

Status and Prospect of Development of Lead-Alloy-Cooled Fast Reactor

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Abstract

STATUS AND PROSPECT OF DEVELOPMENT OF LEAD-ALLOY-COOLED FAST REACTOR. The status and prospect of lead alloy-cooled fast reactor (LFR) have been explained for future development of various types of LFRs that will be required for sustainable development of the world. Advantages of the LFRs compared with sodium-cooled fast reactors (SFR) are emphasized based on inherent safety features of the LFRs. Topics of research studies for the development of LFRs: compatibility of materials with lead alloy, oxygen control technique, etc., have been described referring recent studies. Long-term development plan and research subjects are described.

Keywords: *Lead, Lead-bismuth, Fast reactor, Neutronics, Material, Corrosion, Thermal-hydraulics, Safety*

1. Introduction

Efficient utilization of nuclear fuel using fast breeder reactors is essential for sustainable development of the world in long term future. Lead alloy-cooled fast reactors (LFRs) with inherent safety features have been proposed as innovative small nuclear reactors in IAEA [1] and Generation IV International Forum [2]. The lead alloy means lead (Pb) and lead-bismuth eutectic (LBE, or 45.5%Pb-55.5%Bi). A research and development plan for a lead-cooled Demo reactor, a lead-cooled small reactor and a centralized lead-cooled large reactor is under planning by the GIF-LFR Provisional System Steering Committee (PSSC). As the lead-cooled pool-type reactor, ELSY (600 MWe) has been under conceptual design since 2006, being sponsored by the Sixth Framework Program of EURATOM. The lead-cooled fast reactors, BREST-300/1200, and the lead-bismuth-cooled small reactors, SVBR-75/100 [3] have been proposed in Russia based on the technology of lead-bismuth-cooled thermal reactors for submarines.

Japanese government has chosen sodium-cooled fast reactor (SFR) for the development of future demonstration fast reactors. The other types of fast reactors including LFR have been excluded from the candidate of the future fast reactor since 2006. However, it is requested to present a concept of the SFR that meets the requirement of competitiveness to the light water reactors in performances and economy by 2015. The design of the sodium-cooled demonstration reactor that has been proposed has some innovative design concepts for economical competitiveness to the conventional LWRs. However,

their feasibility has not been demonstrated. Therefore, if the concepts cannot be adopted for the demonstration SFR, it may be obvious that the demonstration SFR loses the economical competitiveness to the LWR. In this case the revival of LFR may be probable in the future [4].

The competitiveness of LFRs to LWRs in performances and economical cost will be achieved because of various inherent safety features of the LFRs. An issue of the compatibility of materials with lead alloy coolants has been pointed out for the development of the LFR. However, corrosion-resistant material has been already developed in Russia, and some modified steels have been tested in Japan and Europe with successful compatibility. Therefore, it may be probable for the LFRs to be chosen for deployment of fast reactor in long term future for sustainable utilization of nuclear fuel.

2. Potential Utilization of LFRs

Potential application of LFRs are presented in Table 1. There may be potential needs for small reactors that have inherent safety in developing countries that have insufficient electrical grids, infrastructure and nuclear technology. For instance, small reactors with electric power of 20-100MW may be suitable as energy source in the islands of Indonesia [5]. Medium size nuclear power plants may be necessary for reduction of CO₂ emission. Present authorized total thermal power used in the factories is 6.7GW in Japan [6].

Table 1. Potential application of LFRs

Reactor types	Application
Medium size and modular type	Central power reactor
Small and medium size reactor	Steam and electricity supply for chemical process
Small reactor with long life core, Nuclear battery	Power reactor in developing country and special region remote from electrical grids
Small reactor	Supply of hot water, Desalination
High temperature small reactor	Hydrogen production
High burn-up core	High uranium utilization efficiency
Actinide burner	Burn and transmutation of minor actinide

3. Advantages Of LFR

The LFR has the advantages in safety, proliferation resistance, sustainability (efficient utilization of uranium resource), radioactive waste management (reduction of environmental load) and economy [7].

3.1 Inherent Safety

The neutron cross section of elastic scattering is larger for lead alloys than for sodium. In other words, the mean free path of neutron is shorter, which leads to lower leakage of neutrons, and higher reflector effect for neutron in the LFR than in SFR. These features of LFR provide good neutron economy, which make it easier to design a small reactor.

The reactor can be designed to have low initial excess reactivity, or low Pu enrichment, and low reactivity loss during operation, which means that the LFR has an inherent safety feature.

The void reactivity of the small LFR core can be easily made negative. That is because a decrease of coolant density and/or voiding enhances the leaks of neutrons due to high elastic scattering cross section in coolant. Therefore, the small LFR has the feature of inherent safety rather than a large size SFR that has positive void reactivity.

Coolant void reactivity does not increase so much even if a volume ratio of coolant is increased. This makes it possible to have large ratio of the fuel pin pitch to the fuel pin diameter (P/D). The large P/D provides low pressure loss in the core, which

makes natural circulation cooling easier. This enhances the inherent safety in comparison with the SFR that has to have tight P/D .

On the other hand, the lead alloy with high mass number has smaller elastic scattering cross section than sodium with low mass number. This means that the slowdown ability of neutrons is lower, and then the neutron spectrum deteriorates in the region of lower neutron energy. This decreases the Doppler coefficient, which is disadvantage for inherent safety. This spectrum can be improved by adding hydrogen and beryllium moderator into metal fuel.

The inelastic scattering cross section of the lead alloy is large in the region of high neutron energy. This causes higher slowdown ability, and as a result the neutron spectrum deteriorates in the region of neutron energy higher than 1MeV.

The LFR has more inherent safety features in comparison with the SFR as mentioned above.

Table 2 shows the comparison of the properties of the lead alloys (Pb-Bi, Pb) and sodium (Na). As the lead alloys are chemically inert even if they contact with air and water, the LFR is superior to SFR in safety. However, when air enters the primary loop and contacts with the lead alloys at high temperature, solid lead-oxide PbO is formed, which causes channel blockage. Even if pipe break occurs in steam generators and steam contacts with lead alloys, chemical problem is not so serious because the formation energy potential of steam is lower than that of PbO. However, it is necessary to control oxygen potential in the steam system appropriately by dissolving hydrogen in the feed-water.

Table 2. Comparison of liquid metals

	44%Pb-55%Bi	Pb	Na
Density (kg/m ³)	10150	10500	847
Melting point (°C)	125	327	98
Boiling point (°C)	1670	1737	883
Expansion coefficient in melting (%)	0.0	3.6	2.5
Scattering cross section (b)	6.9	6.4	3.2
Average logarithmic energy decrease	-	0.0097	0.0852
Cost/t (1998)	\$0.55	\$0.25	\$0.17
Chemical activeness	Low	Low	High

The coolant sodium does not have so high boiling point, 880°C. There is possibility of coolant boiling at the unprotected loss of flow (ULOF) and the unprotected loss of heat sink (ULOHS). A large size SFR has positive void reactivity. Therefore, the boiling losses self-control characteristics. On the other

hand, the lead alloys have high boiling points as shown in Table 1. Therefore, the boiling cannot occur in LFRs even at the ULOF and the ULOHS. This implies that the LFR has additional important inherent safety feature.

3.2 Proliferation Resistance

The good neutron economy mentioned above enables the design of breeder core and long life core. The long refueling interval of the long life core is good for proliferation resistance. The proliferation resistance is promoted furthermore in case of a transportable small reactor which is fabricated in a factory, carried to the utility site, operated and carried back to the factory for refueling. This concept for proliferation resistance may fit to developing countries that do not have enough infrastructure and nuclear technology.

3.3 Sustainability (Efficient Utilization of Nuclear Resource)

Because good neutron economy in the LFRs, the breeding ratio can be higher than unity without radial blanket. In addition, breed & burn and CANDU burning is possible, which can realize efficient utilization of uranium by once-through without reprocessing.

3.4 Radioactive Waste (Reduction of Environmental Load)

LFRs are suitable for transmutation of the actinide, which is effective for reduction of the environmental load. The accelerator driven transmutation system (ADS) using lead alloy target and coolant has been studied.

3.5 Economy

Because lead alloys are chemically inert even if the lead alloys contact with air and water, the intermediate system to be necessary in SFRs can be eliminated in LFRs. It reduces the work and expense for maintenance and repair of the intermediate system. According to preliminary cost evaluation, the construction cost is decreased by 8% by the elimination of the intermediate system. In addition, the safety systems for the accident of sodium-water reaction to be necessary in SFRs can be eliminated in LFR. Therefore, the construction cost of LFR is lower than that of SFRs.

The development of the small and module type reactors do not need a development stage from a prototype reactor to a large size reactors. It can reduce development work and expense considerably.

Furthermore, the deployment of the small and module reactors has lower investment risk because initial investment cost is low and the construction term of works is short. This is also economical advantage of small LFR.

4. Status of Research and Development

4.1 Conceptual Design of PBWFR

There are steam generators and a primary coolant circulation pumps in a reactor vessel in conventional design of LFRs. However, it may be better to simplify the inner structure and components that are in contact with flowing lead alloy as much as possible. That is because it is possible to avoid the damage of the inner structure and components due to corrosion and erosion.

There is the possibility to eliminate the primary pumps and steam generators in the reactor vessel in case of LFR by introducing a steam lift pump. Lead alloys are chemically inert even if they come in contact with water directly, it is possible to feed water directly into a hot primary coolant at the core exit, and to generate high pressure steam for turbines [8,9]. Simultaneously, the primary coolant can be circulated by buoyancy of steam bubbles because specific gravities of lead alloys are very large. If the primary pumps and steam generators are eliminated, the reliability of reactor components in the reactor vessel is improved, and the maintenance and repair of the primary pumps and steam generators can be omitted.

Since the heat transfer efficiency of the direct contact heat exchanger is very high, the Pb-Bi-feed-water direct contact method allows us to design a compact steam generation system.

This type of LFR is called the Pb-Bi-cooled direct contact boiling water fast reactor (PBWFR). The specifications of the PBWFR are presented in Table 3 [10].

By a mass production effect, the construction cost of the PBWFR is estimated to be 350,000 yen/kW, and the generation cost is 5.1 yen/kWh. The PBWFR is economically competitive with conventional light water reactors if a transmission cost can be ignored by constructing the PBWFR near the electricity demand site.

Table 3. Specifications of PBWFR

Thermal power (MWt)	450
Electric power (MWe)	150
Thermal efficiency (%)	33
Pb-Bi inlet temperature (°C)	460
Pb-Bi outlet temperature (°C)	310
Max. cladding temperature (°C)	619
Pressure drop in core (MPa)	0.04
Pb-Bi mass flow rate (t/h)	73,970
Steam temperature (°C)	296
Steam flow rate (t/h)	863
Steam pressure (MPa)	7
Feed water temperature (°C)	220
Refueling interval (y)	10

4.2 Research on Thermal-hydraulics of PBWFR

In the operation of the PBWFR, the steam lift pump start-up for Pb-Bi circulation initially. Then, steam generates in Pb-Bi-water direct contact boiling two-phase flow in chimneys above the core. These operating process was experimentally demonstrated at practical conditions: system pressure of 7MPa, steam temperature of 296°C, Pb-Bi outlet temperature of 460°C, and feed-water temperature of 220°C [11].

The basic phenomena of the carry-over of Pb-Bi droplets and Pb-Bi-water boiling two-phase flow in a chimney were experimentally investigated. The performance of the Chevron steam dryer and the electrostatic precipitator were evaluated by means of basic experiment for the prevention methods of the carry-over of the Pb-Bi droplets [12].

The distribution of void fraction, bubble chord length and bubble rising velocity in the chimney were investigated in small scale rectangular channel by injecting Ar gas into Pb-Bi experimentally [13].

An ultrasonic flowmeter has been developed for high temperature sodium flow. It was tested in a Pb-Bi flow loop, and as a result the applicability of the ultrasonic flowmeter to Pb-Bi flow was demonstrated [14].

4.3 Development of Corrosion-resistant Materials for LFR

Compatibility of materials with lead alloys is one of the key issues for development of LFR. Corrosion of materials in lead alloys occurs with the following mechanisms: (i) penetration of Pb and Bi into materials, (ii) dissolution of constituents in the materials into lead alloys, i. e. dissolution corrosion, and (iii) chemical reactions between impurities in

lead alloys and constituents in the materials such as oxidation corrosion. Selective dissolution of nickel has a large influence on the corrosion of austenite stainless steels in lead alloys.

Zirconium was added to the lead alloy for the prevention of the steel corrosion in U.S.A. from 1950's to the beginning of 60's. The Zr reacts with nitrogen in steels to form a protection film of ZrN on steel surfaces [15]. In this case, oxygen concentration in the lead alloys was decreased by addition of Mg to lead alloy for prevention of oxidation corrosion.

In Russia, a self-healing type oxide film was formed on steel surfaces for the prevention of the corrosion [16]. It was achieved by controlling oxygen concentration in lead alloys. According to the Russian study, this method provided good corrosion resistance up to 450°C for austenite stainless steel, to 500°C for ferritic-martensitic steel, up to 650°C for ferritic-martensitic steel with Si addition: EP-823 (1.0-1.3%Si), up to 650-700°C for EP-824 and EP-900 [16].

Recently, the corrosion resistance for the Si-added iron specimens was investigated in USA [17], and Al-Fe alloying surface by means of pulsed electron beam heating has been proposed for the prevention of steel corrosion in Germany [18].

The authors performed a Pb-Bi flow corrosion experiment at temperature of 550°C [19]. It has been found that if the steel contains higher content of Cr, single- or multi-oxide layers is formed well on steel surfaces, and a thin and stable inner spinel layer is effective for corrosion resistance. The multi-layered oxide film was formed, and the liquid metal corrosion was not observed for 12Cr-steels: SS405, HCM12A, HCM12 and ODS. For 18Cr-steel: SS430, a dense and thin oxide film with much Cr was formed on the surface, and corrosion resistance was good. Ferritic-martensitic steels with Cr content of 9-12% were corrosion resistant under the conditions of steam injection into Pb-Bi as in the chimney of the PBWFR [19]. Therefore, 12Cr-steels are applicable to the structural materials. It has been found that high Cr steels with Si and Al addition are corrosion resistant because of the formation of a thin, dense and stable inner oxide layer [20].

As for fuel cladding material, it is found that uniform coating of Al-Fe alloy on steel surface by means of sputtering is effective for protection of base metals [21]. The Al-Fe alloy-coated steels were corrosion resistant in the 1,000 h-exposure test in Pb-Bi at 700°C. No penetration of Pb and Bi into the steels was observed, and the coating layer remained after the exposure. Refractory metals of W and Mo, and ceramics of SiC and Ti₃SiC₂ were also corrosion resistant in the Pb-Bi at 700°C for 1,000 h, although Nb was damaged [21].

It has been confirmed that the control of oxygen concentration to adequate level is effective for the prevention of corrosion through the formation of oxide layer on the steel surfaces.

From the above-mentioned result, we have chosen high Cr steels: HCM12A, HCM12 and Mod. 9Cr-1Mo steel as candidates of structural materials exposed to Pb-Bi at temperature in the range of 400-550°C. Fuel cladding materials should be compatible with lead alloy at the maximum surface temperature of 650°C. It is necessary to add Si or Al in steels to improve corrosion resistance at the high temperature. The coating of Fe-Al alloy to the steel surface by means of sputtering is recommended since it protects base metal from penetration of Pb and Bi successfully. Refractory metals of W and Mo, and ceramics of SiC and Ti₃SiC₂ may be applicable to the materials for flow regulating structure for prevention of erosion in flowing field.

The issue of corrosion and erosion in the LFR may be solved by using these corrosion-resistant materials adequately.

4.4 Control of Oxygen Potential, Purity Control and Measurement Technique

When lead alloy coolant is oxidized, solid oxide (PbO) blocks coolant channels and stops coolant flow in the core. This prohibits the heat removal in the core. In order to prevent the problem, oxygen potential in a lead alloy should be kept lower than the oxide PbO formation potential. At the same time, a self-healing type oxide film should be formed on the materials surface as corrosion prevention measures. This can be achieved by keeping the oxygen potential in a lead alloy higher than the formation potential of Fe₃O₄.

There are two methods for the control of oxygen potential in the lead alloys. One is to inject a mixture gas (inert gas, hydrogen and steam) with predetermined oxygen potential into lead alloy. Another is to control the temperature of solid PbO particles packed in a tank in lead alloy-flowing channel. Oxygen dissolves from solid PbO particles into the lead alloys or oxygen in the lead alloys precipitates on the solid PbO particles.

The both methods were performed, and adequate oxygen control was realized successfully [23]. The mixture gas was injected into a Pb-Bi circulation loop. The temperature of the solid PbO particle tank was controlled. It is confirmed that temperature control method for PbO particle is suitable for the control at high oxygen concentration of around 10⁻⁵wt%, and that the formation of Pb slug, or solid Pb oxide, is prohibited.

An online monitoring of oxygen potential in lead alloys is necessary. For this purpose, the characteristic of the oxygen sensor was investigated. Reliable zirconia solid electrolyte oxygen sensor was demonstrated, and the long time-measurement using the sensors was achieved.

4.5 Radioactivation Measure

Bismuth Bi²¹⁰ is generated by neutron absorption of Bi, and alpha radioactive nuclide polonium Po²¹⁰ of 5.1MeV is generated by the reaction Bi²¹⁰(n, β)Po²¹⁰[24]. Radiation measure for Po is necessary in cases of a leak of the coolant, fuel exchange, maintenance and repair. Thus, surface decontamination of Po by the baking method was studied [25].

The concentration of Po²¹⁰ is in the magnitude of the forth order lower in natural Pb-cooled reactor than in Pb-Bi-cooled reactor. β-decay of Pb²⁰⁹ mainly generates stable Bi²⁰⁹ among nuclide: Pb²⁰⁹, Pb^{207m} and Pb^{204m} generated by neutron irradiation. In addition, radio-activation by the longer life nuclei becomes the problem [26]. There are nuclei which need around 100 y to decay to the clearance level. Therefore, the disposal of the coolant is a problem, and recycling of the coolant is necessary. As these measures, there is a method to use Pb²⁰⁶ as a coolant in stead of natural Pb [26].

Po of 99.8% becomes PbPo in Pb-Bi. H₂Po is generated when steam is injected into Pb-Bi. According to the estimate of Po for the PBWFR, Po concentration in the primary coolant of Pb-Bi is 1.06x10¹¹ Bq/kg, and that in flowing steam is 4.63x10⁵ Bq/kg. If it is assumed that all the Po in the flowing steam deposits inside the steam system, the radioactivity on the inner surfaces of pipes is 6.2x10⁸ Bq/cm², and that on the surfaces of turbine blades is 1.5x10⁹ Bq/cm². It is extremely higher than the surface contamination density limit of α radioactive material in management area 4 Bq/cm². Therefore polonium decontamination is necessary for prevention of workers' radiation exposure at maintenance and repair for steam systems.

5. Targets and Advantages of LFR Development

5.1 Conceptual Design of LFR

The PBWFR is the fast reactor concept that has safety and economy corresponding to those of the conventional light water reactors. The purpose of the work is to clarify a roadmap to the practical use of this design as a basic concept. Figure 1 shows a diagram of the PBWFR system for supply of both of

electricity and steam. In case that steam is generated by direct contact of water with Pb-Bi by feeding water into Pb-Bi, turbine system may be Po-contaminated. One of the prevention of Po-contamination, the steam system is isolated from the primary Pb-Bi cooling system by steam generators as shown in Figure 2. The function of the steam lift pump remains [27]. This is called the steam lift pump type LFR (SLPLFR). High temperature and high cycle efficiency system and the other working fluid such as CO₂ for the gas lift pump are possible in the SLPLFR.

5.2 Solution of Problems

(i) High density

The penetration of lead alloy to material brings embrittlement, which weakens the material locally. The weakened surfaces of the materials are easily eroded when the materials contacts with flowing lead alloy with high specific gravity. Therefore, the velocity is limited below 2m/s for prevention of the erosion.

The inner structure of the reactor should have mechanisms to prevent parts from floating because the specific gravity of the lead alloy is large.

The power of the coolant circulation pump in the LFR is six to seven times higher than that of the SFR. For the seismic measure, the reactor size is limited to small, and a three-dimensional earthquake proof is taken.

(ii) Thermal-hydraulics

Heat transfer performance deteriorates due to oxide film formation on the heat transfer surface. For the accident where heat transfer pipes of the steam generator break and steam leaks out into the primary loop, pressure increases in the primary loop and an ingress of steam into the core is prohibited. In the lead-cooled reactor, the temperature of the feed water to the steam generator should be higher than the melting point of 327°C by 100°C to prevent solidification of the lead. Because the expansion coefficient of Pb-Bi in melting is 0%, and that of Pb 3.6%. Volume expansion measure should be taken for the lead-cooled reactor.

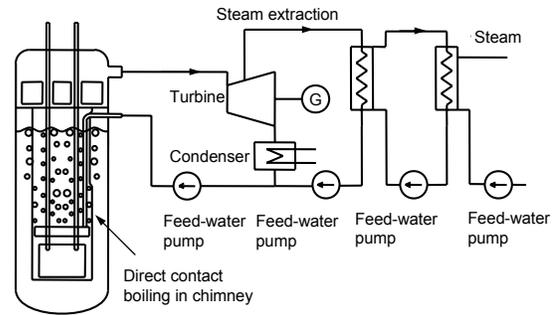


Figure 1. Direct contact boiling type (PBWFR) for supply of steam and electricity

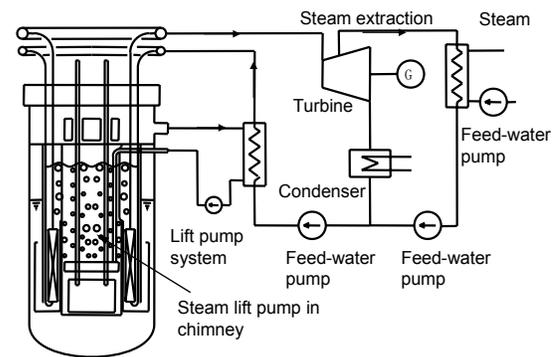


Figure 2. Steam lift pump type (SLPLFR) for supply of steam and electricity

(iii) Resource of Bi [28]

The annual production of the lead in mines is 3,000,000t, but the quantity of natural resources of the bismuth is 260,000t, the deposits 110,000t, and the annual production in mines is only 4,000t. 130 PBWFR plants can be operated at these deposits, and the total power generation becomes 19GWe. One unit of the ADS needs 4,000 t of Bi. The quantity of natural resources of Bi is short even if Bi is reused when Pb-Bi is used for the LFRs. Therefore, when a lot of fast breeder reactors are necessary in the long future, a shift from Pb-Bi-cooled reactors to Pb-cooled ones is possible.

6. Subjects for Research and Development

The subjects for the research and development of the LFR (PBWFR) are presented in Table 4.

Table 4. Subjects for research and development of LFR (PBWFR)

1. Fuel and Core
(1) Fundamental Technology Nitride fuel, Coating technology, Neutron irradiation data, High temperature materials data, corrosion data, Strength standard, Corrosion-resistant cladding material
(2) Fuel and Material Tests Criticality simulation test, Cladding tube test, Pellet irradiation test, Fuel pin irradiation test, Fuel assembly irradiation test, Fuel assembly thermal-hydraulic test/float prevention examination, Control rod/control rod driving mechanism test
(3) Development of fuel assembly analysis code
2. Conceptual Study of Reactor, Safety and Cost Evaluation ULO analysis, ULOHS analysis
3. Development of Reactor Structure
(1) Inner structure Droplet separator/dryer, Chimney, Floating prevention structure, Pump (Electro-magnetic /Steam lift /Mechanical types), Flow test in core, Large-scale Pb-Bi loop construction, Coolant purity control, Fuel assembly thermal-hydraulic test, Material corrosion test, Thermal-hydraulic test for feed-water injection, Performance of chimney, Analytical method
(2) Plant technology Droplet removal with electrostatic precipitator, Protection of turbine blades from Pb-Bi attack, Separator/Dryer, Heat removal performance of PRACS/RVACS, 3-d earthquake proof
4. Measurement technology Oxygen sensor, Ultrasonic flowmeter, Fuel failure detection, ISI for inner structure
5. Fuel Handling Technology
6. Pb and Pb-Bi Technology Oxygen/impurity control technology, Polonium removal, Polonium decontamination

7. Loadmap for LFR Development

Figure 3 shows a long term plan for deployment of LFRs in comparison with the light water reactors and SFRs. In Japan, it is expected that a present light water reactors will be replaced by next generation type of light water reactors from 2030, and that the demonstration SFR will be constructed from about 2030. Commercial SFRs will be deployed from about 2050. The development of the LFRs is delayed by 10 years. It is expected that the LFRs will be deployed from about 2060.

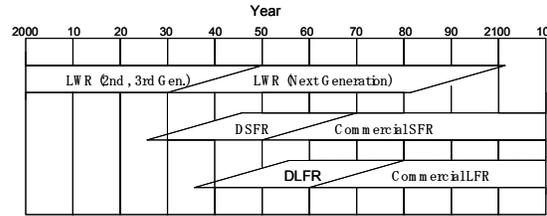


Figure 3. Future plan for introduction of next generation LWRs, SFRs and LFRs

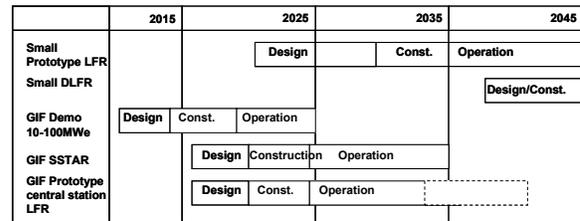


Figure 4. Roadmap for development of LFR

Figure 4 shows a tentative plan for development of small LFRs, compared with tentative GIF-LFR plan. If the conceptual design can start for the small prototype LFR around 2020, it will be constructed in 2030's, and probably a small demonstration LFR (DFR) and a commercial LFR (CLFR) will be constructed and operated. It is expected that core characteristic test and fuel irradiation test data will be available from the startup of the DEMO reactor of GIF in 2020's. If SSTAR of GIF and the construction of the large-size reactor go ahead, the development of the LFR will be accelerated.

For the research and development, basic experiments such as the material development should be continued until 2015. The technology standard that can bear the practical use for the LFR should be established through demonstration test for ten years from 2015 to 2025.

8. Conclusion

The lead alloy-cooled fast reactor (LFR) has possibility to meet various energy demands in the world since its inherent safety characteristics are good. It is expected that the problems specific to lead alloy such as corrosion can be settled by the results in recent studies. It is suggested that the technology of the LFR should be developed to cope with a change of the situation of the fast reactor development after the next 2015, while watching development of the SFR.

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