to special applications, such as small reactors or to a limited number of fast reactors. For this reason lead cooling is also being studied in the IPPE, Kurchatov Institute, and other organization [2.19–2.26].

The thermohydraulic features of lead-bismuth and lead coolants are high boiling temperatures and the relative inertness compared with sodium. The melting and boiling points of sodium are respectively 98°C and 883°C. For lead-bismuth eutectic the respective figures are 123.5°C and 1670°C and for lead 327°C and 1740°C at atmospheric pressure. The boiling points are well above cladding failure temperatures. The specific heats per unit volume of lead-bismuth and lead are similar to those of sodium but the conductivities are about a factor of 4 smaller.

The γ-radioactivity induced in Pb and Pb-Bi coolants is low so that access can be made to the coolant circuit after a shutdown period of about 24 hrs. However the production of α-radioactive ^{210}Po from bismuth, and to a lesser extent from lead, poses problems because of its migration from the coolant to the cover gas and formation of aerosols. ^{210}Po is volatile, so that any leakage from the cover gas poses a hazard to the plant operators and the environment. In the early stages of development, the formation of deposits of lead oxide and other impurities posed problems.

A careful control of the purity of the coolant is required to avoid the formation of such deposits. It was necessary to develop corrosion resistant steels and to pre-treat the surfaces of components and also to use special inhibitors in the lead-bismuth coolant. More extensive studies are required for lead coolant to demonstrate the corrosion-resistance of structural material.

It was found [2.27], that the specific α -activity of the typical lead-bismuth coolant is defined by the Bi-210m (half-life =3.6×10^6 years), which generates in reaction Bi-209 (n,γ) Bi-210m. The long-lived β-activity of Bi-208 (half-life = 3.65×10^5 a) generates in reaction Bi-209 (n, 2n) Bi-208.

The most important contributor to specific long-lived residual radioactivity of a lead coolant is Pb-205 (half-life = 1.51×10^7 a), which generates in the reaction Pb-204 (n, γ)Pb-205. The specific β-activity of a pure lead coolant is significantly less than one of lead-bismuth. Activation of lead-bismuth and lead coolants will increase in every cycle of reuse, if reuse is possible in principle [2.27].

Lead-bismuth reactors [2.25–2.30]. Two design concepts have been studied:SVBR-75 (Fig. 2.4) and ANGSTREM. SVBR-75 is designed to produce 75 MW(e). The study explores the feasibility of designing an SVBR-like reactor core to operate for 10 years without refuelling. A transportable version of the reactor called ANGSTREM can produce 30 MW(th) or 6 MW(e) or a combination of heat and electricity. A version producing up to 25 MW(e) has also been studied.

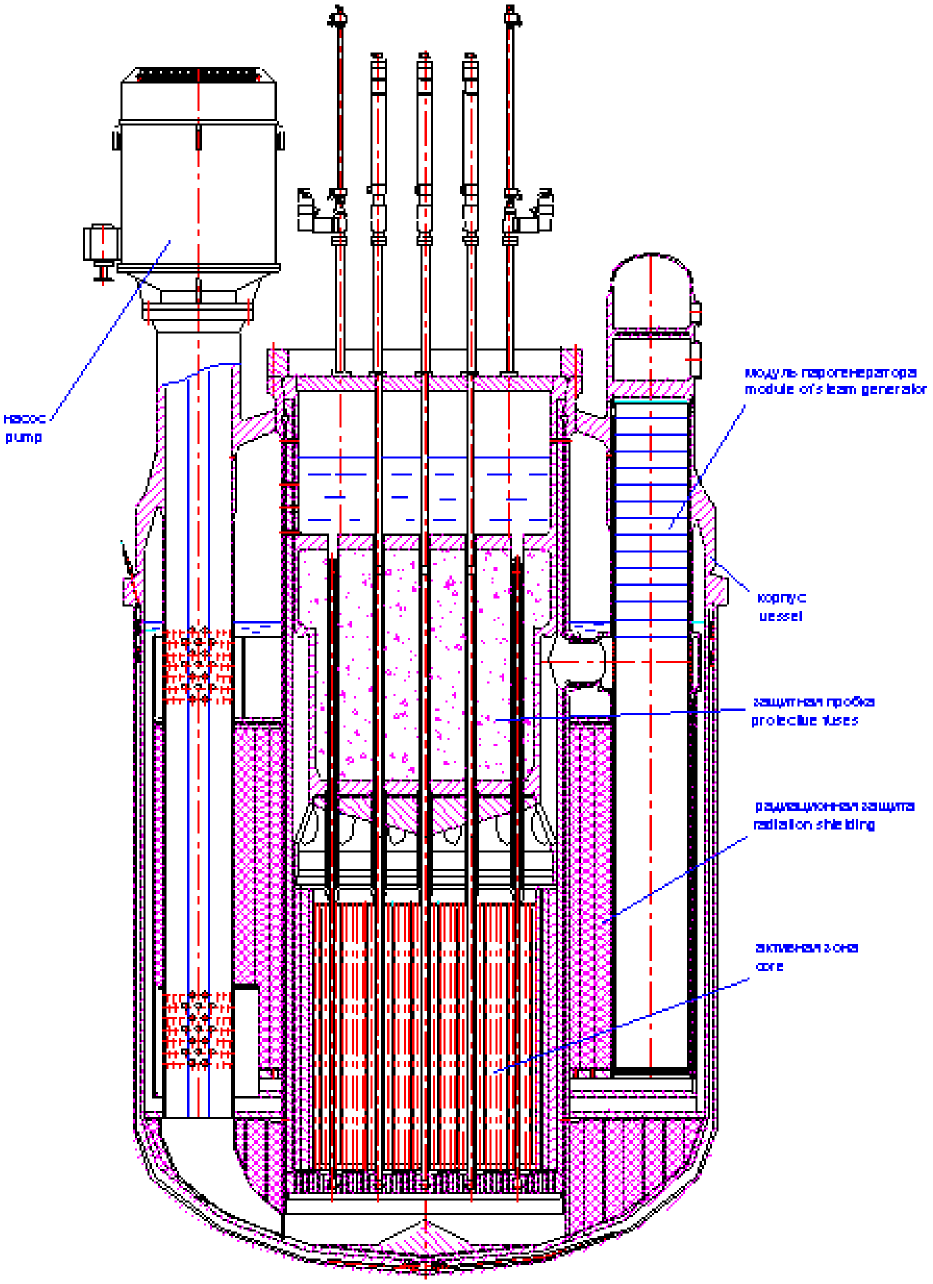

Fig.2.4. Lead-bismuth cooled reactor [Source: SSR IPPE, EDO GIDROPRESS].

The project of BRUS-150 integral type reactor of 500 MW thermal power and 150 MW electric power has been developed. The core, steam generators, pumps and all lead-bismuth loops are located in the reactor vessel, so that leak-proof vessel contains total amount of lead-bismuth. BRUS-150 reactor can be also used for transmutation of minor actinides accumulated in WWER type reactors and for utilization of weapon grade plutonium (for the main characteristics -see Table 2.2)

Lead and lead-bismuth alloy are inflammable in air or water. In principle, a Pb-Bi and Pb cooled reactor would not have to have an intermediate circuit between the primary coolant and the steam, however it is known, that there had been severe incidents with Pb-Bi cooled reactor due to water/steam leaks occurred in the submarine steam generator [2.30]. As a result of one leak some areas of the reactor core were plugged by products of water and Pb-Bi interaction, causing meltdown of the core. Therefore the elimination of the intermediate circuit between the primary coolant (Pb,Pb-Bi) and the water/steam is questionable and needs additional R&D efforts.

Lead-cooled reactors. The main concept of lead cooled fast reactor is integral (or semi-integral) design with nitride fuel. Two-circuit reactor plant design with supercritical pressure steam-water cycle is considered. The BREST-300 and BREST-600 reactor projects (rated power of 300 MW(e) and 600 MW(e), respectively) were developed in Russia by RDIPE in co-operation with RRC Kurchatov Institute (Fig. 2.5). Two-circuit design with steam/steam reheating and feed water preheating up to 340^{o}C with live steam of supercritical parameters have been developed. Heat removal from the reactor core is accomplished by lead forced flow provided by the pumps installed in four parallel loops. Lead coolant flows through the fuel subassemblies where it is heated up to 540^{o}C. In the steam generator lead is cooled to about 420^{o}C.

The total weight of the BREST-300 reactor and coolant is 8000 tons, 600 m^3 of lead being filled into preheated reactor vessel made of austenitic steel. The main characteristics of the reactors BREST- 300 and BREST-600 are presented in Table 2.3.

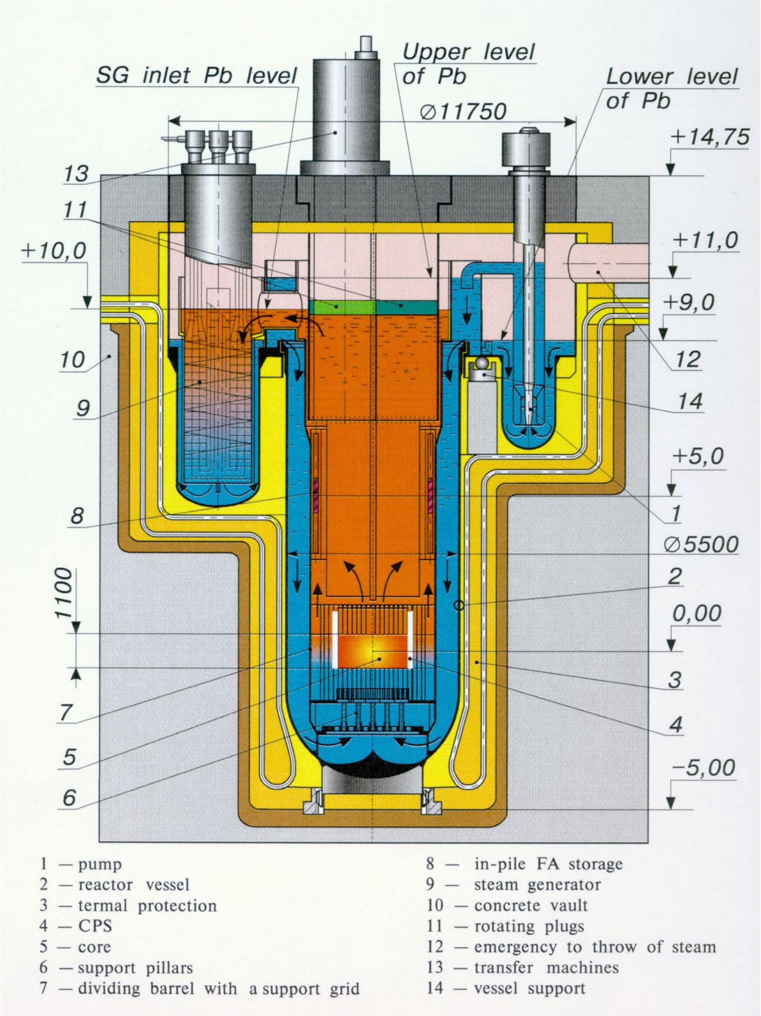

Fig.2.5. BREST-300 reactor design [Source: RDIPE, MINATOM, Russia].

TABLE 2.2. MAIN PHYSICAL CHARACTERISTICS OF BRUS-150 REACTOR

Characteristics	Units	Variant with $\Delta p=0$	Variant with $\frac{G^{Pu^{+238}}}{G^{Pu}} \le$
Power	MW(th)	500	
Core D_{equiv} /H	mm	1962 / 1300	
Fuel		PuO_2+UO_2+actinides	
Initial plutonium nuclides composition Pu-239 / Pu-240 / Pu-241 / Pu-242	%	60/25/10.4/4.6	
Initial actinides composition Np-237 / Am-241 / Am-242m / Am-243 / Cm-242 / Cm-244 / Cm-245	%	50/31/003/13.5/0.5/0.03/5/0	
Loading of Pu-239+Pu-241 at the beginning of reactor run under steady state conditions	kg	1290	1329
Loading of MA at the beginning of reactor run under steady state conditions	kg	1646	991
Annual loading of actinides from spent fuel of WWER	kg	100	48
Annual loading of plutonium	kg	427	443
Maximum burn-up	%	5.2	6.3
Operating period of fuel subassemblies (load factor ~0.8)	year	3	3
Refuelling interval (reactor run)	year	1	1
Reactivity change between refuellings	ΔK/K,%	0	–0.28
Pu-238 content in unloaded fuel	%	7.2	5.0
Temperature reactivity coefficient	10^{-5}, 1/K	-(1.12–0.86)	-(1.50–1.09)
Void reactivity effect under fuel leakage of coolant	%	–0.6	–1.8
Delayed neutron effective fraction	%	0.37	036
Maximum neutron flux for operating period of fuel subassemblies	n/cm^2 (dpa)	$1.3 \cdot 10^{23}$	(30)

Ferritic-martensitic 12 percent chromium steel having corrosion resistance in molten lead, low radiation induced swelling and acceptable strength and creep characteristics was selected as a candidate material for fuel cladding and elements of fuel subassemblies. Results of out-of-pile corrosion tests conducted within 400–650^{o}C temperature range during 8000 h were extrapolated for 35000 h (5 year operation of fuel subassembly) gave ~120 μm and 60 μm reduction of cladding wall thickness at 650^{o}C and 550^{o}C respectively.

TABLE 2.3. CHARACTERISTICS OF LEAD COOLED REACTORS

Characteristics	Units	BREST-600	BREST-300
Thermal power	MW(th)	1400	700
Electric power	MW(e)	600	300
SA number in core	-	357	185
Core diameter	mm	3190	2300
Core height	mm	1100	1100
Fuel element diameter	mm	9.1; 9.6; 10.4	9.1; 9.6; 10.4
Fuel element pitch	mm	136	13.6
Reactor core fuel	-	UN+PuN	UN+PuN
Fuel loading (U+Pu)	t	28	16
Loading of Pu/Pu-239+Pu-241	t	3.73/2.72	2.2/1.6
Fuel lifetime	years	5–6	5
Refueling interval	years	1–2	~1
Core breeding ratio	-	~1	~1
Lead inlet/outlet temperature	^{o}C	420/540	420/540
Peak cladding temperature	^{o}C	650	650
Maximum lead velocity	m/s	1.8	1.8
Power reactivity effect	%ΔK/K	0.17	0.16
Total reactivity effect	%ΔK/K	0.33	0.32
Delayed neutrons fraction, β_{eff}	%ΔK/K	0.35	0.35
SG outlet steam temperature	^{o}C	520	520
SG outlet steam pressure	MPa	24.5	24.5
Net efficiency	%	~ 44	~ 44

Ductless fuel assemblies are used in the core. Each of three radial zones of the core consists of fuel pins with different diameters (9.1/9.6/10.4mm) with the same fuel composition and fuel pin pith: small pins in central and big in outer zone. A rather large fuel pin to pin distance (13.6 mm) provides the large coolant flow cross section, suitable core hydraulic resistance. To reach core breeding ratio equal one, MOX fuel was replaced by U-Pu nitride.

Low moderation and absorption of neutrons in lead gives benefit in the breeding ratio. Partially owing to this, core breeding ratio value equal to one can be achieved in lead cooled reactors with nitride fuel. However, use of lead coolant results in some negative effects. For instance, pressure drop in the core is much higher (about 7 times) than that in case of sodium coolant with other conditions being equal (reactor power, coolant flow cross section area in the core, coolant temperature rise and fuel element length). This pressure drop increase is caused by considerably lower thermal capacity of lead as compared to that of sodium, and cannot be compensated by its higher density. In order to decrease core hydraulic resistance, flow cross section area in the core should be increased by means of both reduction of volumetric fuel fraction and increase of the core diameter.

That is why overall core dimensions of 300 MW(e) BREST-300 lead cooled reactor are rather large: D/H = 2.3/1.1 m with less fuel volumetric fraction (0.23–0.32), while in the sodium cooled 800 MW(e) BN-800 reactor (LMFR), this ratio is D/H = 2.5/0.88 m, fuel volumetric fraction being equal to ~0.4. Simple extrapolation made for BREST type lead cooled reactor of 800 MW(e) power shows that overall dimensions of its core would be D/H = 3.7/1.1 m.

Analysis has shown that owing to increase of the BN-800 type reactor core diameter from 2.5 m to 3.7 m the following benefit can be achieved:

- coolant velocity in the core becomes about two times lower causing almost five times decrease of pressure drop value (down to ~1 atm), thus providing significant (up to 12–15%) increase of natural flow rate of the coolant required for passive removal of the reactor decay heat under abnormal operating conditions;

- fuel element diameter and fuel volumetric fraction in the core can be increased, assuring BR_{core}~1 even in sodium cooled reactor (on condition that higher density nitride fuel is used), in spite of elimination of both radial and axial fertile blankets.

Studies made at the Institute for Physics and Power Engineering have confirmed the possibility of achieving $BR = BR_{core} \cong 1$ with typical for LMFRs volumetric fraction of nitride fuel in the core of sodium cooled 800–1600 MW(e) reactor, its core overall dimensions being much lower than those of lead cooled reactor [2.31].

The above considerations confirm the fact that comparison of physical and economical characteristics of the reactors with different coolants should be made on the basis of identical input data, such as core dimensions, power rating, as well as the fuel type. Careful comparison does not reveal any advantages of lead cooled reactors as compared to LMFRs from the standpoint of achievement of $BR_{core} \sim 1.0$ value and assurance of the reactor decay heat removal using passive means under abnormal operating conditions.

Also, it should be noted that sodium cooled reactor is nuclear technology system, having maximum flexibility extent, i.e. it is capable of assuring the following operation modes in accordance with the user requests:

- fuel breeding ($BR>1$ with T_2 maintained at the acceptable level);
- fuel self-supply ($BR\sim1.0$);
- controlled plutonium build-up ($BR<1$).

Thus, sodium cooled reactors as well would fully come up to the concept of development of nuclear technology systems having no enriched uranium, declared at the UN Millenium Summit. Moreover, no Pu stocks will exist outside LMFR with attached plant for fuel reprocessing and fabrication (e.g. similar to that developed at the ANL for the IFR).

Preliminary design study of lead cooled fast reactor with nitride fuel assemblies has been performed by the Japanese specialists to improve uranium resource utilization and transmutation of HLW nuclides. Plant size limitations caused by seismic resistance requirements and integrated reactor design with compact helical-coil steam generator were studied. The core having inherent safety with negative void reactivity and very small burn-up reactivity of $0.3 shows high breeding ratio. The core, support structure and the primary heat removal system components are located in the reactor vessel. The steam generator and its helical coil tubes are located around the core and core diagrid. Once-through steam generator with helical coil tubes would assure minimum size of the reactor vessel with maximum free lead surface area that is helpful in case of lead-steam interaction. The space between the outer shell of SG and the reactor vessel is low temperature area in the reactor vessel. The main characteristics of the considered reactor are presented in Table 2.4.

TABLE 2.4. CORE PARAMETERS OF JAPANESE LCFR REACTOR

Reactor power	MW(th)	1500
Core height	mm	1000
Core diameter		3550
Blanket height		300
Pin diameter	mm	10.2
Pitch/diameter ratio	-	1.51
Fuel, 12%Pu	-	$(U, Pu)^{15}N$
Loaded Pu amount	t	4.0
Burn-up	GWd/t	92
EFPD	-	2000
Burn-up swing	$\%\Delta K/K$	0.14
Breeding ratio	-	1.26
Void reactivity	$\%\Delta K/K$	–1.5
Doppler coefficient	$(10^{-3}T\Delta K/\Delta T)$	–3.7
Pb density coefficient	$\%\Delta K/K$	$3.55 \cdot 10^{-4}$
Fuel temperature coefficient	$\%\Delta K/K$	$-7.88 \cdot 10^{-4}$
β_{eff}	$\%\Delta K/K$	0.44

2.3. CONCLUSIONS

In conclusion it should be pointed out, that the drawbacks inherent in the lead and lead-bismuth coolants give rise to the following possible problems:

- damage of components and fuel elements caused by high corrosivity of heavy metals;

- freezing of *lead* in the steam generator in case of failure of high pressure feed water heaters of the turbine plant and supply of low temperature feed water (150–170°C below the melting point of lead) from low-pressure feedwater tank(LPFT) to SGs (it is for the first time in the power engineering practice, that melting point of proposed coolant is higher than the temperature of water in LPFT);
- problems concerning repair and maintenance work and remotely controlled refueling operations in *lead-cooled* reactor carried out at rather high *lead* temperature (over 400°C);

- blockage of coolant flow cross section in the fuel subassembly caused by the products of water/steam interaction with coolant in SG, as well as solid inclusions and structural material corrosion products suspended in the heavy coolant;
- problems with production by heavy metals of long-lived isotopes.

If these will be eliminated on the basis of choice of available structural materials and/or development of new ones and innovative design approaches, and if operational reliability of two-circuit pool-type reactor with supercritical SG located inside the reactor vessel will be demonstrated then *the most important advantage of lead and lead-bismuth-cooled reactors will be the possibility to eliminate the safety concerns of LMFR caused by sodium chemical reactivity with air and water with explosion potential*.

All other supposed advantages of fast reactor with heavy metal coolant, namely:

- minimum reactivity excess (BR_{core} ~1);
- transmutation of actinides and long living fission products;
- proliferation resistance by elimination of fertile blankets and application of pyrometallurgical methods of on-site fuel reprocessing and fabricating;
- safely termination of accidents on the basis of passive mechanisms; and
- economical competitiveness;

have been confirmed in practice or justified within the framework of advanced designs of sodium cooled fast reactors [BN-600 (Russia), aLMR(USA), EFR(Western Europe) and BN-800 (Russia)].

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3. PHYSICAL AND CHEMICAL PROPERTIES OF REACTOR COOLANTS

3.1. PHYSICAL PROPERTIES

The highest purity extent of commercial-grade sodium is 99.8%, this value corresponding to the purity of sodium that passed through the cold or hot traps [3.1, 3.2]. However, sodium of 99.995% purity can be obtained using distillation.

Sodium has one stable isotope, namely: ^{23}Na. Characteristics of other isotopes of sodium are presented in Table 3.1. The main isotopes result from the following reactions:

$$^{23}\mathrm{Na}(n, 2n) \rightarrow {}^{22}\mathrm{Na}(\beta^{+},\gamma);$$

$$^{23}\mathrm{Na}(n) \rightarrow {}^{24}\mathrm{Na}(\beta^{-},\gamma) \rightarrow {}^{24}\mathrm{Mg}.$$

It is just ^{24}Na isotope that determines the main contribution to the radioactivity of the coolant flowing in the circuit.

Crude lead contains 93–99% of the basic metal. Among the basic impurities are Cu (1–5%), Sb, As, Sn (0.5–3%), Bi(0.05–0.4%), Al and Au. The highest purity extent of lead is 99.992%. Lead has four stable isotopes, namely: 204, 206, 207 and 208. The last three are the products of U, Ac and Th decay. Characteristics of lead isotopes are given in Tables 3.2 and 3.3.

The main thermophysical properties of sodium, lead, bismuth and lead-bismuth eutectic alloy (44.5% Pb-55.5% Bi) are presented in the Table 3.4. [3.3]

Other thermophysical properties of these materials vs temperature are given in Tables 3.5–3.7 [3.3; 3.4].

The main thermophysical properties of basic sodium compounds are given in Table 3.8. [3.2].

TABLE 3.1. SODIUM ISOTOPE CHARACTERISTICS

Isotopes	Atomic mass	$T_{1/2}$	Energy, MeV
20	19.99	0.4 s	β^+ (5.5; 7.15; 11.2)
21	20.99	23 s	β^- (2.5)
22	21.99	2.6 years	90% β^+ (0.54; 2.8;); 10% (ec) γ
23	22.99	-	-
24	23.99	14.97 h	β^- (5.5); γ (1.38; 2.76)
25	24.99	60 s	β^- (3.7; 4.0); γ (0.37; 0.58; 0.98)

TABLE 3.2. STABLE LEAD ISOTOPES

Pb isotopes	Content, % mass	Neutron capture cross section, barn	Integral resonance, barn
204	1.48	0.661±0.070	1.7±0.5
206	23.6	0.0305±0.008	0.2±0.1
207	22.6	0.0709±0.010	0.4±0.2
208	52.3	0.487±0.030	-
Natural Pb		0.170±0.002	0.16±0.05

TABLE 3.3. UNSTABLE LEAD ISOTOPES

Isotopes	$T_{1/2}$	Energy, MeV
205	1.5×10^7 years	
209	3.3 hours	β^- (0.6)
210	22 years	β^- (0.029); γ (0.046)
211	36 min	β^- (1.35); γ (0.065–1.265)
212	10.64 hours	β^- (0.16–0.58); γ (0.1–0.3)
214	26.8 min	β^- (0.65–1.03); γ (0.53–0.35)

TABLE 3.4. THERMOPHYSICAL PROPERTIES OF SODIUM, LEAD, BISMUTH AND LEAD-BISMUTH EUTECTIC ALLOY (44.5% PB; 55.5% BI)

Properties	Na	Pb	Bi	PbBi
Atomic number	11	82	83	-
Atomic mass	22.99	207.2	208.98	-
Melting temperature, ^{o}C	98	327.4	271.4	125
Boiling temperature, ^{o}C	883	1745	1552	1670
Heat of melting, kJ/kg kJ/mole	114.8 2.6	24.7 5.1	54.7 11.4	38.8 8.07
Heat of vaporisation, kJ/kg kJ/mole	3871 89.04	856.8 178	852 178	852 178
Density, kg/m^{3} at 20^{o}C (solid) at 450^{o}C (liquid)	966 845	11340 10520	9780 9854	10474 10150
Heat capacity, kJ/kg K at 20^{o}C (solid) at 450^{o}C (liquid)	 1.230 1.269	 0.127 147.3	 0.129 150	 0.128 146
Thermal conductivity, W/m K at 20^{o}C (solid) at 450^{o}C (liquid)	 130 68.8	 35 17.1	 8.4 14.2	 12.6 14.2
Kinematic viscosity, m^{2}/s at 450^{o}C	$3 \cdot 10^{-7}$	$1.9 \cdot 10^{-7}$	$1.3 \cdot 10^{-7}$	$1.4 \cdot 10^{-7}$
Prandtl number at 450^{o}C	0.0048	0.0174	0.0135	0.0147
Surface tension, mN/m at 450^{o}C	163	480	370	392
Volume change with melting,%	+2.65	+3.6	–3.3	~ +0.5

TABLE 3.5. THERMOPHYSICAL PROPERTIES OF LEAD

t, ^{0}C	ρ, (x10^{3}) kg/m^{3}	C$_{p}$, J/kg·K	λ, W/m·K	a, (x10^{-6}), m^{2}/s	ν, (x10^{-7}), m^{2}/s	Pr, (x10^{-2})	σ, (x10^{-3}), N/m	ρ$_{e}$, (x10^{-8}), Ohm·m
330	10.67	147.30	15.83	10.07	23.90	2.37	445.81	93.77
400	10.58	147.30	16.58	10.64	20.99	1.97	441.33	97.02
500	10.46	147.30	17.66	11.46	17.63	1.54	434.93	101.67
600	10.34	147.30	18.74	12.31	15.19	1.23	428.53	106.32
700	10.21	147.30	19.82	13.18	13.69	1.04	422.13	110.97
800	10.09	147.30	20.90	14.07	13.13	0.93	415.73	115.62

TABLE 3.6. THERMOPHYSICAL PROPERTIES OF PBBI EUTECTIC

t, ^{0}C	ρ, (x10^3) kg/m^3	C_p J/kgK·	λ, W/m·K	a, (x10^{-6}), m^2/s	ν, (x10^{-7}), m^2/s	Pr, (x10^{-2})	σ, (x10^{-3}), N/m	ρ_e, (x10^{-8}), Ohm·m
130	10.55	146.00	11.0	7.17	29.41	4.10	415.65	104.41
200	10.46	146.00	11.7	7.68	24.85	3.24	410.73	108.07
300	10.33	146.00	12.7	8.43	19.52	2.32	403.70	113.30
400	10.21	146.00	13.7	9.20	15.58	1.69	396.67	118.53
500	10.08	146.00	14.7	9.99	13.03	1.30	389.64	123.76
600	9.96	146.00	15.7	10.80	11.87	1.10	382.61	128.99
700	9.83	146.00	16.7	11.63	12.10	1.04	375.58	134.22
800	9.71	146.00	17.7	12.48	13.72	1.10	368.55	139.45

TABLE 3.7. THERMOPHYSICAL PROPERTIES OF SODIUM

t, ^{0}C	ρ, (x10^3) kg/m^3	C_p, J/kg·K	λ, W/m·K	a, (x10^{-6}), m^2/s	ν, (x10^{-7}), m^2/s	Pr, (x10^{-2})	σ, (x10^{-3}), N/m	ρ_e, (x10^{-8}), Ohm·m
100	0.925	1382.3	84.92	66.42	7.367	1.11	200.06	9.49
200	0.904	1343.3	81.02	66.68	5.001	0.75	189.19	12.8
300	0.881	1309.4	77.12	66.85	3.876	0.58	178.86	16.6
400	0.856	1282.6	73.19	66.64	3.238	0.49	168.96	20.8
500	0.832	1263.9	69.28	65.91	2.836	0.43	159.40	25.6
600	0.808	1253.5	65.37	64.57	2.562	0.40	150.07	31.1
700	0.784	1251.7	61.46	62.62	2.368	0.38	140.88	37.2
800	0.761	1258.6	57.55	60.10	2.226	0.37	131.73	44.3

TABLE 3.8. THERMOPHYSICAL PROPERTIES OF BASIC SODIUM COMPOUNDS

Compounds	Properties					
	Temperature, ^{0}C		Heat of formation, kJ/mole	Density, kg/m^3 at 20^{o}C	Heat capacity, J/mole·K	Heat of melting, kJ/mole
	melting	boiling				
NaH	420	decom-position	65.8	1400	–	–
Na_2O	1350	decom-position	420	2270	70	29.7
Na_2O_2	596		596	2600	–	16.7
$NaOH$	318		426	2130	59.5	6.4
$NaCl$	801	1390	410	2165	50.5	28
Na_2CO_3	858	1490	1129	2530	112.3	28
Na_2S	1168	decom-position	359	1856	79.5	30
Na_2SO_4	884	890	1388	2663	118	24.3

3.2. CHEMICAL PROPERTIES OF SODIUM

Sodium is the most electropositive metal. Since it is ahead of hydrogen in electric tension row, Na displaces H_2 out of water with production of hydroxide: NaOH. When interacting with dry hydrogen sodium forms NaH hydride, which is soluble in sodium:

$$2Na(s) + H_2(g) \rightarrow 2NaH(s) + 57.4\ kJ/mole,$$

At the temperature of 420°C NaH is decomposed with release of hydrogen. This fact should be taken into account when gas-tight vessel is heated.

When sodium interacts with the small amount of oxygen, Na_2O oxide is produced, whereas its burning in the air results in Na_2O_2 peroxide:

$$4Na(s) + O_2(g) \rightarrow 2Na_2O(s) + 416\ kJ/mole;$$

$$4Na(l) + O_2(g) \rightarrow 2Na_2O(s) + 436\ kJ/mole;$$

$$2Na(s) + O_2(g) \rightarrow 2Na_2O_2(s) + 499\ kJ/mol.$$

In the molten sodium, only Na_2O oxide is stable, while Na_2O_2 dissociates as follows:

$$Na_2O_2 + 2Na \rightarrow 2Na_2O.$$

When sodium interacts with water, some reactions occur and, depending on reaction temperature, impurities of different composition would be present in sodium.

Hydroxide is produced by the following reaction at 201–300°C:

$$2Na + 2H_2O \rightarrow 2NaOH +142\ kJ/mole.$$

Oxide as well as hydroxide is produced at 350–400°C:

$$2Na + NaOH \rightarrow 2Na_2O + NaH;$$

$$2Na + H_2 \rightarrow 2NaH.$$

At t>420°C, hydride dissociates and reaction goes escaping intermediate stages:

$$2Na + H_2O \rightarrow Na_2O + H_2.$$

Burning reaction is characterized by a zone of small flames at the sodium-air interface, formation of Na_2O on the sodium surface and vigorous emission of high density white oxide fumes.

However, this burning causes relatively low heat release, namely 420 kJ/mole by Na_2O or 500 kJ/mole by Na_2O_2. This is equivalent to about 10 kJ (~2 kcal) per gram of sodium burnt, being approximately equal to that for 1 gram of sulphur, one third of gram of magnesium or aluminium, and less than one quarter of gram of gas/oil. With the same volume of fuel, sodium burning results in the energy release equal to 50% of that for sulphur, 30% of that for gas/oil, 25% of that for magnesium and slightly over 10% of that for aluminium.

Saturation concentration of element in solution (C_s, wt%) follows the Arrhenius equation:

$$lg\, C_s = A - B/T, \tag{3.1}$$

where A and B are experimentally determined constants: A-entropy of solution, kJ/mole and B-heat of evaporation, kJ/mole. Therefore, Eq. 3.1 can be used to estimate solubility of chemical elements in liquid metals as well as material corrosion resistance in the liquid metals. Solubility of different elements in liquid sodium is given in Table 3.9 [3.4].

TABLE 3.9. SOLUBILITY OF DIFFERENT ELEMENTS IN LIQUID SODIUM

Elements	Constants of Eq. 3.1		Temperature range, K
	A	B	
C	7.2	5465	873–1223
Cu	5.45	3055	573 973
Fe	5.16	4310	573–1173
H_2	6.067	2880	273–673
Mo	3.27	3962	1073–1278
Na_2O	1.2	1777	373–873
Ni	2.07	1570	573–1173
O_2	6.257	2444	383–823

3.3. CHEMICAL PROPERTIES OF LEAD

In dry air solid lead is not practically oxidized, whereas in humid air it is coated with oxide film of PbO. This film under air exposure is transformed to basic carbonate $3PbCO_3 \cdot Pb(OH)$. However, oxidation of molten lead by air occurs resulting in its transformation initially into Pb_2O and then into PbO oxide. At the temperature of 450°C the latter is transformed to Pb_2O_3 and then at 450–470°C to Pb_3O_4. Being unstable all these compositions dissociate into PbO and O_2.

Lead interacts with water producing hydroxide $Pb(OH)_2$. In hard water lead is coated with film, preventing water from being polluted with soluble lead compounds. Detectable lead amount dissolves in distilled water resulting in water poisoning.

High lead resistance to diluted sulphuric acid is of practical value. Lead practically does not dissolve in diluted hydrochloric acid, whereas it readily dissolves in nitric acid because of high oxidizability of lead.

Lead dissolves in alkaline medium, although its solubility rate is low. Lead does not interact with either nitrogen or carbon. Lead resistance to chlorine exceeds that of Al, Cu and Fe up to 300°C, because lead chloride exhibits protective properties. Interaction of lead with most electropositive metals (Li, Na, Mg, Ca, Ba, Zr, Hf, etc.) results in formation of intermetallic compounds.

Solubility of different elements or their corrosion resistance in liquid lead and eutectic PbBi alloy is shown in Table 3.10 [3.5] and Table 3.11 [3.1].

TABLE 3.10. SOLUBILITY OF DIFFERENT ELEMENTS IN LIQUID LEAD

Elements	Constants of Eq. 3.1		Temperature range, K
	A	B	
C	1.026	3850	350–1000
Co	2.60	4400	350–1650
Cr	3.74	6750	908–1210
Cu	2.72	2360	327–1000
Fe	0.34	3450	330–910
H_2	–1.946	2360	500–900
Mn	2.02	1825	327–1200
Mo	solubility <10^{-3} wt.% at 1000°C		
N_2	no solubility		
Nb	solubility <10^{-5} wt.% at 1000°C		
Ni	2.78	1000	330–1300
O_2	–0.106	2176	350–850
Si	3.886	7180	1050–1250
Ti	solubility ~$5.6 \cdot 10^{-4}$ wt.% at 500°C		
U	3.921	5121	400–800
Zr	solubility ~$1.2 \cdot 10^{-9}$ wt.% at 500°C		

TABLE 3.11. SOLUBILITY OF DIFFERENT ELEMENTS IN LIQUID EUTECTIC Pb-Bi ALLOY

Elements	Constants of Eq. 3.1		Temperature range, K
	A	B	
C	–1.36	1870	-
Cr	–0.02	2280	400–500
Fe	2.01	4380	-
Ni	1.70	1000	450–550
O_2	0.106	2176	327–1000
Zr	0.15	3172	350–750

REFERENCES TO SECTION 3

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4. PRODUCTION OF SODIUM, LEAD AND BISMUTH

4.1. SODIUM

Sodium is among the most abundant elements. Its content in the earth’s crust is about 2.5 wt.%. The most important sodium compound, produced in the amount of millions of tons, is NaCl, the content of NaCl in seawater being about 3%.

Sodium can be produced by high-temperature reaction from almost any its compound (NaCl, NaOH, Na_2CO_3 and Na_2S). The method of sodium production that has enjoyed the widest application is electrolysis of the molten salts, for example:

$$4NaOH \rightarrow 2Na + 2NaOH + H_2 + O_2.$$

Since water interacts with the one-half amount of sodium produced in this reaction, sodium yield cannot exceed 50% of theoretical value. In other electrolysis reactions, the yield of sodium could be even lower. Metallic sodium production by electrolysis of molten NaCl having melting point about 800°C is the most wide spread method. In order to reduce melting temperature such salts as $CaCl_2$, Na_2CO_3, etc. are used. For instance, mixture consisting of 40% NaCl and 60% $CaCl_2$ has melting temperature of 580°C, while mixture of 35.6% NaCl and 64.4% Na_2CO_3 melts at 600°C.

In electrolytic vat, the anode is usually made of graphite, the cathode being made of metal (Fe). Chlorine being by-product of this process is either discharged into atmosphere or accumulated for further using. Electric energy needed for sodium production is about 15 kW-h/kg Na.

Sodium produced by electrolysis contains 99.7 to 99.8% of basic metal and impurities (<0.1%K, <0.001% Fe, <0.15% Ca and others, such as Ba, Zr and Sn). It should be noted that the use of graphite electrodes causes sodium to be polluted with carbon. The worldwide sodium production data is given in Table 4.1 [4.1, 4.2].

TABLE 4.1. WORLD SODIUM PRODUCTION (thousand tonnes/year)

Years	1930	1952	1970	1990	2000 (prediction)
Quantity	25	140	270	354	340

4.2. LEAD

Lead content in the earth’s crust is about $4\cdot10^{-3\%}$. The most important lead minerals are: PbS (86.6% Pb), $3PbS\cdot Sb_2S_3$ (58.8% Pb), $PbCO_3$ (77.55% Pb), $2PbS\cdot Cu_2S\cdot Sb_2S_3$ (42.4% Pb) and $PbCl_2\cdot PbCO_3$ (76.0% Pb). Lead bearing ores such as PbS composition are the most abundant. In the oxidized (carbonate) ores lead exists in the form of $PbCO_3$ compound.

Only sulphide ores ($PbSO_4$, PbS) are reprocessed in the lead-zinc industry using two ways, namely: hydrometallurgy and pyrometallurgy. Preference is given to the pyrometallurgical method, based on lead extraction by reducing or smelting. The molten lead contains copper, antimony, arsenic and sulphur impurities. Moreover, it usually contains silver extracted by lead purification. In order to remove impurities from the ore, the re-smelting is performed providing access for air (pyrometallurgical method). Impurities are oxidized with the products floating up to the surface and dumped away gradually [4.3–4.5]. The world lead production data is given in Table 4.2 [4.2, 4.6].

TABLE 4.2. WORLD LEAD PRODUCTION (M TONNES)

Year	1900	1950	1960	1970	1980	1990
Quantity	0.87	1.75	2.4	3.6	4.15	3.9

Commercial-grade (C1) lead contains about 99.985% of basic metal. Among the main impurities are Bi, Ag, Cu, Zr, As, Sn, Sb and Fe. Bismuth is withdrawn using Ca, Mg and Sb. Such impurities as Ca and Mg are injected into the vat filled with lead, resulting in production of Bi_2CaMg_2 compound, which is then removed, residual content of Bi in lead being equal to 0.008–0.01%. Upon adding Ca+Mg+Sb mixture and removal of slag, 0.003–0.04% Bi content is left in lead. Ca, Mg, Sb and Zn impurities are withdrawn by $NaNO_3$ oxidation in the presence of NaOH.

Being highly toxic lead may cause poisoning. Its maximum permissible concentration (MPC) values are 0.003 mg/m^3 and 0.03 mg/L respectively in air and water. The main contributors to the environmental pollution are releases from metallurgical plants, sewage (discharged to the world’s ocean) and car engines. The value of annual lead release into the environment is presented in Table 4.3.

TABLE 4.3. AMOUNT OF ANNUAL LEAD RELEASE INTO ENVIRONMENT (THOUSANDS OF TONNES)

Source of release	Metallurgical plants	Car engines	Sewage
Amount	89	260	430–650

4.3. BISMUTH

Bismuth content in the earth's crust is about $2\cdot10^{-5\%}$. Bismuth deposits are few and far-between, bismuth recovering from the bismuth bearing ores being very complicated multistage process.

The major portion of bismuth is obtained by reprocessing polymetal copper ores and on the refining stage of lead production. Bismuth is extracted by adding Ca and Mg to molten lead. This results in formation of Bi_2CaMg_2 compound, which is considered as raw material for bismuth production.

Bismuth is applied in ferrous and non-ferrous metallurgy as admixture for stainless steel, pig iron and non-ferrous alloys. Alloys of Bi-Pb-Sn-Cd system are known for having 50–70°C and even lower melting temperature. The world bismuth production is given in Table 4.4 [4.2, 4.3, 4.8].

Market values of the aforementioned liquid metal coolants are summarized in Table 4.5.

The world-wide production of Bi is ~4000 t/a at a cost of ~$ 12 per kg, its explored reserves as of 1972 being ~160 000 tons (coolant inventory of 1GW(e) reactor is 15.000 tons). Respective data for lead is: 4×10^6 t/a and $\sim100\times10^6$ tons. Bismuth is expensive, its resources being limited. It is possible that its use could be confined to a limited number of reactors.

TABLE 4.4. WORLD BISMUTH PRODUCTION (tonnes/year)

Year	1959	1968	2000
Quantity	2355	3800 (except for Russia)	~ 5000 (forecast)

TABLE 4.5. MARKET VALUE OF LIQUID METAL COOLANTS (US$/tonnes)

Coolants	1960	1998
Commercial-grade sodium	360	~ 700
Reactor-grade sodium*)	2500	4000
Commercial-grade lead	260	550
Reactor-grade lead	1000	3000
Bismuth	5000	12 000

*) Stored in special tanks with argon cover gas.

REFERENCES TO SECTION 4

[4.1] M. SITTIG, "Sodium, its Production, Properties and Use", New York, 1959

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5. SODIUM AND LEAD TECHNOLOGIES

5.1. INTRODUCTION

The term "coolant technology" means a complex of methods and means assuring operation of facility under rated conditions and possible abnormal conditions. The main tasks related to coolant technology are as follows:

1. Development of coolant quality standards;
2. Analysis of impurities condition, their sources and accumulation rate in the circuit;
3. Analysis of corrosion and mass transfer;
4. Development of methods and equipment for keeping impurity content in the coolant within acceptable limits;
5. Analysis of different operating procedures and evaluation of related impurities input;
6. Control of coolant quality during operation.

Coolants produced by the industry (Na, Pb, etc.) do not always meet technological requirements. Therefore additional procedures are performed prior to filling circuit with the coolant in order to bring the coolant to required condition. Mounting and repair works are always required during facility life although resulting in pollution of the coolant. Another cause of changes in the coolant composition is corrosion, resulting in deterioration of mechanical properties of structural materials. Corrosion products are transported along the circuit, and depositions are formed that can affect hydrodynamics and heat transfer, and hence reliability of facility. All circumstances mentioned above lead to the necessity of permanent control over the impurities content and corrosion processes in the circuit.

The main elementary corrosion processes in the liquid metal are as follows:

1. Interaction of coolant with passive or oxide films-passivation or activation of materials.
2. Dissolving of steel components and their chemical interaction with non-metal impurities (oxygen, hydrogen, etc.).
3. Penetration of liquid metal into solid materials causing frontal and inter-granular corrosion.
4. Transport of structural material components along the circuit.

Interaction of Na, Pb and Fe with oxygen occurs by the following reaction:

$$n\,Na + k\,Me + m\,[O] \rightarrow Na_nMe_kO_m + .2\,Na + 1/2\,O_2 \rightarrow Na_2O$$

$$Pb + 1/2\,O_2 \rightarrow PbO$$

$$3Fe + 2O_2 \rightarrow Fe_3O_4$$

Direction of corrosion process is determined by thermodynamic properties of the system. The reactivity is determined by the change in isobaric-isothermal potential or Gibbs energy *(ΔG):*

$$\Delta G = RT \ln a, \quad (5.1)$$

where: R-thermodynamic constant, J/kg K; $a = \gamma N_i$-thermodynamic activity of i-th component, N_i-mole fraction of the component, γ — its activity factor. Component dissolving takes place, if $\Delta G < 0$, while crystallization occurs with $\Delta G > 0$.

In order to predict the direction of corrosion and mass transfer, it is essential to have data on thermodynamic properties of chemical compositions and steel components as a function of temperature. If the liquid metal is flowing at high velocity, the material is subject to erosion. Formation of the film (consisting of both steel and liquid metal coolant components) on the structural metal surface is another type of corrosion, since this is not protective film. Due to the difference in chemical activity between sodium and lead, technologies of these coolants are quite different, although some methods share a number of common features.

5.2. SODIUM TECHNOLOGY

5.2.1. Impurities sources

Typical composition of commercial-grade sodium determined by standard methods of analysis is given in Table 5.1, where 1 ppm=$10^{-4\%}$.

Among permanent contributors of impurities under normal operating conditions are corrosion related hydrogen from steam generators, tritium from the reactor core, oxygen and hydrogen from cover gas and corrosion products. Sources of impurities in liquid sodium determined in the BN-350 and BN-600 reactors under operating conditions are shown in Table 5.2 [5.1]. They have the following characteristics:

1. Initial impurity content due to oxygen absorption on the circuit surface is 3.4 g/m^2 (I circuit) and 2.2 g/m^2 (II circuit), that is in a good agreement with the experimental data (1.4–2.4 g/m^2). Total amount of oxygen absorbed is about 30 kg.

2. Impurities introduced during repair operation are Na_2O, NaOH and Na_2CO_3. Assessments have shown that during operation (20 years) about 200 kg of substances aforementioned is introduced [5.2.].

3. Impurities added during subassembly loading. Assuming 2 g/m^2 specific content, total amount is equal to about 200 kg.

4. Impurities caused by diffusion from other sources. Diffusion rate is evaluated to be about 50 g O_2/day. Total amount of impurities is about 720 kg.

TABLE 5.1. TYPICAL COMPOSITION OF COMMERCIAL-GRADE SODIUM

Components	Content (mass)
Total (Na)	99.9%
Metal (Na)	99.8%
Potassium (K)	200 ppm
Calcium (Ca)	250 ppm
Barium (Ba)	< 5 ppm
Iron (Fe)	< 20 ppm
Chlorine (Cl)	< 10 ppm

TABLE 5.2. SOURCES OF IMPURITIES IN LIQUID SODIUM

Sources	Oxygen	Water, kg/year	Hydrogen	Corrosion products from reactor, kg/year	Tritium from the core, g/hour
Cover gas	1 kg/year*	0.1–0.5	$(3\div6)\ 10^{-2}$ g/h**	20**	$6.3\cdot10^{-5}$*
Repair works	6 kg/year*	0.6	0.5–1 g/h**	60*	
Steel (Cr18Ni10Ti)	0.01 g/m^2	-	$4.4\cdot10^{-3}$ g/kg		
Steel (11/4 Cr2Mo)	0.01 g/m^2	-	$6.4\cdot10^{-3}$ g/kg		

Note: * — BN-600 reactor, ** — BN-350 reactor.

5.2.2. Purification of sodium and cover gas

At the present time, sodium purification technique with the use of cold traps based on decreasing solubility of the major part of impurities in sodium with temperature decrease has found the most wide practical application [5.3, 5.4]. In the cold trap sodium is cooled within the settling tank and the section upstream the filter. This results in reduction of oxygen and hydrogen concentrations respectively to 1 and 0.05 ppm. Cold trap is capable of retaining impurities in amount up to ~30% of its volume [5.5].

Carbon is confined in the cold trap as suspension, the purification process being 10 times longer than that for oxygen. In order to catch caesium, graphite based purification technique was developed [5.6, 5.7]. Upon 10 years of the BN-350 reactor operation capacity of the secondary traps was exhausted and their hydrogenation was carried out. The basic amount of sodium transformed to caustic phase was removed at 420°C. Thus, normal operation of traps was restored [5.3].

As regards the cover gas, it is necessary to purify source argon eliminating impermissible release to the environment of $^{133,\ 135}$Xe and $^{85,\ 87,\ 88}$Kr, short-term total activity of which in the BN-600 reactor reached $6.7\cdot10^9$ Bq/L value. This problem was solved using special filters and activated coal as absorbent [5.1, 5.3].

5.2.3. Control of impurities content in the coolant and cover gas

Control of the content of non-metal impurities in the coolant (O_2, H_2 and C) and cover gas (H_2O, CH_4 and N_2), as well as ^{90}Sr, ^{131}J, ^{137}Cs, ^{54}Mn and $^{58, 60}$Co in the primary circuit is most important.

Oxygen control. In order to make measurements of oxygen activity in sodium the electrochemical control technique based on galvanic cell has been mastered with sodium flowing over electrolytic pellet of thorium and yttrium sealed into the metal tube. Reference electrode is located inside the tube. E.m. F. generated depends on the temperature and oxygen concentration: e.m. F. = f (T, C_{O2}). Service life of such device is over 10^4 hours.

Hydrogen control. The necessity in hydrogen control was caused by the need for detection of water leak into sodium resulting in the following reaction:

$$Na + H_2O \rightarrow Na_2O + H_2.$$

Diffusion technique that has found the widest application is the method using metal membrane permeable to hydrogen in combination with different secondary devices (mass-spectrometer, magnetic discharge pump, etc.). So, the flow rate of hydrogen passing through the nickel membrane into vacuum cavity is measured by the system equipped with magnetic discharge pump, where gas ionization takes place. Automatic hydrogen detector is capable of detecting 10–30 g water leak to 100 t of secondary sodium. Description of other techniques can be found in [5.3].

Carbon control. Carbon needs to be controlled for evaluating carburization of structural materials because of possible impact on their mechanical properties. Diffusion and electro-chemical cells similar to those mentioned above are used for carbon control, with the salt mixtures (Na_2CO_3-Li_2CO_3 and CaC_2-LiCl) used as electrolytes.

Control of initial temperature of impurity crystallization (plugging indicator). Operation of plugging indicator is based on impurity deposition from supersaturated solution in the narrow gaps. Super-saturation is achieved by decreasing temperature of the coolant flowing through the indicator. As the temperature becomes lower than the saturation point, the flow rate through the indicator decreases, this being registered by the indicator. Actually, indicator measures the temperature at which rate of impurity deposition or solubility is sufficient to detect change in the flow rate. Presence of different impurities in the coolant makes it possible to detect several "plugging temperature" values. It should be noted that saturation temperature is determined within the accuracy of 3°C.

Radioactive impurity control. Prompt control of radioactive impurities in the primary sodium is made using three methods:

- measurement of γ-radiation from piping 10–12 days after reactor shutdown;
- measurement of activity of fission and corrosion products on special-purpose bypass section 10 days after its disconnection from the primary circuit;
- measurement of concentration of selected products (Cs, I) in graphite compound.

5.2.4. Corrosion processes in sodium

By now, comprehensive studies have been performed and reliable industrial experience has been gained on material corrosion in sodium. Corrosion intensity in sodium is significantly lower than that in water or lead-based coolants [5.9]. In sodium, as well as in the other liquid metals, corrosion rate depends on many factors (temperature level, coolant velocity, impurity content, temperature difference, time, etc.). When evaluating corrosion rate, the major part of researchers took into account only the most contributing factors. Empirical equations for corrosion rate were most commonly derived for 316 steel at the coolant velocity of > 4 m/s and oxygen content of ≤ 10 ppm. The most reliable results were obtained in [5.10, 5.11] for corrosion rate K, mg/cm^2 h, that can be expressed as follows:

$$K = 0.61(C_o)^{1.5} \exp\left(-\frac{18000}{RT}\right). \qquad (5.2)$$

It was found that chromium alloyed and austenitic steels have high corrosion resistance. The equivalent corrosion rate of two steels such as HT-9 and Fe9Cr1Mo was measured after their exposure during 4000 hours at 600–650°C temperature, 6 m/s flow velocity and 1 ppm oxygen content. The results of measurements are given in Table 5.3 [5.12, 5.13, 5.14].

TABLE 5.3. EQUIVALENT CORROSION RATE OF AUSTENITIC AND CHROMIUM STEELS

Materials	Equivalent corrosion rate, μm/year	
	600°C	650°C
HT-9	1.15	3.1
Fe9Cr1Mo	0.7	2.3

The operational experience of a number of LMFRs during long time (20–30 years) has shown, that sodium is practically non-corrosive with respect to stainless steel, the content of impurities, mainly oxygen and carbon, being held at acceptable low level by the cold traps.

5.3. LEAD TECHNOLOGY

5.3.1. Basic issues of technology

Basic technological challenge of using lead as reactor coolant is assurance of such quality of the coolant (as well as that of the surfaces contacting coolant), that the following conditions are realized:

- sufficient corrosion resistance of structural materials;
- stable hydrodynamics and heat transfer during life time.

Presence of impurities in lead coolant is injurious for at least two reasons, namely:

1) possible partial or full plugging of the coolant flow cross section area, that disturbs hydrodynamics and hence heat transfer;
2) deposits formed on heat transfer surfaces (especially on the fuel elements of the reactor core causing rise of cladding temperature).

5.3.2. Impurities contributors

The following impurities are present at the initial stage of circuit filling:

- residual oxygen, remaining after evacuation, and water vapours;
- gas adsorbed on the inner surfaces;
- steel corrosion products;
- casual impurities (chip, welding hail, etc.).

Under operating conditions, the basic factors causing increase of impurities content in the lead circuit are:

- structural material corrosion;
- erosion and abrasion of materials;
- penetration of grease from pump seals and bearings;
- cover gas entrainment by the coolant;
- admixtures to the coolant aimed at forming protective films.

Impurities can be located in such parts of circuit as cover gas plenum and stagnant sections, as well as on the free surface of molten lead (since density of practically all impurities is lower than that of lead) and structures (as deposits). During facility operation the impurities are transported along the circuit. The methods of impurity control in lead and cover gas are shown in Table 5.4.

TABLE 5.4. METHODS OF IMPURITY CONTROL IN LEAD AND COVER GAS

Impurities	Sources	Methods of control
Water vapour in cover gas	Steam generator leak	1. Condensation of vapour from cover gas 2. Permanent chromatography control of cover gas
Hydrogen in cover gas	Cleaning of coolant and circuit from impurities	Measurement of hydrogen content in cover gas: • thermal conductivity method • chromatography
Oxygen in lead	Maintaining required oxygen content	Galvanic cell (measurement of e.m. F. induced in solid electrolyte with ion conductivity)

5.3.3. Cleaning from slag

Oxide-based slag (PbO) can be removed from the coolant by settling and reducing with hydrogen. Slag originating dispersed impurities caused by lead interaction with structural materials (Fe, Cr and Ni) are removed by mechanical filter or by settling. Oil and pyrolysis products can be removed using organic solvents or water vapour injected into the circuit.

5.3.4. Corrosion processes in lead [5.15–5.20].

The development of corrosion resistant structural materials is apparently the main problem in lead cooled reactor technology. Lead exhibits strong erosion-corrosion effect on structural materials, such as material dissolving, embrittlement, thermal transport of mass and inter-granular penetration of lead.

The most resistant to lead are refractory metals followed by chromium steels and austenitic steels being less resistant because of high solubility of incorporated nickel. Stabilization of austenitic steels by Ti, Nb and Mo enhances their resistance to lead.

The main type of corrosion damages in liquid Pb, Bi and Pb-Bi is the dissolution of structural materials (steels) and their components in these coolants. The kinetics of dissolution processes can be of different nature. For example, in some cases the dissolution is localized on boundaries of grain, causing interstructure infiltration of liquid metal (Pb, Pb-Bi) into steel.

The basic kind of corrosion damage, which is the most dangerous for structural materials both in Pb-Bi and in Pb coolants, is local corrosion of materials appearing as the separate corrosion-erosion centres (“pittings”). Local through corrosion damages of structural elements may appear at temperatures over 550^0C after holding for some hundred hours under the following conditions: unbalance of alloying elements and impurities in steel, poor quality of metal, absence of coolant quality control and non-optimal coolant flow regimes. The typical corrosion rate in such cases is estimated as 2.55 mm/year.

The principle solutions ensuring high corrosion resistance of structural materials in heavy liquid metal coolant were found using oxygen dissolved in the coolant. It has been shown as a result of long-term studies that this corrosion resistance essentially depends on concentration of dissolved oxygen.

Upon reaching certain level of concentration of dissolved oxygen corrosion processes is stopped due to protective oxide film formed on the steel surface. At high temperatures an indispensable condition of corrosion inhibition is presence of silicon in steel as additional alloying element. The silicon content in steels is varied within 1–3.5% range depending on steel type.

Oxide films formed on the steel surface prevent it from interaction with liquid lead. Since breakdown of oxide films is possible during operation, precautions must be taken for resuming and maintaining their thickness and density.

Thus, steel corrosion in molten lead can be significantly slowed down by the oxide film formed on the steel surface. The main technological problem is maintaining such oxygen content in the coolant which, on the one hand, would provide stability of oxide film (Fe_2O_4) on the steel surfaces, but, on the other hand, would preclude generation of lead oxide (PbO) in the coolant, that could result in the circuit slagging.

There are some ranges of content of oxygen dissolved in lead meeting these two conditions, for instance ($\sim 5 \cdot 10^{-6}$–10^{-3} wt%) range. Oxygen content in lead can be controlled by injecting gaseous oxygen or dissolving solid PbO.

Required oxygen content in lead can be maintained in two ways: (a) bubbling of argon, hydrogen and water vapour mixture or gaseous oxygen through molten lead; (b) lead oxide filling through which molten lead is pumped.

In order to change oxygen content and remove surplus PbO, reactions with water vapour or hydrogen can be used. To determine oxygen content in molten lead (similarly to Pb-Bi technology development) galvanic cell can be used.

The problem of hyperthermal corrosion resistance of structural materials was got over by development of preliminary protective coatings for the working steel surfaces. In particular, the most important structural units of circuit, e.g. fuel rod claddings and steam generator tubes, are covered by these coatings at the final stage of their manufacture. Additional barriers are also formed directly on the inner surfaces of liquid metal circuit under effect of the coolant in the early stage of the reactor operation.

The best results of using technology of preliminary oxidation of circuit components were achieved by application of media with low partial pressure of oxygen, namely: Pb-Bi-O, H_2O+H_2 and CO_2. These methods, first of all, make it possible to avoid critical kinetic stage of preliminary passivation of uncoated surfaces of steel structures of the circuit. Moreover, they prove to extend the range of permissible decrease of oxygen concentration in the coolant.

Therefore, the basic factors ensuring high corrosion-erosion resistance of structural materials in heavy liquid metal coolant (Pb, Pb-Bi) are as follows:

- application of silicon alloyed steels;
- passivation by oxygen using special regime of coolant;
- using additional corrosion barriers such as oxide films formed on working surfaces of circuit components under reactor start-up conditions.

In reaction of PbO reduction, water vapours are efficiently removed from the circuit. Small amount of moisture acts as diluted oxidizer preventing from achieving reduction conditions for oxide films on the steel surface.

Parameters of all these processes have to be developed with necessary control of hydrogen content in cover gas and oxygen activity in liquid lead.

Preliminary studies have shown that principal possibility exists to develop the technology mentioned, but it should be noted that conditions of experiments did not correspond to those of real operation.

It was found in the tests performed on the experimental and industrial facilities, that corrosion rate for chromium steels in PbBi alloy is 6–60 mg/m^2h at 450–500^oC. It can vary with temperature, coolant velocity, oxygen content and other parameters. In lead, this value is about 0.026 mg/m^2h at 600^oC with no mass transfer [5.20].

5.3.5. Comparison of PbBi technology for submarine and Pb technology for NPP

There are problems of applicability of data gained on lead-bismuth cooled submarine facilities to lead cooled nuclear power plants (NPP), which are caused by the following factors:

1. The inner surface area of the primary circuit contacting liquid metal in fast reactor NPP exceeds by a factor of 10 that of submarine facility (see Table 5.5).

TABLE 5.5. INNER SURFACE AREA OF THE PRIMARY CIRCUIT IN CONTACT WITH LIQUID METAL

Coolant	Type of plant	Electric power, MW	Surface area, m^2
PbBi	Submarine facilities	~30	~2000
Na	BN-600	600	21600
Na	Super Phenix	1240	24600
Na	EFR	1580	22800

2. NPP operating period is about 6000–7000 h/year that is at least one order greater than that of submarines.

3. The average temperature of lead circuit is 150–200°C higher as compared with that of PbBi circuit.

4. Some other effects causing corrosion take place in the NPP, such as iron diffusion through passivated protective oxide film, high steam generator pressure, etc.

Therefore, it can be concluded that direct transfer of already developed PbBi technology to the stationary NPP operating conditions is impossible, and additional expensive studies are required for many years.

Evaluations have shown that the increase of corrosion products amount in the circuit could be as high as hundreds of kg per year. Thus, it is obviously needed to develop special-purpose mass transfer equipment designed for catching corrosion products with permanent passivation of steel surface in contact with liquid metal.

CONCLUSIONS

1. Sodium technology developed up to the industrial scale for fast reactors has demonstrated that existing methods and control means are capable of solving the majority of the problems concerning future nuclear power. The possibility of continuous improvement of the technology already brought into commercial use ensures that such work will not require significant additional expenses.

2. Scientific and engineering approaches to be used in heavy metal coolant technology are essentially clear, but their implementation calls for time and financial resources yet difficult to estimate.

3. Direct application of data gained on lead-bismuth alloy to lead coolant is doubtful, and it should be noted, that further long-term studies are required.

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6. THERMOHYDRAULICS OF REACTOR CORE AND SOME ASPECTS OF FAST REACTOR ENGINEERING

6.1. INTRODUCTION

High power rating and temperature impose special requirements to fast reactor thermohydraulics. Analysis of thermohydraulic issues assumes reliable hydraulic and heat transfer relationships to be worked out. As a result, distributions of the coolant flow rate, its velocity, and finally, fuel and core structure temperatures would be obtained. All mentioned parameters are required for evaluation of core integrity and mechanical behaviour.

The fuel subassembly life is affected by peak cladding temperature and temperature distributions in the coolant, fuel and subassembly structures. These temperature distributions should be such that the fuel burnup goals can be met without seriously impairing core life and performance. For constraining temperature levels by design measures, it is important that the fuel subassembly temperature distributions be accurately predicted and the governing thermal-hydraulic phenomena be understood. An example of temperature affected performance is that the estimate made for existing LMFR designs indicates that fuel burnup can be increased by about 10% (in MW·^{D}/t) per one Kelvin degree reduction of hot spot cladding temperature.

Method developed for predicting temperature distributions should be capable of accounting for the effects of geometrical differences and the wide range of operating conditions of the core and blanket subassemblies, as well as control rods of LMFRs.

For prediction of subassembly coolant flow rate and temperature distributions a wide range of coolant flow and thermal convection regimes must be considered including laminar and turbulent flow; natural, forced and mixed (forced + natural) convection; and steady state and transient reactor conditions.

Thermohydraulic performance prediction for the core subassemblies begins with calculation of the subassembly flow rate necessary for determining the limiting design parameter (e.g. lifetime or maximum local cladding midwall temperature) in the limiting subassembly of each flow orificing zone.

Total calculated subassembly flow rate, as well as design data on core neutronics are input to subchannel analysis codes that predict coolant flow and temperature distribution in the subchannels of the core subassemblies. The peripheral subchannel temperatures and flow rates used for duct temperature prediction, as well as peak subchannel coolant temperature used for prediction of hot spot temperature of the hottest fuel elements are of particular interest.

For detailed cladding and coolant temperature information the distributed parameter codes are used, which calculate two-dimensional temperature fields in the coolant, cladding and fuel.

Within this Report it is impossible to discuss all details and strategies of LMFR thermal hydraulic design. Therefore, only the most important aspects will be treated in the following subsections.

6.2. BASIC ASSESSMENTS

LMFR core orificing is important because fuel lifetime strongly depends on maximum cladding temperature, which is determined primarily by the core coolant temperature rise (ΔT) and inlet temperature.

Subchannel analysis is commonly used for thermal hydraulic analysis of single fuel subassemblies. Bulk average values characterize the fluid dynamics and thermal coolant conditions in each subchannel. Thermal and hydraulic interactions between subchannels are taken into account.

All subchannel codes are used to solve mass, momentum and energy transport equations for turbulent fluid flow. Since the governing equations are very complicated, in each code simplifying assumptions are made to expedite solutions. Empirical input by either experimental data or analytical results is required for the codes to obtain reliable computer results. The correlations strongly depend on the geometry considered, the spacer concept (grids or wire-wraps), etc.

Experimental investigations, therefore, are always necessary on the actual geometry for an accurate design. However, some correlations, which are widely used, are mentioned in the following subsections.

Qualitative comparison of some thermohydraulic characteristics of sodium and lead coolants is presented in [6.1] with BREST reactor design taken as a reference [6.2]. Results of optional assessments performed by the author [6.2] are in a good agreement with the data presented in [6.1]. In the analysis, the following parameters are assumed identical: reactor power, average temperature in the circuit, cross section for coolant flow, coolant temperature rise in the reactor, primary circuit pressure drop and coolant volume.

Analysis has shown the following ratios:

Coolant velocity in the core	$W_{Na}/W_{Pb}=3.5–6.0$
Core pressure drop	$\Delta P_{Na}/\Delta P_{Pb}=0.16–0.2$
Pumping power	$N_{Na}/N_{Pb}=0.23–0.3$
Heat accumulated in the primary circuit prior to achieving boiling temperature $T_{Na} = 880^{o}C$, $T_{Pb} = 1745^{o}C$	$Q_{Na}/Q_{Pb}=0.7–0.4$

The latter assessment has no special sense for just logical reason, namely that lead boiling temperature cannot be achieved in LCFR, in contrast to that of sodium in LMFR, because reactor vessel would be necessarily broken before lead boiling onset (melting temperature of Cr18Ni10Ti, Cr15Ni11MoNb and other steels is about $1500^{o}C$). Because of this, there is no reason for considering high boiling temperature of lead as an advantage.

Thus, in terms of thermohydraulics, sodium cooled reactor parameters such as pressure drop (5–6 times lower) and required pumping power (3–4 times lower) are preferable.

Thermohydraulic parameters of BREST type plant should follow the following four requirements:

- Maximum permissible coolant velocity (based on corrosion and erosion conditions) is ~ 2.5 m/s;
- Low primary circuit pressure drop (under accident conditions, 15% flow rate should be provided);
- Temperature of the fuel element ferritic steel cladding is ~923 K ($650^{o}C$);
- Minimum release of gaseous fission products (maximum fuel temperature should not exceed $1000^{o}C$).

6.3. HYDRODYNAMICS

As a rule, hydrodynamics of Na and Pb is quite similar, since both sodium and lead are Newton liquids. Therefore the data available on hydraulic resistance of the reactor core incorporating pin bundles can be used for adequate evaluation of hydraulic friction loss in the core [6.3–6.6].

Data on pressure drop for pin bundles were determined by summarizing a great number of experimental and analytical results. The following relationships have been proposed:

$$\Lambda_{tr}=0.57+0.18(x-1)+0.53[1-\exp(-a)], \qquad (6.1)$$

where $a=0.58\{1-\exp[-70(x-1)]\} + 9.2(x-1)$, and

$$\Lambda_{sq}=0.59 + 0.19(x-1) + 0.52\{1-\exp[10(x-1)]\}, \qquad (6.2)$$

which is valid for $1<x<10$; $2 \cdot 10^4<Re<2 \cdot 10^5$.

In this case, Λ is ratio between friction factor of a bundle (λ) and that of round tube (λ_o), subscripts "*tr*" and "*sq*" refer to triangular and square arrays respectively, and $x=s/d$ is relative pitch of the pin bundle.

Equivalent hydraulic diameter values are calculated as follows:

$$d_{tr} = d\left(\frac{2\sqrt{3}}{\pi}x^2 - 1\right),\ d_{sq} = d\left(\frac{4}{\pi}x^2 - 1\right)$$

Instead of equation (6.1), more simple relationship can be used:

$$\Lambda_{tr} = 0.21/Re^{0.25}[1+(x-1)^{0.32}]$$

for $1<x<1.5$; $6 \cdot 10^3<Re<2 \cdot 10^5$.

Equation (6.2) is based mainly on the results of analysis and limited experimental data. For more specific consideration, additional studies and new results are required, for example those obtained by other authors.

Local pressure drop in the subassemblies (for instance that due to spacing elements) can make significant contribution to the total pressure drop [6.7, 6.8]. It is unlikely, that spacing elements used in the fuel subassemblies of sodium cooled reactors (wire, etc.) could be applied for lead cooled reactors. Therefore, if the new spacers are designed, the available data cannot be applied. This would require special-purpose experiments not only for determining hydrodynamics, but to test structure for vibration and attrition.

There is one point which is most important for the development of lead cooled reactors. When carrying out studies on the models of lead-bismuth cooled reactor flow path (including full-scale model), it was found that in parallel with the toroidal eddies appearing at the core diagrid inlet, sufficiently stable horizontal eddies may arise near the bottom of the diagrid. These eddies result in reduction of local inlet flow rates and flow stability, thus facilitating local concentration of particles suspended in the core diagrid area and, hence, offering risk of their penetration into the core.

6.4. HEAT TRANSFER

6.4.1. Basic relationships

In general, comprehensive studies have been made on heat transfer of different liquid metals (Hg, Na, NaK, PbBi, Li, etc.). Unless special measures are taken for liquid metal purification, heat transfer coefficients follow the relationship (for tubes):

$$Nu = 3.3 + 0.014\, Pe^{0.8}. \tag{6.4}$$

Careful purification makes heat removal more intensive, as it is described in [6.12–6.14]:

$$Nu = 5.0 + 0.025\, Pe^{0.8}. \tag{6.5.}$$

Low heat transfer coefficient (6.4) is caused by thermal contact resistance at the wall-liquid boundary. Thermal resistance is influenced by surface wetting with coolant, oxide films formed on metal surface, as well as deposits of oxides and other impurities. Thermal contact resistance can hardly be evaluated, only possible upper limit can be specified [6.15, 6.16]:

$$\left(R_c \frac{\lambda}{d}\right)_{\max} = 200 \cdot \mathrm{Re}^{-0.75}. \tag{6.6}$$

Fig. 6.1 shows the data available and the relationship presented.

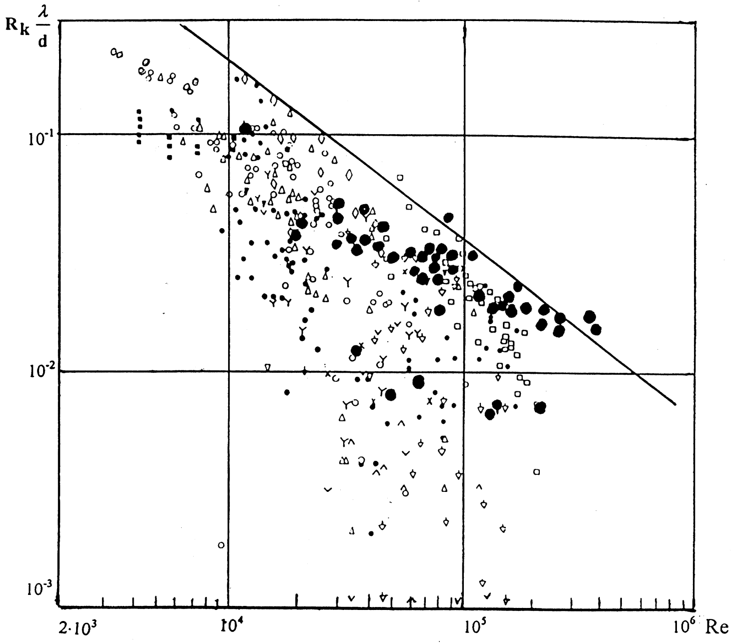


FIG. 6.1. Dimensionless thermal contact resistance as Re function [6.15, 6.16]:
● — PbBi; other sympols: Li, Na, NaK, Hg
---- — equation 6.6.

Thus, common conclusion cannot be reached on value of thermal contact resistance. As for lead, this question remains open and the data must be checked.

Normal purification of liquid metal leads to almost complete elimination of thermal contact resistance due to depositions.

Typical thickness of passivation film in PbBi alloy is about 10 μm and 20 μm respectively for austenitic and perlitic steel, though scattering within the range from 10 μm to 100 μm. The film thermal conductivity is about 5 W/(mK). There is a good reason to believe that the film is not continuous because of its saturation with liquid metal. Thus, thermal resistance is rather low and temperature difference across the film is about 5K.

It is clear that the coolant must be purified. While this is not a problem for sodium, in case of lead one should bear in mind presence of permanent oxides in the circuit taking into consideration corrosion activity of lead and necessity of protective oxide films formation on the metal surface. Comparison of heat transfer in tubes with sodium and lead-bismuth described by (6.1) and (6.2) equations is shown in the Table 6.1.

TABLE 6.1. COMPARISON OF HEAT TRANSFER COFFICIENT FOR DIFFERENT COOLANTS (α – w/(M^2k), T = 400^Oc, D = 10 mm)

Na

Re	Pe	Nu_1	Nu_2	α_1	α_2
20000	90	3.512	5.915	25018	42131
30000	135	3.709	6.265	26416	44628
50000	225	4.066	6.904	28964	49178
100000	450	4.856	8.315	34593	59229

Pb

Pe	Nu_1	Nu_2	α_1	α_2
348	4.511	7.699	7723	13181
522	5.091	8.733	8715	14951
870	6.146	10.618	10522	18177
1740	8.477	14.781	14513	25305

PbBi

Pe	Nu_1	Nu_2	α_1	α_2
294	4.321	7.358	6135.4	10449
441	4.827	8.262	6854.0	11732
735	5.749	9.909	8163.4	14070
1470	7.786	13.547	11056	19236

6.4.2. Pin bundle heat transfer

Lead cooled fast reactor design assumes the subassembly with fuel pins arranged in square lattice with large pitch-to-diameter ratio (s/d ~ 1.4–1.5). In LMFR a more tight arrangement of fuel pins is adopted (s/d ~ 1.1–1.18):

Reactors	BR-10	BOR-60	BN-350	BN-600	BN-800	Phenix	Super Phenix
x=s/d	1.10	1.10	1.16	1.16	1.17	1.178	1.18

Both experimental and analytical studies of heat transfer in Hg, PbBi, NaK and Na coolants have been carried out. The following relationships for evaluation of heat transfer in triangular lattice bundle were obtained experimentally:

1. V.M. Borishanski, et al (triangular arrays) [6.20]:

$$Nu1 = 24.151 \ \lg[-8.12 + 12.76x - 3.65x^2] + 0.0174[1 - \exp(-6 \ (x-1))](Pe - 200)^{0.9}$$

2. H. Graber, (triangular arrays) [6.21]:

$$Nu2 = 0.25 + 6.2x + [0.32x - 0.07]Pe^{(0.8-0.024x)}$$

3. H. Calamai, et al [6.23]:

$$Nu3 = 4 + 0.16x^5 + 0.33x^{3.8}(Pe/100)^{0.86}$$

4. A.V. Zhukov (rod bundles with spacers, square arrays) [6.22]:

$$Nu4 = 7.55x - 14x^{-5} + 0.09Pe^{(0.64+0.264x)}$$

A.V. Zhukov (rod bundles without spacers, square arrays) [6.22]:

$$Nu4 = 7.55x - 14x^{-5} + 0.07Pe^{(0.64+0.264x)}$$

It has been demonstrated that Nusselt number for square lattice is lower than that for triangular lattice bundles [6.17, 6.18]. This is illustrated by the data in Fig. 6.2.

In Table 6.2 and Fig. 6.3 relationships *Nu=f(Pe)* taken for triangular and square lattice with fixed pitch-to-diameter ratio (*x*=1.4) are compared.

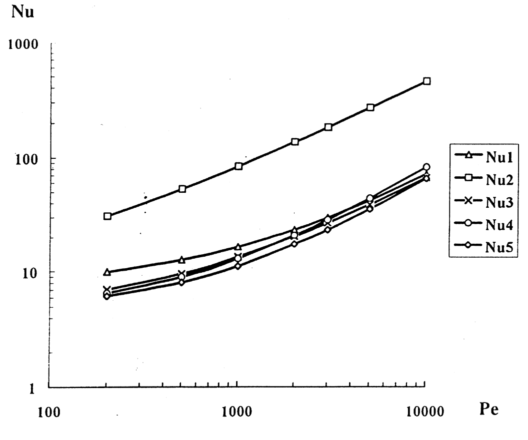


FIG. 6.2. Correlation for liquid metal heat transfer in rod bundles (s/d = 1.4).

TABLE 6.2. COMPARISON OF HEAT TRANSFER IN ROD BUNDLES WITH TRIANGULAR (TR) AND SQUARE (SQ) ARRAYS, X=1.4

Pe	100	200	300	500	1000	2000	3000	5000	10000
Nu(tr)	11.26	11.98	12.64	13.86	16.61	21.46	25.90	34.08	52.44
Nu(sq)	8.62	9.25	9.89	11.24	14.25	20.4	26.50	38.61	68.60
Nu(sq)/Nu(tr)	0.766	0.772	0.782	0.804	0.858	0.950	1.023	1.133	1.308

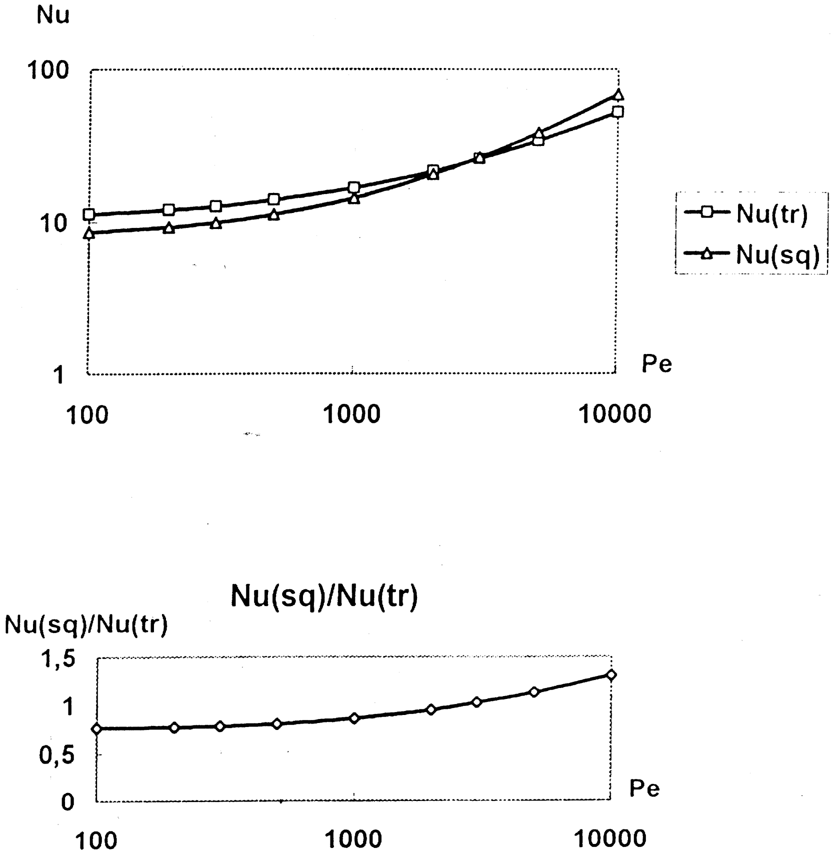


FIG. 6.3. heat transfer in rod bundles with triangular and square arrays.

Heat transfer in square bundles is roughly estimated, so special experiments are needed for verification. By now there are no convincing methods of extrapolation of the data on temperature non-uniformity in the area around fuel pin to the square grid bundle. As a first approximation, it can be assumed that values increase in the same way as in case of laminar flow. Unfortunately, this cannot be adopted for the peripheral pins where temperature pattern non-uniformity is significant. Therefore, additional experimental studies are needed.

Available experimental results on temperature distribution in plugged SA were analyzed to determine the main characteristics of velocity and temperature profiles of single-phase flow, namely: recirculation zone length, coolant flow distribution in SA, coolant temperature rise in the wake, etc. At the same time, it is clear that data currently available provide only rough estimations of SA thermohydraulic characteristics in case of blockage as a function of Reynolds number and blocked flow area.

In-pile and out-of-pile test results show that boiling occurring behind the blockage does not lead to rapid propagation of failure in the fuel pin bundles, while the gas release taking place as a result of pin failure behind the blockage may cause more pin damages within the SA. Studies of various factors influencing temperature profile (flow rate, power rating, Reynolds and Peclet numbers, blocked fraction of flow cross section area) call for carrying out additional comprehensive experimental and analytical studies. Special attention should be given to studies of thermohydraulics of SA with porous heat generating blockages, initial dry-out conditions with boiling occurring behind the blockage and SA cooling limits.

As for local thermohydraulic characteristics, such as hot spot factors, temperature non-uniformity in the irregular arrangement of pins etc., of critical importance are the processes of inter-channel exchange. Comprehensive studies of inter-channel heat and mass transfer in the bundles of smooth pins with ribs or wires provided a basis for development of calculation techniques.

Mixing factors in case of wire-to-pin contact in the bundle of wire spaced fuel elements turned out to be higher than those for wire-to-wire contact. Counter-direction wires providing lateral coolant flows would be most effective.

Issue of spacing elements development for lead cooled SA is rather complicated because of high dynamic pressure head, possible vibrations, etc. This is the subject of further detailed studies. A delusion still exists that rather high thermal conductivity would smooth temperature non-uniformity. It is only true for stagnant liquid metals that are equivalent to the solid body. Experiments and evaluations show the necessity of considering conjugate tasks of heat removal from fuel elements, i.e. taking into account properties of fuel elements.

As it was demonstrated by experimental studies, considerable temperature fluctuations take place in both coolant and channel walls. Preliminary estimations showed that such pulsation would not impact structural strength. However, this does not relate to the situation when two flows of different temperature are mixed. In this case thermal stress can result in structure failure, that happened at Phenix IHX.

The problem cannot be resolved without implementing a comprehensive study program, including analysis of thermal stripping, experimental studies on computer code verification, development of codes and techniques for measurements of the wall surface temperature fluctuations, etc.

6.5. FAST REACTOR ENGINEERING

6.5.1. Sodium-water interaction

Because of sodium radioactivity in the primary circuit and the possibility of sodium-water interaction in the steam generators, LMFRs are equipped with non-radioactive intermediate (secondary) sodium circuit. Intermediate heat exchangers thus constitute an internal containment barrier. In case of failure of water filled pressurized steam generator tube, water is injected at high pressure into sodium, and violent sodium-water interaction occurs. This sodium-water interaction would give rise to peak pressures that must be accommodated by straining the steam generator shell and the pipelines in the secondary circuit. Moreover, if the pressure in the system becomes too high, a special rupture discs in sodium and/or gas sections will be broken, thereby relieving the pressure and ejecting sodium hydroxide, sodium oxide and hydrogen into a special collector vessel. This pressure relief system is very important, since it protects intermediate heat exchanger from damage. The sodium-water interaction is immediately detected by instrumentation and the plant would be shut down if a large leak occurred. The basic physical processes are discussed below.

Sodium-water steam generator design features. One of the principal problems in the development of the sodium-water steam generator (SG) design is the choice of the tube bundle characteristics.

Specific requirements are also given to sodium-water SG structural materials. Corrosion-erosion resistance of materials contacting with sodium-water reaction products becomes of great importance. From this point of view, high-nickel alloys (30–40% Ni) have the best characteristics; chromium-nickel austenitic steels of X18H9 type being somewhat inferior to them. Perlitic steels, such as $2^1/_4$Cr1Mo and chromium steels with 9–12% Cr content are most susceptible to corrosion-erosion failure caused by reaction products. It should also be taken into account that perlitic steels are low-cost and relatively workable but have low corrosion resistance and are prone to pitting on the steam-water side.

Besides, there are certain limitations on temperature conditions for some steels (for example, stability of mechanical properties of $2^1/_4$Cr1Mo steel can only be guaranteed at temperature not exceeding 520°C). It should be also noted that practically all of the above steels have adequate corrosion resistance in pure sodium and all corrosion problems take place on the steam-water side. So it is seen that choosing tube material is rather complicated problem.

Taking into account various properties of steels and contradictory requirements imposed to them, the SG design features, such as unit heat capacity, tube bundle characteristics, maintainability, etc. sometimes play decisive role in the choice of structural materials. To date, there are examples of implementation of all the above steels.

Speaking about design features, small tube-to-tube distance in the tube bundle should be mentioned, since micro defect present in one tube would result in rapid failures of the adjacent tubes.

Physical phenomena of sodium-water reaction. Sodium-water chemical interaction proceeds in two stages: at the first stage the reaction proceeds at a high rate with release of gaseous hydrogen and heat:

$$Na + H_2O = NaOH + 1/2\ H_2 + 140,\ kJ/mole. \qquad (6.7)$$

This reaction is practically irreversible because of a very high equilibrium pressure of hydrogen.

At the second stage chemical interaction takes place between the products of the first stage of the reaction and excess sodium (excess sodium is a characteristic of water leak in the steam generator):

$$2Na+NaOH = Na_2O + NaH \qquad (6.8)$$

$$Na + 1/2\ H_2 = NaH \qquad (6.9)$$

The total reaction is described by the following equation:

$$4Na + H_2O = 2NaH + Na_2O \qquad (6.10)$$

Thus, sodium-water interaction is rather complicated multistage process including successive reactions with sodium hydroxide and hydrogen formation followed by their interaction with sodium. The final concentration of water-sodium interaction products is determined by thermodynamic equilibrium conditions and the time of reaching equilibrium state in the course of the reaction.

Small water leaks into sodium occur in the tubes with formation of chemical reaction products jet due to coolant interaction with water. The rate of corrosion-erosion damage of the adjacent tube material by reaction products formed in the jet depends, to a considerable degree, on the tube-to-tube gap value.

When choosing tube-to-tube gap value, the designer should be governed, on the one hand, by the limitations on the overall bundle dimensions, and, on the other hand, he should take into account the consequences of accidental contact of coolants caused by small water-into-sodium leaks. Practice has shown that for the single module design of small heat capacity, for which repair problems can be solved by simple replacement of failed module by the new one, optimum tube-to-tube gap value is within 12–15 mm range. For larger capacity SG using corrosion-resistant steels (for instance, Incalloy 800), tube-to-tube distance is sometimes increased up to 30–40 mm.

The second design feature comes from a desire to ensure the integrity of the steam generator vessel and sodium circuit under conditions of large water-into-sodium leaks (appreciable tube failures). As experimental studies have shown, an efficient means to reduce the rate of hydrodynamic effects in case of contact of large amounts of sodium and water is the use of expansion gas volumes. For the designs of large unit heat capacity the expansion (damping) gas volume is provided, as a rule, within the SG vessel.

It should be noted that the problem of sodium circuit hydraulic resistance is of vital importance due to some features of accident processes. Accidental pressure in the sodium volume in the vicinity of large water leak into sodium depends not only on the leak rate but also on the hydraulic "compliance" of the sodium circuit. The experience shows that, in view of the above circumstances, sodium circuit pressure drop within the boundaries of "SG-expansion gas volume inlet" section should not exceed 0.3 MPa value.

In some experimental and prototype NPPs with sodium-cooled fast reactors water/steam-sodium contacts took place in SG. The requirements to sodium-water steam generator from the standpoint of its tightness should be much higher than those to any other heat exchanger. When considering specific problems, on the basis of experience obtained on some NPPs the following conclusions can be drawn:

1. Operating experience of "Enrico Fermi" NPP SG (the first SG with single wall separation of sodium and water) has shown that tubes vibration led to their very rapid failures and, as a result, to large leaks. Insufficient attention was given to design optimization of some units and systems. It was also concluded that the choice of tube wall thickness of 1 mm

proved to be inappropriate solution and it cannot be adopted for the sodium-water steam generators.

2. As it is known, the cause of a number of large and small water leaks into sodium in the BN-350 NPP steam generators in 1972–1975 was their poor quality of fabrication. Nevertheless, the experience gained from incidents in the BN-350 NPP steam generators has also some positive results. First (and this is most important), it has been shown that in principle an ingress of large amount of water into sodium (in one of the incidents, about 800 kg of water got into the sodium circuit) does not lead to catastrophic consequences. Therefore, integration of sodium and water within one plant is quite justified from the engineering point of view. Second, steam generators and sodium circuits, even under conditions of substantial water-into-sodium leaks, remain repairable. Third, steam generator safety systems, even being under severe accident conditions, are capable of ensuring plant safety.

3. Water-into-sodium leaks that occurred in the steam generators of the BN-350 and other reactors were of much less scale than those occurred on the PFR NPP. However, these leaks have also confirmed the main conclusions drawn on the basis of the BN-350 and PFR NPP experience from the viewpoint of emergency conditions caused by chemical activity of sodium to water.

4. Operating experience of the BN-600 NPP steam generators has revealed the necessity of more careful choice of materials and refinement of fabrication techniques (in particular, of weld joints). However the main conclusion is that sodium-water steam generator design can be developed that would be practically fully capable of eliminating impact of steam generator leaks on the operation of the plant as a whole. This can be sectional-modular design, with single modules of the evaporators, superheaters and reheaters integrated into several sections, and each section can be isolated while all other sections are kept in operation. Of course, such design is more expensive as compared to the integral concept. However, during 20 years of operation this design fully paid for itself.

6.5.2. Burning and freezing of coolants

Sodium burning. Metallic sodium in its pure form does not exist in nature. The reason for this is its high chemical activity, especially with respect to air and water. Sodium interacts with water very intensively in the open air. In contrast to it, sodium reaction with air is slow. Sodium is solid at normal temperature, its surface being coated with the oxide. It is not possible to define the exact value of sodium ignition temperature because it depends on impurities content in sodium, air humidity and sodium-air interface condition. According to the results of studies performed by different investigators the ignition temperature values lie in the range from 140°C to 320°C.

As for burning process intensity, sodium takes one of the last places among other combustibles. This can be illustrated by comparing sodium and petrol pool burning characteristics [6.24]. Temperature distribution in the area above burning sodium and petrol is presented. Burning conditions were equal in both cases, burning surface area was about 1 m^2. Sodium combustion rate was 45 kg/m^2hour. In the point 1 m above the sodium surface the temperature was lower than 100°C. Reaction zone (flame zone) was situated close over the sodium surface. In the case of petrol, the flame was as high as 4 m. Time averaged temperature measured in the point 2 m above the petrol surface was over 600°C, and petrol combustion rate was 4 times higher than that of sodium. This is owing to different physical properties of sodium and petrol. Sodium boiling temperature is 880°C at atmospheric pressure, and evaporation latent heat value is 4340 kJ/kg. This results in its relatively low evaporation rate.

On the other hand, petrol boiling temperature is 80°C and its evaporation latent heat value is 12 times lower as compared to that of sodium. Therefore in the burning petrol boiling point is achieved soon resulting in sufficient vapour amount over its surface. That is why high flame is formed over the petrol. Sodium-air reaction heat is 10900 kJ/kg that is 4 times lower as compared to that of petrol. Thus, energy release rate during sodium burning is approximately 15 times less than that in case of petrol.

The intensity of processes in the reaction zone is determined by two mechanisms: (a) air oxygen diffusion into reaction zone, and (b) opposite direction sodium vapour diffusion from sodium surface into the reaction zone.

The former mechanism basically controls processes at temperatures lower than 650°C. With temperature increase the latter mechanism becomes more and more significant. The higher sodium temperature, the more vapour generation rate. The reaction zone extends from the sodium surface resulting in the decrease of heat transfer to sodium. This, in its turn, decreases evaporation rate. Studies have shown that steady state is achieved at the pool sodium temperature of 720°C to 745°C. During burning process 15 to 25% of the combustion product mass leaves the reaction zone in the form of fume. Nevertheless sodium mass remains almost constant because of simultaneous mass increase due to sodium oxidation.

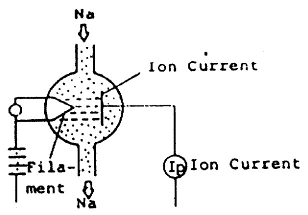
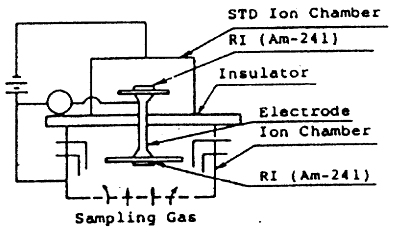
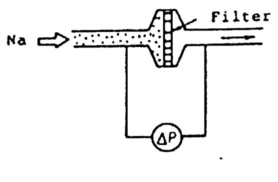
All this concerns burning of sodium spilled over the horizontal plane. However sodium drop burning can also take place. Specific rate of falling sodium burning is higher than that of sodium stationary pool. This also applies to the other burning effects. However in order to spray outflowing sodium, definite conditions are needed which are seldom met in practice. Drop burning effect should be taken into account in case of outflowing jet spraying caused by some barrier (e.g. floor or walls of the room). Continuous sodium spray does not burn because of time deficiency. As in any fire, smoke is one of the most dangerous factors in the sodium burning process. Sodium fume consists of combustion products that have become aerosols. These aerosols are sodium oxides (NaO and Na_2O) coming immediately into interaction with atmospheric components, steam and carbon dioxide producing sodium hydroxide-alkali (NaOH) and sodium carbonate (Na_2CO_3).

The process of oxide into hydroxide transformation takes few seconds after particle formation. Sodium carbonate is formed more slowly. This process rate depends on atmosphere humidity and takes several minutes. Both chemical and physical transformations of particles take place during the fume propagation, as well as conglomeration of particles formed in the burning process. With relative air humidity over 35%, dry particles are transformed into fluid drops. There are changes of their density and size distribution mode. Sodium particle radius varies within the wide range: from several hundredths of micron to tens of microns. In the process of aerosol propagation they deposit on the floor, ceiling and walls of the room, as well as on the surfaces of equipment and ventilation piping. This particle precipitation takes place mainly because of gravity: up to 80% of all particles are deposited on the floor. Aerosol particles can cause equipment damage, concerning first of all electrical equipment and instrumentation. These particles are also dangerous for people.

Sodium leak detection methods. Sodium metal leaking through a small crack in the component or piping wall in LMFR may drop off or flow out along the wall, producing aerosols as a result of its interaction with atmospheric oxygen and moisture.

There are two methods of sodium leak detection. One of these methods is based on detection of certain minimum amount of leaking sodium accumulated in one location. The other method is referred to as gas sampling method, based on aerosols detection in the gaseous atmosphere evacuated continuously by pumping. Table 6.3 illustrates gas sampling method and detection principle. Laser Type Sodium Leak Detector is based on gas sampling, and if sensitivity of gas sampling detector is adequately increased, it is feasible to provide monitoring throughout the whole plant.

TABLE 6.3. TYPE OF GAS SAMPLING AND DETECTING PRINCIPLE

Type	Principle of detection	Sensitivity
Sodium Ionization Detector (SID)		$10^{-10}g/cm^3$
Radiative Ionization Detector (RID)		$10^{-10}g/cm^3$
Differential Pressure Detector (DPD)		2mg

Heavy coolant freezing. One of the most dangerous events resulting in coolant freezing is the secondary steam header depressurization. Rarefaction wave propagates at sound velocity in the secondary circuit on the initial stage of such accident.

Secondary circuit water boiling up and abrupt increase of heat removal from the primary coolant occurs to result probably in its freezing. Steam turbine drive of feed water pump and by-pass pipelines connecting steam generator inlet and steam header through orifices are provided in the secondary circuit to decrease consequences of such accidents.

Evaluations have shown that coolant freezing does not occur in presence of by-pass pipelines in the secondary circuit. If there are no such pipelines, lead freezing begins at 15th s of emergency discharge.

Engineering features are provided in the reactor design to prevent lead freezing due to malfunctions or personnel errors in start-up and transient modes.

Relatively low melting temperature of PbBi eutectic (125°C) required study to be carried out on the primary circuit components and reactor as a whole in case of a change of phase state of the coolant under conditions of accident or scheduled cooling down and the posterior primary circuit heating up («freezing» and «unfreezing» conditions).

Variation of the eutectic volume during its solidification and comparability of its mechanical properties with those of structural materials leads to additional loading of the primary circuit components.

Study of change in PbBi eutectic density within the limits of possible deviation from eutectic within the temperature interval from –50°C to +130°C has been carried out at the IPPE.

It has been demonstrated, in particular, that in case that Pb content deviates by +5% from that of the eutectic composition the volumetric effect of fusion neither varies significantly, nor deviates from the corresponding value for eutectic by more than +0.17%.

Variation of the eutectic volume during its solidification and comparability of its mechanical properties with those of structural materials leads to additional loading of the primary circuit components.

The experimental studies have shown that due to limited solubility of components in the solid state, PbBi alloy is subject to phase-structural transformations, in the course of which its properties change. It has turned out that it is possible to describe these changes using kinetic relationships that have been determined for such most significant properties as density, elasticity and creep.

Experimental studies of features of the alloy-structure interaction were carried out by tests of mock-ups, models and real primary components. Fuel subassemblies, control and safety system components, primary pumps, steam generators, sections of main and auxiliary pipelines and their valves have been tested. In the course of tests requirements have been worked out for the temperature conditions of heating up and cooling down modes, as well as for separate structures designed for avoiding damages at multiple condition changes.

On the basis of experimental and analytical studies algorithms have been developed to implement conditions of particular plants.

The research work carried out and the experience gained in the field of realising freezing-unfreezing conditions facilitate solution of the problem of different type plants to be applied for various purposes and to realise the conditions repeatedly according to the schedule.

CONCLUSIONS

1. Analysis of experimental results on LMFR has served the basis for approximate evaluation of the main characteristics of coolant flow velocity and temperature profiles in the fuel subassemblies with blockage of single-phase flow, i.e. recirculation zone length, coolant flow distribution in SA, coolant temperature rise in the wake, etc. On the other hand, it is clear that currently available data provide only rough estimations of SA thermohydraulic characteristics in case of blockage depending on the Reynolds number and blocked flow cross section area.

2. Results of in-pile and out-of-pile tests demonstrate that boiling occurring behind the blockage does not lead to rapid propagation of failure within the pin bundle. Gas release taking place as a result of pin failure behind the blockage may be the cause of more pin damages in the SA.

3. Studies of various factors influencing temperature profile (such as flow rate, power rating, Reynolds and Peclet numbers and blocked fraction of flow cross section area) call for carrying out additional comprehensive experimental and analytical studies. Special attention should be given to the investigation of thermal hydraulics of SA with porous power generating blockages, as well as initial dryout conditions with boiling occurring behind the blockage and SA cooling limits.

4. Pressure drop in the pin bundles can be evaluated using relationships available.

5. Evaluation of local pressure drop in LCFR core (spacing grids and other structural elements of SA) requires additional studies to be carried out on each modification stage.

6. Heat transfer ratios for square/triangular lattice of the pin bundles (LMFR) are as follows:

   - for laminar flow, Pe=100, x=1.4: $\alpha(sq)=0.6\alpha(tr)$;
   - for turbulent flow, Pe=1000, x=1.4: $\alpha(sq)=0.65\alpha(tr)$

   Taking into account velocity profile and properties of different coolants (Na, PbBi and Pb) the inherent Peclet numbers in LCFRs are 3 times higher as compared to those in LMFRs, thus assuring approximately equal heat transfer coefficients for these reactor cores (provided coolant is purified). If impurities are present in lead or PbBi coolant, one should take into account possible impact on heat transfer, and additional thermal resistance can be evaluated by (6.3).

7. Hot spot factors associated with (a) uncertainty of coolant parameters distribution at the SA inlet and (b) probable distortion of SA configuration during operation, that may impact LCFR parameters, should be taken into consideration, particularly in case of high values of pin bundle relative pitch.

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7. RADIOACTIVITY AND TOXICITY OF COOLANTS

7.1. SODIUM COOLANT RADIOACTIVITY

“Operational” radioactivity. The natural isotope of sodium is ^{23}Na (abundance 100%). Neutron capture processes in sodium (n,γ reaction resulting in 1.4 MeV γ-quanta emittance) lead to formation of ^{24}Na isotope with half-life time of 15 hours, while sodium is flowing through the core. Besides, there is (n, 2n) threshold reaction producing ^{22}Na with 2.6 years half-life time. ^{22}Na emits 1.3 MeV γ-quanta; its activity is proportional to thermal power of the reactor plant.

Neutrons are not practically generated by sodium radionuclides with half-life time exceeding 2.6 years value, even after 50 years of exposition to intensive fast neutron flux.

^{24}Na is the main isotope giving rise to requirement of protection against γ-radiation. Approximately 10 days after reactor shutdown the primary circuit activity is mainly determined by ^{22}Na.

This feature, along with the fact that sodium interacts chemically with water and air, results in three circuit reactor power plant design, including:

- primary circuit containing radioactive sodium heated up in the core,

- secondary non-radioactive sodium circuit coupled with the primary circuit by the intermediate heat exchanger,

- tertiary water circuit producing steam for electricity generation.

Within the primary circuit radioactive coolant is protected against air by the steel barriers and cover argon gas. Radioactive sodium of the primary circuit is separated from non-radioactive sodium of the secondary circuit by the steel tubes of the intermediate heat exchangers.

The operating experience of different reactors has shown, that the coolants separated by the wall of heat exchanging component of the reactor heat removal circuit (steam generator in PWR, turbine condenser in BWR and intermediate heat exchanger in LMFR) should be the same to assure nuclear plant safety and reliability (or at least highly compatible to each other in order to avoid plugging of the core flow channels by the products of coolants interaction). The rise of the total NPP cost owing to this measure (application of identical coolants in the primary and the secondary circuits) and use of tertiary water-steam circuit is insignificant, since standard operation and maintenance of steam generator, water circuit and turbine plant are applied.

Operational experience gained on LMFRs, such as BN-350, Phenix, PFR, BN-600 etc. has proved, that sodium is practically non-corrosive to stainless steel, and content of impurities, mainly oxygen and carbon, are maintained at acceptably low level by the cold traps installed in the bypass of the main coolant circuit. Radioactive hazardous isotopes (caesium, tritium, strontium and iodine) are retained by sodium.

The primary sodium activity in the reactor under operation, mainly determined by ^{24}Na (BN-350 reactor, Kazakhstan) is about 10 Ci per kilogram of sodium [7.1], while after the reactor has been shut down for decommissioning residual activity of ^{22}Na is ~1×10^{-4} Ci per kilogram of sodium [7.2].

If any component of the primary sodium circuit is to be removed from the reactor for the purpose of repair/maintenance, sodium sticking to the surface must be removed because of (a) chemical reaction of sodium with oxygen and moisture in air, and (b) radioactivity of sodium. The process of radioactive sodium removal has been selected for the reasons of safety, effective cleaning of different components, economics, etc.

Cover gas is another source of LMFR radioactivity. Primary gas activity is to considerable extent determined by the impurities in sodium and activation of ^{40}Ar and ^{41}Ar. As a result of (n, p) reaction, radioactive ^{23}Ne with short half-life of 38 s is produced from ^{23}Na [7.3].

Residual radioactivity. As it was pointed out in [7.4], the amount of the long lived radioactivity generated in sodium by neutrons is negligible. Activation of sodium reaches equilibrium state in about ten years of the first cycle of its use and will never exceed this level. The long-lived radionuclides furnished by fission products, sodium impurities and corrosion activation products are chemical elements alien to sodium, that makes possible its external contamination at the reactor plant decommissioning stage.

Two strategies have been considered for the disposal of contaminated sodium:

- separation of radioactive products from sodium, reuse of the purified sodium and conversion of the concentrated waste into an inert form for permanent repository,

- retaining of radioactive sodium on the plant site for later use.

Analysis has shown [7.4], that sodium coolant of 1 GW(e) LMFR after 50 years of operation and ~50 years of retaining may be exempted for free use in the industry or returned to the nature.

It was pointed out at the IAEA consultants meeting on the BN-350 reactor (Kazakhstan) decommissioning (Obninsk, Russian Federation, 23–27 Feb. 1998), that the main radionuclides determining sodium radioactivity are Na-22 ($T_{1/2}$=2.6 a), Co-60 ($T_{1/2}$=5.3 a) and Cs-137 ($T_{1/2}$=30y).

7.2. LEAD-BISMUTH AND LEAD COOLANTS RADIOACTIVITY

“**Operational” radioactivity.** The natural isotope of bismuth is Bi-209 (abundance ~100%). Neutron capture results in the formation of Po-210 by the following reaction:

$$^{209}Bi(n,\gamma)^{210}Bi\xrightarrow[5.0d]{\beta^-}{}^{210}Po\xrightarrow[138.8d]{\alpha}{}^{206}Pb_{st}$$

The small amount of Po-209 is formed from Po-210 by (n, 2n) reaction.

Po-210 having 138 days half-life undergoes α-decay, and Po-209 decays in a similar way with a half-life of 120 years. Polonium is volatile at coolant operating temperatures, some amount of it migrating to the cover gas, where it forms aerosols [7.6, 7.6a]. Leaks of cover gas or coolant may cause contamination problems and maintenance of components requires special measures to protect personnel.

Even in case of lead coolant the problem of polonium contamination exists because of Bi-209 formation by neutron capture in Pb-208 (abundance 52.3%) [7.5]:

$$^{208}Pb(n,\gamma)^{209}Pb \xrightarrow[0.14d]{\beta^-} {}^{209}Bi(n,\gamma)^{210}Bi \xrightarrow[5.0d]{\beta^-} {}^{210}Po \xrightarrow[138.3d]{\alpha} {}^{206}Pb$$

Although the rate of production of Po-210 by this two-step process is much lower (1000 times lower), fraction of polonium migration out of the coolant is higher (perhaps, by ~100 times) because of higher coolant temperature.

In the report [7.5] by V. Orlov, et al. it is pointed out, that in the reactor cooled by lead-bismuth polonium activity value is determined by reaction on ^{209}Bi isotope, and equilibrium activity is ~10 Ci/kg. As for lead cooled reactor ($5\cdot10^{-4\%}$ Bi) polonium activity is determined by reaction on both ^{209}Bi and ^{208}Pb isotopes and may reach $5\cdot10^{-4}$ Ci/kg value by the end of the reactor service life.

Authors of the report [7.5] have noted, that such lead-bismuth coolant activity gives rise to the problems even under normal operating conditions. They consider that in case of the cover gas leak rate of 0.01% of its volume per day, release of ^{210}Po to the central hall may cause (if gas circuit is not cleaned from polonium) 200 fold exceeding of its maximum permissible concentration (mpc) [7.1]. To ensure that mpc value is not exceeded for personnel in the central hall, it is necessary to comply with very strong requirements for the reactor cover gas circuit leak-tightness.

Residual radioactivity. Experts in nuclear technology believe that selection of coolant for the future wide-scale nuclear facilities with fast neutron spectrum should be done taking into account management and disposal of the radioactive waste arising from NPP decommissioning. Specialists suppose that post operational characteristics of a reactor plant including spent coolant waste should be considered as highly important as those related to the operation stage. The primary circuit coolant of the NPP could be one of the main potential sources of radioactive waste.

Long term residual activity of sodium, lead-bismuth and lead coolants of fast reactors have been studied and compared by V. Ousanov, D. Pankratov, et al. [7.4]. It was found, that specific α-activity of typical lead-bismuth coolant is determined by Bi-210m (half-life = 3.6×10^6 years) generated in reaction Bi-209 (n,γ) Bi-210m. The long lived β-activity of Bi-208 (half-life = 3.65×10^5 years) is produced in the reaction Bi-209 (n, 2n) Bi-208.

The most important contributor to specific long-lived residual radioactivity of lead coolant is Pb-205 (half-life = 1.51×10^7 years) generated in the reaction Pb-204 (n, γ) Pb-205. The specific β-activity of the pure lead coolant is significantly lower than that of lead-bismuth. Activation of lead-bismuth and lead coolants will increase in every stage of recycling, if recycling is possible in principle [7.4].

Thus, the residual activity of lead-bismuth and lead coolants is expected to be as high as millions of years. As it is pointed out in [7.4], purification of lead-bismuth and lead coolants from the long lived radionuclides of bismuth and lead (if it is possible) would be too expensive.

In the summary [7.4] it was concluded that "the sodium coolant appears to be the most appropriate for realisation of the non-waste goal among three liquid metal coolants considered".

Lead-bismuth purification from polonium. Some studies in this field have been carried out at the IPPE (Obninsk, Russian Federation). In particular, the possibility of polonium alkaline extraction from lead-bismuth was studied, coolant flow rate of ~0.1% being sufficient to decrease polonium activity in the circuit by over 4 orders and transform extracted polonium into non-volatile compounds. Periodic polonium removal from the circuit is possible for instance before scheduled repair.

It is important to assess evaporation rate of the media, where Po is present. Po can be either in elementary form or in chemical compounds, such as oxide (PoO), inter-metalloid (PbPo), hydride (PoH), etc. Evaporation rate depends on the type of composition, its temperature, density, gas content and other parameters. Rate of evaporation into vacuum follows the Lenghmure law:

$$G_o = kP\sqrt{M/T}\ , \tag{7.1}$$

where: P-pressure, K = 0.0044, M — molecular mass, T — temperature. If the substance is in solution, evaporation rate is proportional to mole part "m": $G=mG_o$.

Feuerstein experiments [7.7, 7.8] on Po evaporation from eutectic Pb-Li-17 have shown that within 300–800°C temperature range, evaporation rate of Po into vacuum is 10^3 times lower than that of elementary Po. Inert gas atmosphere (Ar, Ne) reduces evaporation rate by 10^3 times more. Similar data was obtained by Tupper [7.9] in the experiments on Po evaporation from PbBi.

Thus, Po may be thought of as evaporated from PbBi in the form of PbPo or BiPo. Saturation pressure for polonide vapour follows the relationship:

$$lgP=6.94-(7270/T). \tag{7.2}$$

Calculations made using the relationships presented give the following polonium yield rates:

From PbBi with activity of 1.85 10^{13} Bq/L at 300°C	-	$6.7\cdot10^5$ Bq/kg
From Pb with activity of 1.85 10^{10} Bq/L at 500°C	-	$1.15\cdot10^6$ Bq/kg

Although Po activity in lead is 1000 times lower than that in PbBi, polonium yield from lead is approximately the same due to higher temperature.

Polonium and reactor engineering. In the overviews by B. Gromov, D. Pankratov, et al [7.9, 7.10] general conclusion was made on that Po-210 activity cannot be considered as the main barrier preventing use of PbBi coolant. This conclusion is based to considerable extent on the operating experience of NPP, nuclear submarines and their ground-based prototypes. Results of studies on beyond design accidents on PbBi cooled stationary reactors are presented in [7.11].

Experience gained on LOCA studies has shown that application of protective coating makes it possible to confine polonium by preventing its release to the environment. This coating effect is based on sorption and dissolution of impurities in dispersed medium and fixing in the coating.

In case of steam generator tube failure and depressurization of the secondary circuit, PbBi alloy can find its way into the secondary circuit and water can be contaminated by polonium. In this case basic amount of polonium is kept in the alloy, condensate saturation activity of $10 \div 10^4$ Bq/kg is reached, and SG inner surfaces become contaminated owing to polonium sorption from the water. Water evaporation determines radioactivity level in the turbine hall.

Contamination of the inner surface of the secondary circuit gives rise to danger in case of equipment repair, since the alloy kept in the secondary circuit is a permanent source of water contamination. Water replacement and SG inner surface decontamination without any alloy removal would not give the desired result. The secondary circuit decontamination turns out to be possible only after complete removal of the alloy from the secondary circuit. Obviously, this cannot be assured.

If SG tubes failures are assumed possible, the alloy penetration to the secondary circuit should not be neglected. Tupper‘s experiments [7.9] have shown that PbBi evaporation rate is within the limits given by Raoult law. Lead contained in air may produce the same danger for health as ^{210}Po does.

Polonium activity is one of the important operational problems in case of lead-bismuth used as a coolant. To decrease this problem lead has been proposed instead of lead-bismuth. This has the following consequences:

1. Such problems as control and maintenance of coolant quality, its compatibility with structural materials, corrosion strength, etc. should be studied to a great extent at higher temperatures. Though lead is similar to some extent to lead-bismuth, nevertheless it is a different coolant.

2. Some engineering problems are caused by thermal insulation and heating of loops up to the temperatures over 400^{o}C. Related system should have high reliability, assuring required conditions in both operation and repair stages.

3. There is a problem of elimination of coolant freezing under any accident conditions (failure of heaters, quick pressure decrease in the water circuit, etc.), or its subsequent melting without structure damage.

4. Two-circuit heat removal system with steam-water cycle of supercritical parameters is very difficult for realization, because super high pressure steam generator (~250 atm) should be located inside the reactor vessel (BREST concept), and there is a real possibility of the core blockage by the products of water and heavy coolant interaction resulting in the core melt- down. Such incident took place in the propulsion reactor cooled by PbBi [7.11].

The main hazard is α-activity of polonium aerosols present in air and aerosol deposits on the surface. According to the Russian standards, the acceptable specific radioactivity values are specified for personnel: 2.7 Bq/m^3 (air) and population: $4.6\cdot10^{-2}$ Bq/m^3 (air) and 1 Bq/kg (water and food).

Radioactivity of components. Activity of the primary circuit components is determined by not only natural activity of lead, but also by admixtures (basically Zn, As, Cd and Sb) and corrosion products (See Table 7.1). The activity can be reduced by decreasing Sb content in Pb. The purification technique should be developed.

Among steel corrosion products the greatest contribution to activity is given by Cr-51, Mn-54 and Co-60. LMFR experience shows that corrosion products are deposited on the cold surfaces of the primary circuit. Process of these products deposition from lead is to be studied.

TABLE 7.1. EQUILIBRIUM ACTIVITY OF COMPONENTS

Radionuclides	Half-life time	Equilibrium activity, Bq/L
Zn-65	245 days	$2.6\cdot10^{8}$
As-76	26 hours	$0.41\cdot10^{10}$
Cd-115	54 hours	$2.2\cdot10^{8}$
Sb-122	50 hours	$0.48\cdot10^{10}$
Sb-124	60 days	$0.37\cdot10^{10}$

Gas activity. The inert gas (Ar) above lead level in the reactor usually contains impurities such as polonium, some amount of hydrogen from oxygen content control systems and tritium. Purification of radioactive cover gas can be made by hydrogen oxidation (to water), water separation and adsorption. Purification from polonium can be made by alkaline extraction. All these methods are considered to develop.

The use of U nitride fuel and boron control rods in LCFR (BREST concept) will cause tritium formation as a result of triple fission on N-14 and B-10 nuclei. Tritium activity may reach $2.6\cdot10^{8}$ Bq/day, that is rather high value. Because of its penetration into the secondary circuit through SG tube walls, tritium release into the environment is possible. These assessments are thought to be performed not only by calculations but also using experimental studies.

One of the arguments in favour of using lead instead of sodium relates to severe accidents. To justify this argument, an accident causing reactor vessel and building destruction is considered.

The main problem is that of integrity of the fuel elements. Assuming that damaged fuel elements still keep their integrity, and no radioactivity leakage occurs from the fuel, assessments for LMFR and LCFR were made at the RDIPE showing that radioactivity release in LMFR would be 3 to 4 orders higher than that in lead cooled reactor because of sodium burning. However, if the fuel element failures take place (and this seems to be the most probable) coolant radioactivity becomes less important, and fission products would contribute essentially.

7.3. TOXICITY OF LEAD

When in industrial use, lead is released to the atmosphere, and not only its stable isotope, but also radioactive isotopes, such as Pb-210, Pb-212 and Pb-214 are released. Their sources are radioactive gases (including radon).

Lead is highly toxic substance, it is accumulated in the human body and expelled slowly together with the living activity products. Introduced in a human body it attacks the nervous system, marrow, blood and vascular, disturbing albumen synthesis and genetic structure of cells [7.12, 7.13, 7.14].

Maximum permissible concentrations of lead in air and in water are respectively 10^{-5} mg/L and 0.03–0.1 mg/L. In normal reactor operation lead is kept within gas-tight circuit. Enhancement of lead concentration in working rooms is possible as a result of accidents causing leaks in the reactor upper plate, or its disassembly.

TABLE 7.2. HUMAN DISRUPTION INDEX (ENERGY SUPPLY INPUT)

Flow items	Natural baseline flow	Human Disruption Index	Major causes
Lead	25 000 (t/a)	15	Fuel burning associated processes *
Oil to oceans	500 000 (t/a)	10	Oil processing and waste
Cadmium	1 000 (t/a)	8	Associated processes *
SO_2	50 million (t/a)	1.4	Fuel burning
Methane stock	0.8 ppm	1.1	Agricultural activities
Mercury	25 000 (t/a)	0.7	Associated processes *
Nitrous oxides	10 million (t/a)	0.4	Agricultural activities
Particles	500 million (t/a)	0.25	Fuel burning; land activities
CO_2	280 ppm	0.25	Fuel burning

* Associated processes include metals processing and manufacturing, but no burning. Abridged from U.N. DEVELOPMENT PROGRAMME (UNDP), Energy after Rio; Prospects and Challenges, UNDP, New York, NY (1997).

Human Disruption Index. General indicator of the impact on the global environment attributed to today's energy activities is Human Disruption Index (HDI) (see Table 7.2). It is essentially ratio of human generated additions to the natural baseline situation for energy related environmental factors such as CO_2, SO_2, NO_x, cadmium, lead, mercury toxic particles, oil to oceans and methane stock It includes energy chain impacts such as manufacturing that demonstrates the impacts beyond the production stage.

HDI value is 10 for oil put into the oceans, 15 for lead and 0.7 for mercury release corresponding to the human generated movement of 5 million tons of oil put annually into the oceans and also almost 400 000 tons of lead and 20 000 tons of mercury released into the environment. In the context of large natural inventory of radioactive materials in the earth and significant continuous release of natural radon gas to the atmosphere, additions made by nuclear power have a negligible impact on the natural radioactive baseline situation.

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8. COMPARATIVE CHARACTERISTICS OF REACTOR COOLANTS

The comparison of basic characteristics of most of the suitable coolants for fast reactor application was made on the basis of Sections 2–7 and literature data [8.1–8.4] and given below in Table 8.1 The characteristics of steam and gas (helium, CO_2) were included due to their consideration in several countries as a coolants of advanced fast reactors [8.5, 8.6].

It should be pointed out that presently only liquid metal coolant-sodium is widely adopted for fast reactors. Mercury was used for a short period ("Clementine" reactor in the USA and BR-2 in the USSR having, respectively, thermal output powers of 30 and 100 kW). A number of lead cooled fast reactors are being studied presently.

TABLE 8.1 FAST REACTOR COOLANT COMPARISON

Coolant	Advantages	Shortcomings
1	2	3
Sodium	• best thermohydraulic characteristics; • low reactor vessel pressure(near atm.); • good breeding characteristics: high BR, short T_2 could be provided; • non-corrosive to stainless steel and any fuel compositions; • decay heat removal by passive means; • low density, preventing fuel SA from floating up, and allowing passive safety rod operation (by gravity force); all the ‘dirt’ resulted from the assembly and operation is precipitated in the stagnant zones; •retention of caesium, strontium, iodine and tritium hazardous isotopes by sodium; •neutrons do not generate radionuclides with a half life more than 2.6 years (^{23}Na);	- chemical activity with respect to oxygen and water with explosion potenntial; - high γ-activity of ^{24}Na; - intermediate circuit necessity; - freezing temperature is about 100^0C, so heaters are required for maintaining liquid phase of coolant; temperature required for refueling/repairing is ~150–200^0C; - problems with sodium removal and disposal during reactor plant operation and decommissioning.
Lead, lead-bismuth	• lack the high γ-activity; • chemical interaction with water and air without explosion; • low moderation and absorption of neutrons; • good reflecting properties; • good neutronic performance; • reactor decay heat removal by passive means; • high boiling temperature, coolant void reactivity is negative;	- α-radioactive, volatile polonium produced, any leakage from the cover gas poses a hazard to operators and environment(Pb-Bi) - highly corrosive to steels and some fuel compositions; - high density, probability of core pollution by suspensions (assembling and operating ‘dirt’) in heavy coolants;

		- high freezing temperature (327°C for lead); - residual activity of coolant is high ($T_{1/2}$–$1.5\cdot10^7$y of nuclide ^{205}Pb and $3.6\cdot10^6$y for ^{210}Bi)[8.7]; - formation of the products of water and heavy metal interaction (SG leaks) which could block flow channels of the core [8.8],two circuit design is questionable; - a leak-before-break argument in safety analysis is questionable because of a risk of corrosion damage; - problems with radioactive waste management and coolant disposal during decommissioning.
Steam	• possibility of direct cycle or at most two-circuit heat transfer system; • transparency; • commercial application level; • minimum of chemical reactions within the coolant; • liquid state at room temperature • insignificant activation of coolant itself; • less problems with reactor plant decommissioning.	- primary circuit high pressure; - serious problem of reactor decay heat removal in the case of loss of forced convection or pipe breaks; - high pumping power; - high temperature difference between fuel element cladding and coolant; - corrosion activity with respect to fuel element structural materials; - difficulties maintaining coolant quality to decrease as much as possible its corrosive effect; - absence of technological experience of reactor cooling by steam; - low breeding ratio; - possibility of fission product penetration into a turbine in the case of direct cycle.

Gas (helium, CO_2)	• two-circuit system (no intermediate circuit); • no activation (helium); • less moderating than sodium and so results in a core with a harder spectrum, which allows good flexibility with regard to plutonium management and minor actinide transmutation; • minimum void reactivity effect; • good compatibility with structural materials; •inertness; • less problems with reactor plant decommissioning; • transparent gas atmosphere facilitates in-service inspection and maintenance • synergy with the technology of HTRs, which already exist or is under development; • principal possibility of direct cycle realization (helium).	- high system pressure; - high pumping power; - necessity for use of fuel element cladding with improved heat transfer surface provision, e.g. by high degree of roughness; - high temperature difference between fuel element cladding and coolant; - serious problem of reactor decay heat removal in the case of loss of forced convection or pipe breaks; - difficulties concerning gas leak from circuit; -absence of commercial scale experience; -low breeding characteristics (for CO_2).

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CONTRIBUTORS TO DRAFTING AND REVIEW

BOGOSLOVSKAIA, G.P.	Institute of Physics and Power Engineering, Bondarenno sq. 1, 249020, Obninsk, Kaluga Region, Russian Federation
KIRILLOV, P.L.	Institute of Physics and Power Engineering, Bondarenno sq. 1, 249020, Obninsk, Kaluga Region, Russian Federation
KUPITZ, J.	International Atomic Energy Agency, Wagramer Strasse 5, P.O. Box 100, A-1400 Vienna, Austria
HEUSENER, G.	Forschungszentrum Karlsruhe, Postfach 3640, D-76201 Karlsruhe, Germany
NISHI, Y.	Central Research Institute of Electric Power Industry, 1-6-1, Ohtemachi, Chiyoda-Ku, Tokyo, 100-8126, Japan
RINEISKII, A.	International Atomic Energy Agency, Wagramer Strasse 5, P.O. Box 100, A-1400 Vienna, Austria
STANCULESCU, A.	International Atomic Energy Agency, Wagramer Strasse 5, P.O. Box 100, A-1400 Vienna, Austria
WADE, D.	Argonne National Laboratory, Argonne, Illinois 60439, United States of America
YAROVITSIN, V.V.	Institute of Physics and Power Engineering, Bondarenno sq. 1, 249020, Obninsk, Kaluga Region, Russian Federation