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Accelerator Molten Salt Target System for Transmutation of Long Lived Nuclides

Y. Kate, H. Katsuta, T. Takizuka, H. Takada and H. Yosida

Japan Atomic Energy Research Institute
Tokai-Mura, Ibaraki-Ken, 319-11 Japan

ABSTRACT

Preliminary studies on a molten-salt target system for the transmutation of long lived nuclides is carried out. The advantage of this system is in the continuous on-line processing of the target salt. It is able to control the concentrations of TRU(transuranic)elements and some kinds of FP(fission product) in the salt- Some chlorides are discussed as a target salt for MA(Minor Actinoide) transmutation and a conceptional design of an accelerator molten salt target system is proposed. It is estimated that the MA produced from 10 LWRs of each 1000MWe can be incinerated by the system with beam power of 1.5GeV, 25mA.

[Key Words] : Molten Salt, Target System, TRU, Minor Actinoide, MA, Incineration, Transmutation

1 INTRODUCTION

In the concept of the accelerator transmutation fuel cycle, the remarkable features of a molten salt target system compared with the solid target are as follows:

(1) No need to fabricate the solid target.
(2) Continuous on-line processing method of the salt instead of batch-wise reprocessing method for solid target.

Our molten salt target system is planning to incinerate the MA mainly by high energy fission reactions. The FP and spallation fragments are to be separated as mush as possible from the target salt by the continuous on-line processing system.

To estimate the technological capability of our proposed molten salt target system, the neutron spectrum and thermal power density distribution in the core are calculated at first, then the total amount of TRU incineration in the system are evaluated. In this system, compact type heat exchangers and salt pumps are set up in the core to decrease the total inventory of TRU.

Next, the separation system of FP and spallation fragment from the salt is considered. As the separation methods, gas bubble injection method, reductive extraction method and cold trap method are proposed. These methods will be able to apply individually or in parallel.

2 MOLTEN SALT TARGET SYSTEM AND MINOR ACTINOIDE(MA) INCINERATION

2-1 Selection of the Target Salt

The molten salt as a target material is required to have the following features: a) Composed of elements with adequate nuclear characteristics for the transmutation b) Adequate physical and chemical properties as a fluid. c) Sufficient volubility of TRU elements. d) Good compatibility with the structural materials.

It is well known that some molten fluoride salts were used in the molten salt reactor experiments(MSRE) at ORNL and were proved its excellent features as the fuel and coolant salts of fission reactor. If the sufficient volubility of TRU in the molten fluoride salt is actually confirmed, it will be promising as a target salt(’). In this paper,however, following molten chloride salts[2]are considered as the matrix because of its expected high solubilities for TRU elements and composed of high mass nuclides compared with the fluorides :

A) NaCL-$PuCl_3$ (64-36 mol% : MP=453 “C)
B) $PbCl_2$-$PuCl_3$ (85-15 mol% : MP=476 ’C)

We assume that $PuCl_3$ can be replaced with minor actinoides(MA) such as Np, Am, and Cm. The physical and chemical properties and the compatibility of these salts with practical alloys have not been proved. For the compatibility, we expect that the problem will be improved in resent future by some technology now under developing, such as the surface modification of the structural alloys and also as the redox potential control in the salt to decrease the corrosion rate.

2-2 Target Concept

The conceptional figure of the molten salt target system is shown in Fig. 1 and its scales are shown in Fig. 2 which is simplified for the nuclear calculation. To decrease the TRU inventory, inside core heat exchanger system is adopted in this system.

The molten salt containing TRU elements is circulated through the region I to 111 in the core by centrifugal pumps. In the region I, high energy proton beam is injected into the salt. TRU is mainly incinerated in this region by the spallation and fast fission reactions.

The compact type heat exchangers are set up below the salt pumps in the region III and the total capacity of maximum heat removal is designed to about 800MW. The numbers of the heat exchanger units are to be optimized on account of the heat generation in the core. The temperatures of primary salt at the input and output of the heat exchanger are designed to 650 and 550 'C respectively. The region 11 is a cylindrical neutron reflector{ Hastelloy N) to protect the heat exchangers and pumps from fast neutron irradiation damage.

In the region III the volume ratio of [primary salt : pumps and heat exchangers (Hastelloy N alloy) : secondary salt] are assumed to be [5 : 2 : 4].

The region IV is the fast neutron reflector(iron) and outside of it should be the core vessel but is abbreviated here. The region V is metal window for proton beam injection and the most of its inside space is occupied by forced circulating He coolant gas.

2-3 Estimation of the TRU incineration

The detail of the method to calculate the nuclear reaction in the core is almost the same as described in reference [3]. In the process of this calculation, improved code NMTC/JAERI was used to analyze the spallation reaction[4]. The injected proton beam power is planed to be 1.5 GeV, lmA.

For the salt $NaCl-PuCl_3$ (64-36mol%), it is assumed that 85 wt% of the Pu is replaced by MA(NP, Am, Cm) and the total inventory of TRU in the core is about 5.4t. The results of the calculation on the core are as follows :

Keff = 0.92, Thermal out put = 32 MWt, and Total amount of incinerated TRU = 10 Kg/y·mA.

This result means that the MA from 10 LWRS of each 1000MWe can be incinerated by the beam current of about 25mA.

The neutron spectrum and thermal power density in the core are shown in Fig. 3 and Fig. 4 respectively.

If all the Pu in this salt could be replaced by the MA these results are changed as follows:

Keff = 0.76, Thermal output = 11MWt, and Total amount of incinerated TRU = 3Kg/y·mA

Though the optimization of component ratio in this salt has not yet made, above results show the $PuCl_3$ component is effective to incinerate the MA.

For the another kind of the salt $PbCl_2-PuCl_3$(60-40 mol%),

it is also assumed that 85 wt% of the Pu is replaced by MA. The results are as follows :

Keff = 0.91, thermal output = 28MWt, and Total amount of incinerated TRU = 8.8 Kg/y·mA

Some options will be possible for this target system. For example, the salt pumps and heat exchangers can be set up in the outside of the core and the region II, 111 and IV are filled by graphite moderator. In this moderator region some kinds of long lived FP elements will be transmuted by thermal neutron.

3 ON LINE FP SEPARATION

The fundamental line for our molten salt target system is that the FP produced by fission of TRU should be separated from the salt as much as possible. In the separation process, TRU contaminating into the separation elements should be restricted to the lowest level.

The fission reaction of the TRU in the target molten chloride salt creates free chlorine in the salt. The behavior of the FP elements will be affected by the free energy of formation with chlorine ion. The FP can be qualitatively classified into the following 3 groups under the consideration of the free energy of he chloride formation to perform the separation.

[group-l] Exist mainly in the gaseous state(inert gas or volatile chloride):
Xe, Kr, I, Te($TeCL_4$), Zr($ZrCl_2$)
[about 23% of total FPI

[group-21 Exist as the metal elements. The free energy of the formation for these elements is negative and smaller absolute value than that of Cd. These elements will be reduced to metal by Cd in the matrix :
Nb, Te, Mo, Tc, Rh, Pd, Ru
[about 33% of total FPI

[group-31 Exist in the molten salt as the stable chlorides. The free energy of the formation for these elements is negative and large absolute value.
Ba, Rb, Sr, Cs, Sm, La, Pr, Ce, Nd, Y, Eu, Gd [about 44% of total FPI

The on-line partitioning methods for these FP groups are proposed as follows. For the group-1, He purge method will be suitable. In this method, He gas bubbles are blown into the salt and purge out the FP gas with He. $TeCl_4$, $ZrCl_2$ and I are expected to be purged out but the practical capability are uncertain.

For the group-2, reductive extraction method by liquid Cd will be applied. In the extractor, target molten salt and liquid Cd are contacted directly and these chlorides of group-2 are turned to metals by the reduction reaction with Cd and are separated from the target salt. These elements in the Cd can be removed out by on–line cold trapping method. In the

separated metal elements by the cold trap, Tc-99 will be contained. If the further separation of the Tc is required, it will be possible to use the volatilization method of Tc_2O_7[5]. which is selectively produced in the oxygen gas circumstance.

Finally, for the group-3, cold trap method will be expected. These elements of group-3 will instantly become to stable chlorides after the fission reaction and not be reduced by Cd. These chlorides have high melting point far from that of matrix of the target salt. The efficiency of the traPPing depends on the volubility of these salts to the matrix salt. This is the supporting factor of our proposal, but further studies should be made for the practical application.

The separation methods described above and the ratio of each FP element to the total amount of FP are listed in Table 1 and the concept of on-line separation system is shown in fig. 5.

4 CONCLUSION

Accelerator molten salt target system was mainly considered for the transmutation of MA elements and some separation methods of FP were estimated. In this preliminary estimation of the concept, it is concluded on some assumptions that molten chloride target system is one of the promising system to develop the practical one.

It is the subject to get exact data of molten salt such as the solubilities of TRU elements, physical and chemical properties of TRU contained salt. Next subject to study for the salt will be the compatibility with the structural materials and the FP behavior in the salt. If the enough volubility of TRU is actually confirmed for some molten fluoride salts, they will be promising for transmutation of TRU and FP.

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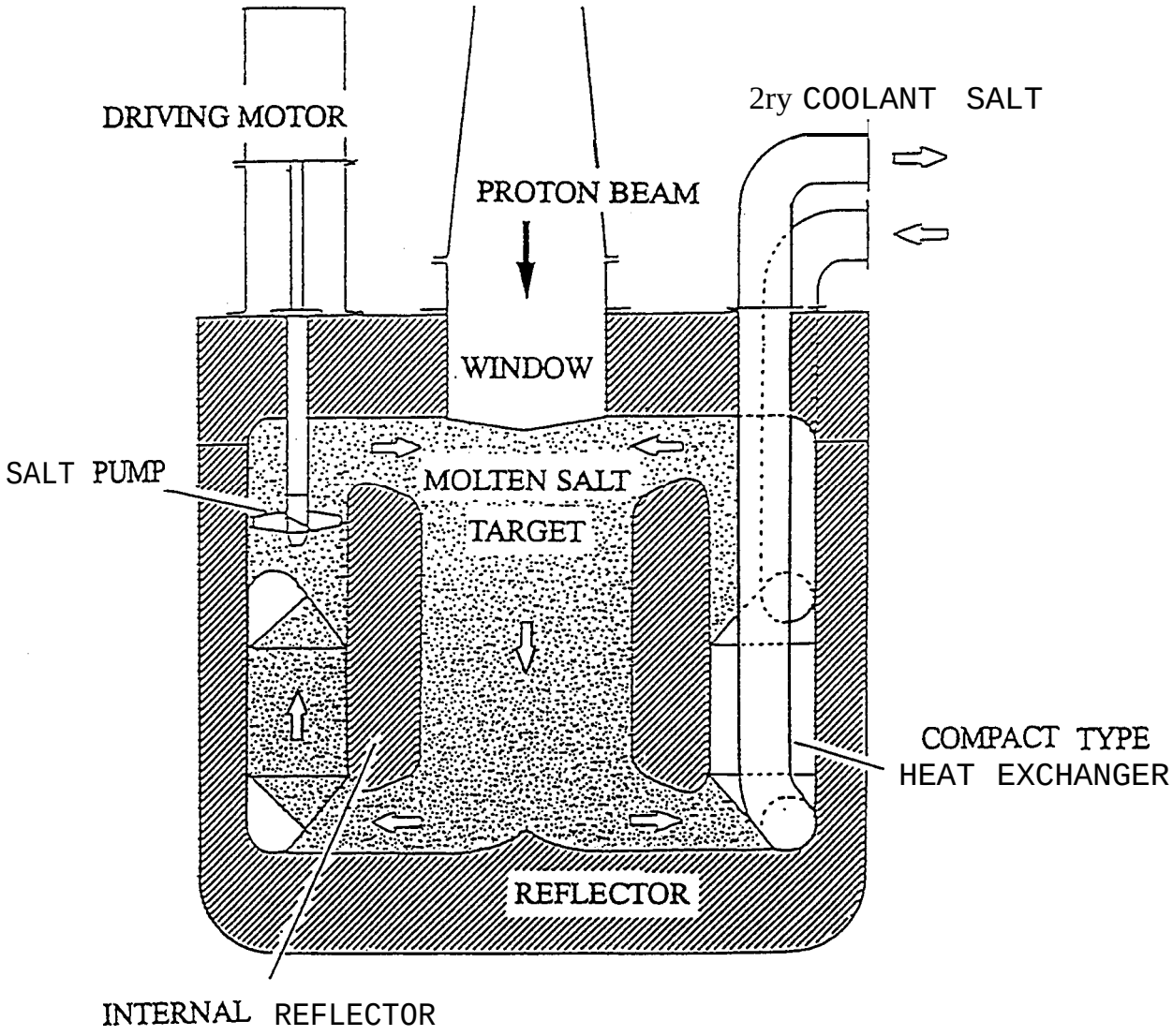
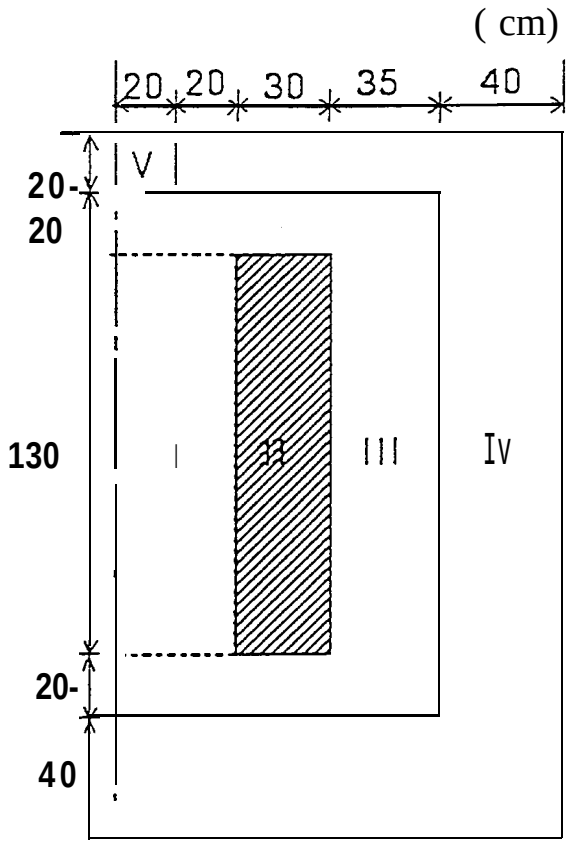


Fig. 1 Concept of the molten salt target system.



REGION

1 : Target Molten Salt
II : Internal Reflector
III: Heat Exchanger Region
N : Reflector
V : He (Beam Window)

Fig. 2 Scales of the Molten Salt Target Core.

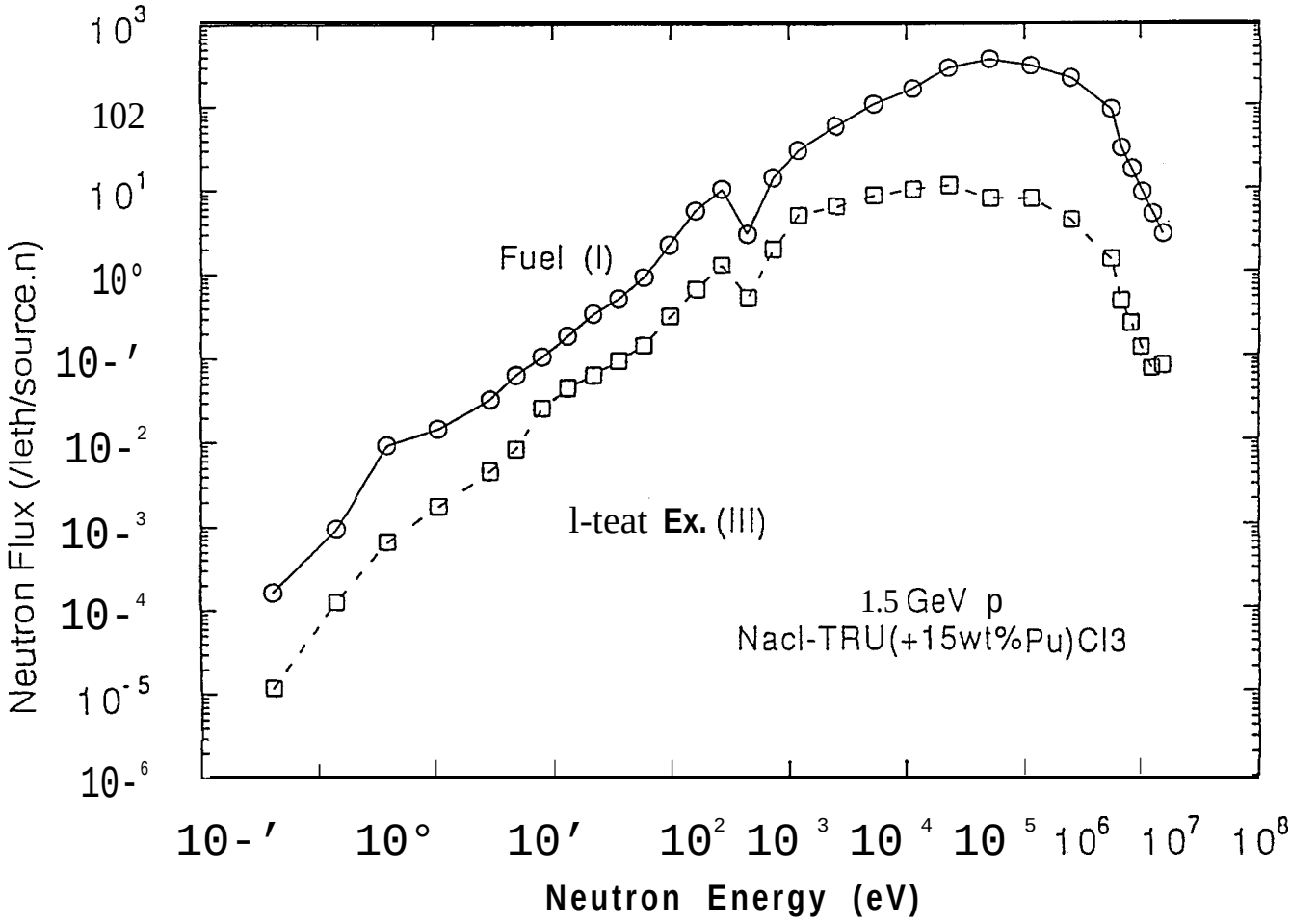


Fig. 3 Neutron spectrum in the target core.

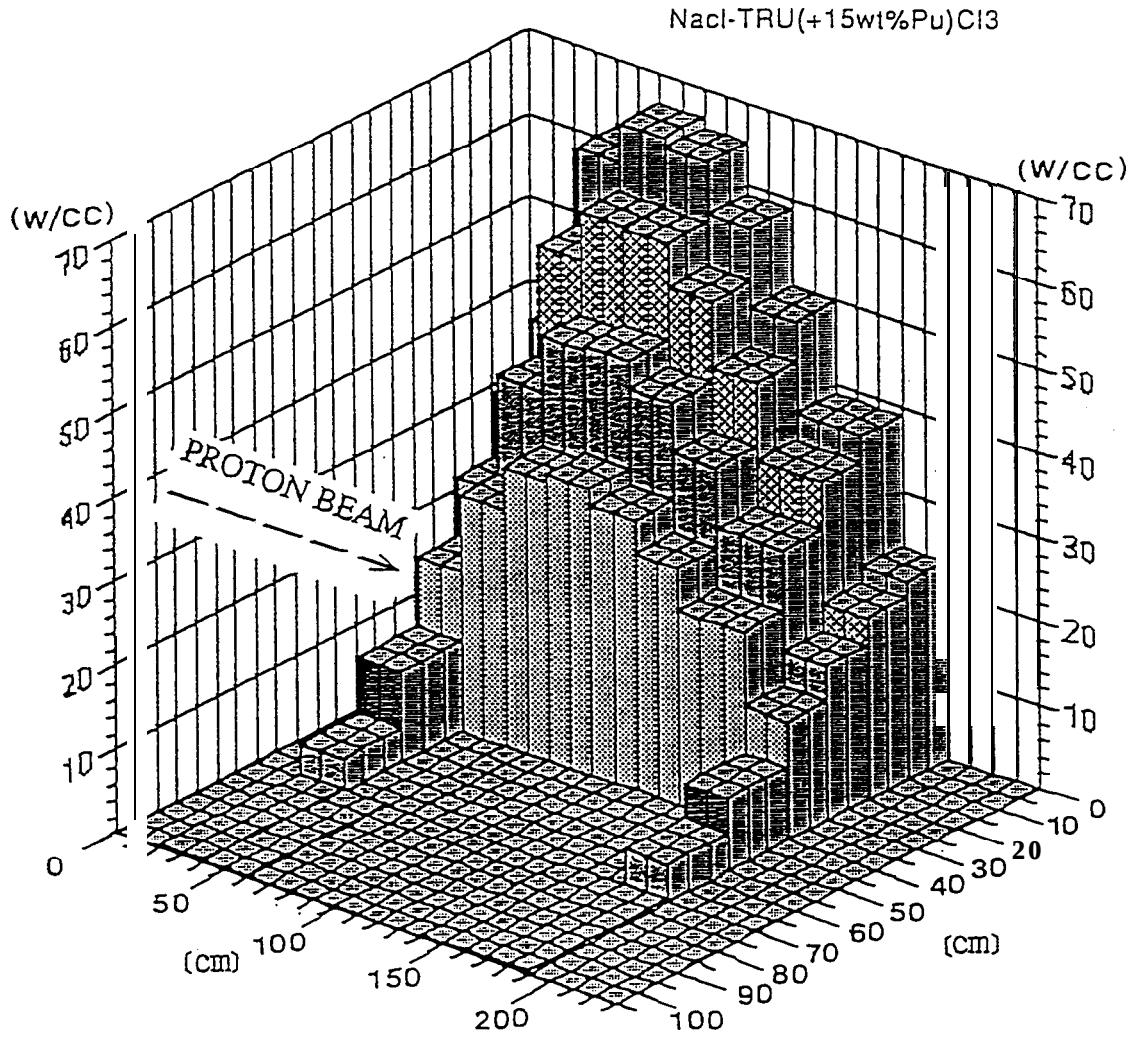


Fig. 4 Power density in the core (< 15MeV, at 1mA proton beam)

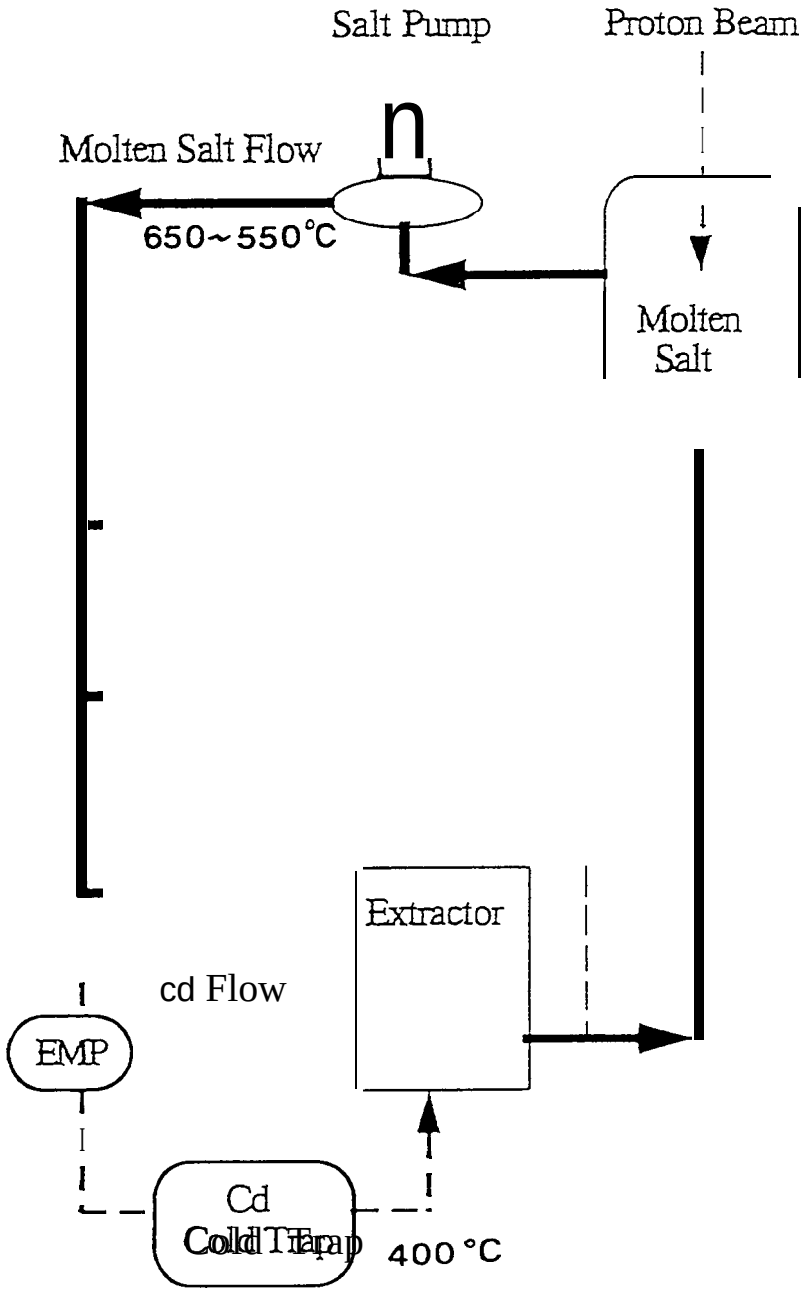


Fig. 5 concept of the on-line separation system

Table 1 Separation Method of FP

He Purge Method (Group -1) 23.8 %		Reductive Extraction Method (Group -2) 33.5 %		Cold Trap Method (Group -3) 44.10/0	
	(c'/o)		(%)		(%)
Kr	0.7	Nb	0.1	Rb	0.6
Xe	13	Mo	11.3	Sr	1.3
Te(TeCl4)	1.8	Tc	2.3	'f	0.7
I	1.0	Ru	8.7	Cs	10.7
Zr(ZrCl2)	7.3	Rh	2.5	Ba	4.4
		Pd	7.3	La	3.3
		Ag	0.7	Ce	6.0
				Pr	3.1
				Nd	10
				Pm	0.2
				Sm	3.1
				Eu	0.3
				Gd	0.4

Values of (°/0) : **WHC-EP-0268(1990)**