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Investigation on the Polonium Problem in Lead-Bismuth Coolant for Nuclear Reactors

Sung Il Kim, Kun Jai Lee

Korea Advanced Institute of Science and Technology
373-1, Kusong-dong, Yusong-gu, Daejon, 305-701, Korea

Abstract

By using lead-bismuth eutectic as a coolant for nuclear reactors and liquid metal targets of accelerator driven system attention is paid to high hazard of alpha-active polonium generated in lead-bismuth eutectic under operation of these installation. Therefore rather high technology culture and special means of radiation safety ensuring are required in handling with radioactive lead-bismuth eutectic. The results of investigation are briefly described on lead-bismuth eutectic purification from polonium using alkaline extraction.

1. Introduction

Very long-lived core, lead-bismuth cooled, fast reactors have continued to be investigated in Japan, and in the United States at the University of California at Berkeley. A lead-bismuth cooled, accelerator-driven, sub-critical actinide-burner (labeled ATW) has been proposed by the Los Alamos National Laboratory (LANL) for burning the actinides and long-life fission products from spent light water reactor fuel.

In Korea, KAERI is setting up a long-term research program called HYPER (Hybrid Power Extraction Reactor) and also SNU (Seoul National University) proposed a new transmutation concept designated as PEACER (Proliferation-resistant Environmental-friendly Accident-tolerant Continuable and Economical Reactor). Both of them include the concept of pyroprocess-based partitioning system and lead-bismuth cooled transmutation reactor. So we needed a study on the problem of coolant activation in lead-bismuth coolant reactor.

The use of molten lead-bismuth eutectic as a heavy liquid metal coolant for fast spectrum nuclear system has been deemed preferable over pure lead (Spencer 2000, LANL 1999, Gromov et al. 1997, Zaki and Sekimoto 1995) due to its lower melting point (125 °C vs. 327 °C), which greatly simplifies coolant handling as well as plant design and operation. However, lead-bismuth displays several disadvantages with respect to lead including coolant availability and cost (bismuth is a relatively rare and costly material), materials corrosion (at given temperature the solubility of steel materials is larger in bismuth than in lead) and the radiological hazards associated with coolant activation. Polonium 210 (an extremely toxic alpha-emitter of approximately 140 days half-life) is formed from ^{209}Bi by neutron capture. In a single-phase lead-bismuth reactor the primary system is sealed and separated from the secondary system. As a result polonium is well retained in the lead-bismuth eutectic during normal operating conditions and can cause problems only if coolant leakage through the steam generator or the vessel occurs. Also, some polonium migrates to the cover gas in the reactor plenum and can diffuse outside the primary system of the reactor pool is not perfectly tight. Considerable experience in dealing with polonium related issues has been gained in the past 30 years in Russia where submarines were equipped with lead-bismuth cooled nuclear reactor. A polonium technology was developed that includes special polonium filters for air cleaning, polonium adsorbing adhesive films for decontamination of large surfaces, special respirators, and pressurized suits for maintenance of contaminated area.

In this study, we investigated on polonium problem in lead-bismuth coolant for nuclear reactors and liquid metal targets of accelerator driven systems (ADS).

2. Mechanism of polonium generation

Polonium has no stable isotopes. However, ^{210}Po, ^{214}Po and ^{218}Po do naturally occur in the decay chain of ^{238}U. The isotope of interest in our case, ^{210}Po has a relatively long half-life and emits alpha particles of 5.3MeV with 100% yield. And is therefore known as a pure alpha emitter. It is produced in the lead-bismuth coolant by neutron activation of ^{209}Bi according to the following reaction chain:

$$Bi^{209} + n \rightarrow Bi^{210} \xrightarrow[t_{1/2}=5days]{b^-} Po^{210} \xrightarrow[t_{1/2}=138.4days]{a} Pb^{206}$$

$$pure\ a - emitter$$

$$E_a = 5.3\ MeV$$

$$specific\ activity\ :\ 1 \sim 10Ci / kg$$

Also active polonium nuclides are formed in a lead-bismuth target as results of reactions $Bi^{209}(p, xn)Po^{210} \rightarrow Po^{210-x}(x = 1 \sim 12)$, $Pb^{208}(a, xn) \rightarrow Po^{210-x+2}(x = 2 \sim 14)$. Figure 1 shows the production type and specific activity of polonium on various reactor types. In case of including bismuth in coolant is the most contaminated by polonium.

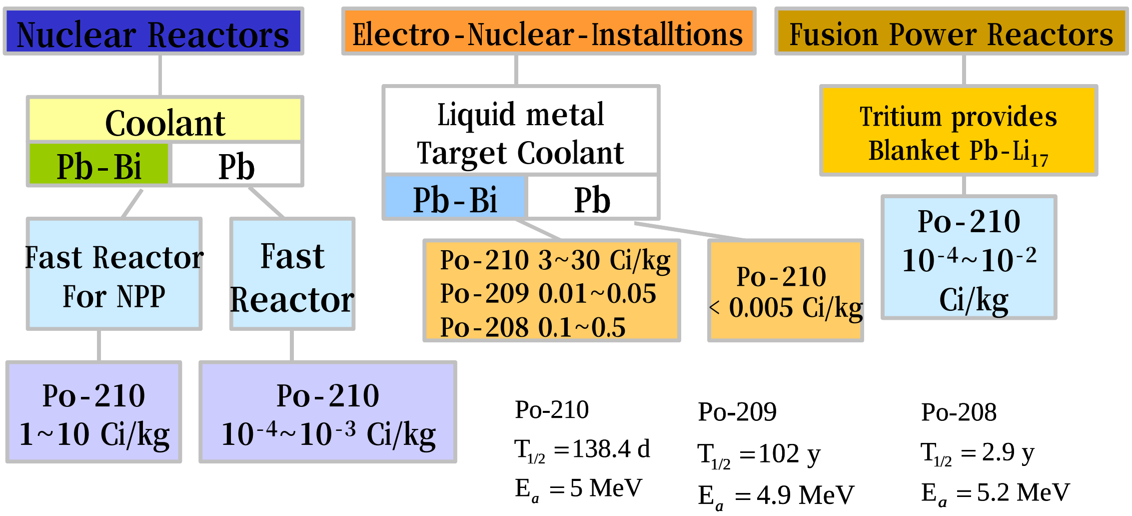

Figure 1. Polonium type and specific activity on various reactor types

3. Polonium hazard

There is no polonium hazard under normal operation conditions when the primary loop is sealed. Hazard appears in the events of the primary loop disclose for repair and maintenance operations, fuel reloading, heat exchanger tightness failure, emergencies with radioactive coolant spills into working rooms and destruction of reactor building. Under lead-bismuth eutectic spilling the main reason of hazard is formation of alpha-active aerosols in working rooms and surface contamination resulted from aerosol sedimentation or mechanical (contact) transport of alpha-activity.

To characterize its radiotoxicity, US made use of the concept of derived air concentration. Because ^{210}Po is an emitter of energetic alpha particles, its derived air concentration is very stringent, 10Bq/m^3 or 3×10^{-10}μCi/cm^3 as reported by the Environmental Protection Agency (EPA 1988), which may raise a potential radiological problem. Also Russia specifies the following permissible limiting levels of ^{210}Po annual intake for the population and personnel :

- Personnel's annual permissible inhalation :

$API_{pers} = 6.7\times10^3 Bq/y$

- Personnel's permissible volumetric activity in air :

$PVA_{pers.\ air} = 2.7 Bq/m^3$

- Population's annual permissible air inhalation :

$API_{pop} = 2.5\times10^2 Bq/y$

- Population's permissible volumetric activity in air :

$PVA_{pop.\ air} = 3.4\times10^{-2} Bq/m^3$

- Population's annual permissible food ingestion :

$API_{pop.\ food} = 1.1\times10^2 Bq/y$

- Population permissible specific water ingestion :

$API_{pop.\ water} = 0.12 Bq/kg$

- Permissible limit surface contamination :

$200/cm^2\cdot\min$

4. Mechanism of polonium release

Polonium exists both in an elementary chemical form and in a form of chemical compounds such as polonium oxide, intermetallic compounds, polonium hydride, ETC. When created by neutron activation in the lead-bismuth coolant, polonium rapidly forms a stable compound with lead, known as lead-polonide. As a result, only up to 0.2% of the polonium present in lead-bismuth is in the unbound elementary form (Po). Upon contact of lead-polonide with water, a very volatile hydride (H_2Po) is formed according to the reaction:

$$PbPo + H_2O \rightarrow H_2Po\uparrow + PbO$$

Volatility of these forms is different and depends on the medium temperature, density and type of the gas atmosphere above the polonium contained melt and other parameters.

A polonium balance over the control volume represented by the total lead-bismuth inventory in the reactor pool yields:

$$\frac{dN_{Po}}{dt} = \Gamma_{prod} - \Gamma_{decay} - \Gamma_{extr} - \Gamma_{rel}$$

N_{Po} : total number of ^{210}Po nuclei

Γ_{prod}: (n,γ)production term

Γ_{decay} : decay term

Γ_{extr}: extraction term

Γ_{rel} : release term

The effect of the polonium extraction system rate on the polonium concentrate on lead-bismuth is remarkable because the extraction term is much larger than the decay and release terms.

5. Methods of polonium removal

Possibilities of polonium removal from a target circuit were analyzed by means of vacuum sublimation and alkaline extraction. Among them the method of alkaline extraction is the most suitable. The first way requires creation of high temperature potentially dangerous devices. The data on high efficiency of alkaline extraction when the coolant was passing through melted mixture of NaOH and KOH were obtained in Russia. One of the important issues of polonium removal system creation is containing and storing polonium removed. Its storage in solidified alkaline is not expedient because of secondary neutron formation as a result of (α,n) reaction on oxygen and sodium nucleus.

The technical experience of polonium removal in Russia is introduced as below.

- Vacuum sublimation

Under the temperature higher than 600°C the lead polonide is dissociated. Volatility of metallic polonium is higher by a factor of 100 than that for bismuth. For that reason, the sublimation method used for extracting polonium out of irradiated bismuth can be used for extracting polonium from lead-bismuth coolant. In the sixties and seventies technology and designs on extracting and concentrating polonium from irradiated lead-bismuth coolant (LBC) by using the distillation methods were intensively developed in Russia. The installation designed for polonium sublimation consists of the apparatus of the so-called stream or "flat" type. Liquid bismuth (or LBC) flows through this apparatus at 700°C and higher temperatures. The capability of one stage of the distillation apparatus provides removal of ~99% of Po under continuous flow rate ranging from ~50 to ~600 kg/hour.

- Alkaline extraction

The method using alkaline melts for extracting polonium out of the liquid metal is the most suitable for organizing continuous purifying lead-bismuth coolant from polonium and from polonium concentrating. This method is based on the reaction of interacting polonium with melted alkali (NaOH) as following:

$$PbPo + 4NaOH \longleftrightarrow Na_2Po + Na_2PbO_2 + 2H_2O$$

The temperature and duration of contacting the alkaline and metallic phases slightly affect the value of the polonium dispersion coefficient. In case of mass relation ("m") of NaOH to LBC + NaOH being ≥0.5, over 99% of polonium is removed into alkaline melt at one-stage extraction. These coefficients are kept in the wide range of polonium and lead-bismuth coolant concentrations that allow achieving the high extent of purifying irradiated lead-bismuth coolant under increasing the number of purification stages or realizing the extraction in the one-through extraction apparatus. The process is carried out in the inert atmosphere because in the air the coefficient of extracting polonium from lead-bismuth coolant decreases sharply.

6. Conclusion

By using lead-bismuth eutectic as a coolant for nuclear reactors and liquid metal targets of accelerator driven system attention is paid to high hazard of alpha-active polonium generated in lead-bismuth eutectic under operation of these installation. Therefore rather high technology culture and special means of radiation safety ensuring are required in handling with radioactive lead-bismuth eutectic. The results of investigation are briefly described on lead-bismuth eutectic purification from polonium using alkaline extraction. But clear conclusion that this purification is expedient may not be made since in this case a system with high polonium concentration would appear in the installation structure. Potential hazard of this system would be rather high. Therefore expedience of its introducing requires additional investigation and substantiation.

Acknowledgement

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