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Impurity control in liquid lithium loop for IFMIF target facility

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Abstract

In the IFMIF Lithium target system, the total lithium inventory is about 21 m^3 and the impurities in the lithium are controlled by a cold trap and a hot trap. From the safety standpoint, the control of tritium and beryllium-7 concentration in the lithium is the most important issue. Both elements are to be controlled by a cold trap. As an option for the tritium, a hot trap is also considered. Other impurities such as oxygen, nitrogen and carbon which will affect the compatibility of lithium and loop materials are also controlled by the cold and hot traps to an acceptable level. A swamping method to control the tritium concentration is analyzed.

1. Introduction

In the IFMIF target facility, liquid lithium circulates in the primary loop to produce the intense neutrons by d–Li reaction and to remove the heat as high as 10 MW deposited in the target lithium jet flow by the deuteron beam (250 mA, Max. 40 MW). The lithium flow rate in the main loop is about 130 l/s and the total lithium inventory is about 21 m^3.

The major impurity product expected, in terms of quantity, is the deuterium deposited in the lithium jet by the beam. Among the deuteron–lithium nuclear reaction products, tritium and beryllium (Be-7) are the most noteworthy elements for radiological safety. These elements, as well as oxygen and carbon, are removed from the main lithium loop by the cold trap. As an option for the system, a hot trap with an yttrium getter is also considered for the tritium removal.

Nitrogen and some corrosion products in lithium are removed by a hot trap with titanium getter. A high nitrogen concentration has a potential to increase the corrosion rate on the construction materials and may build up from air contamination during repair or exchange of the components and adhesion to component surfaces. Carbon can build up from HX tube leakage. The design process for the cold trap is similar to the case for sodium and the technology have been well established. It appears to be more practical to remove tritium by swamping method of cold trapping than with a hot trap which have not yet been put into practice for hydrogen isotopes. The analysis for the swamping method used to control the tritium level with a cold trap has not been performed. In this report, a basic evaluation for the swamping method is provided for control of the tritium level.

2. Production rate of the main impurity elements

Tritium is produced by direct reactions of the beam with the lithium, as well as by the capture of low energy

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back scattered neutrons by Li-6. Total tritium production rate is estimated to be about 10 g/y. The tritium inventory has to be minimized in the system since this could be the dominant source term in the event of a radiological release. Tritium is removed by the cold trap with protium sparging, the so-called swamping method, or, as an option, by a hot trap with an yttrium getter. Using one of these methods, the total tritium inventory in the circulating lithium in the loop will be kept to about 3 g.

The most highly radioactive impurity is expected to be Be-7, (half-life: 53-d, decay γ: 0.48 MeV) produced from (d, n) and (d, 2n) reactions with lithium. If not removed, this product will build up to a saturated activity of about 4.5×10^{15} Bq. The Be will exist as a chemical form Be_3N_2 in the lithium and the solubility of Be_3N_2 is about 0.5 appb at 500 K. The cold trap will remove the Be-7, and its equilibrium concentration in the loop will be kept within 1 appb. However, some of Be-7 is expected to deposit around the loop, and is very likely to limit the remote handling operations.

At the initial lithium charge, about 0.84 kg of oxygen impurity is expected to come from the surface contamination of the loop components. During maintenance (4 times per year), about 1.2 kg/y (= 0.3 kg/y × 4) will be absorbed by the lithium. Also during maintenance, about 4 kg /y (= 1 kg/y × 4) of nitrogen will be absorbed by the lithium.

Table 1
Production rate of hydrogen isotopes

	No. of isotope		Weight	Concentration
	per Deuteron	(Atom/s)	(g/day)	(appm/day)
H	0.06	9.36×10^{16}	1.34×10^{-2}	8.76×10^{-3}
D	1	1.56×10^{18}	4.47×10^{-1}	1.46×10^{-1}
T	0.03	4.68×10^{16}	2.03×10^{-2}	4.42×10^{-3}
Total		1.70×10^{18}	4.81×10^{-1}	1.59×10^{-1}

3. Basic analysis for the swamping method of cold trapping

To make the problem simple, the effect of different hydrogen isotopes on the cold trap performance is neglected. The solubility of hydrogen in lithium at 473 K, the operation temperature of the cold trap in IFMIF target system, is about 440 appm (= 63 wppm) [1]. In the initial operation of the target lithium system, the impurity level of hydrogen is estimated at about 105 appm (= 15 wppm) or so, if the system is constructed under good management conditions. When the beam is turned on, the hydrogen isotope will increase to 440 appm from 105 appm with no effect of the cold trap. The production rate for all hydrogen isotope by the d–Li reaction is about 0.159 appm/day as shown in Table 1.

The swamping method is expected to decrease the tritium in the lithium system. Before the beam on target operation, hydrogen is injected into the lithium until its concentration reaches 441 appm. The concentration of hydrogen isotope $C(t)$ and of tritium $C_T(t)$ will be obtained by solving the following equations:

$$\mathrm{d}C(t)/\mathrm{d}t = S_m - (e/T)(C(t) - \mathrm{Co}), \qquad (1)$$

$$\mathrm{d}C_T(t)/\mathrm{d}t = S_t - (e/T)(C_T(t)/C(t))(C(t) - \mathrm{Co}), \qquad (2)$$

where

$$S_m = (S_h + S_d + S_t + M),$$

with S_h, S_d, S_t: the production rates of hydrogen, deuterium and tritium, respectively; M: the hydrogen injection (swamping) rate; e: the impurity removal efficiency of cold trap; $T = V/f$; V: the lithium inventory (21 m^3); f: the lithium flow rate in cold trap (0.25 l/s); Co: the hydrogen saturation solubility in lithium at 473 K. The initial conditions of Eqs. (1) and (2) are

$$C(t) = \mathrm{Co} \ \text{and} \ C_T = 0 \ \text{at} \ t = 0. \qquad (3)$$

Then the solution of Eq. (1) is given as follows.

$$C(t) = \mathrm{Co} + (TS_m/e)(1 - \exp[-et/T]). \qquad (4)$$

The numeric solution of Eq. (2) can be obtained by using Eq. (4), but the analytically approximate solution is obtained in order to exchange the $C(t)$ with an appropriate constant. When we exchange $C(t)$ for $(\mathrm{Co} + TS_m/e)$, which is the limit of $C(t)$ at $t = \infty$, and the following approximate solution is obtained.

$$C_T(t) = S_t(\mathrm{Co}/S_m + T/e) \times (1 - \exp[-t/(\mathrm{Co}/S_m + T/e)]). \qquad (5)$$

For the constants in Eqs. (4) and (5), the numerical values in the IFMIF conceptual design are given as follows: $S_h = 8.76 \times 10^{-3}$ appm, $S_d = 1.46 \times 10^{-1}$ appm and $S_t = 4.42 \times 10^{-3}$ appm with $e = 0.5, V = 21$ m^3, $f = 0.25 \times 10^{-3}$ m^3/s, $\mathrm{Co} = 441$ appm (= 63 wppm).

The results of the calculation of Eqs. (4) and (5) are shown in Figs. 1 and 2. From Fig. 1, it is clear that even for the case of $M = 5$ appm, $C(t)$ is lower enough than the saturation solubility (= 1365 appm) for hydrogen at 523 K which is the minimum operating temperature of the lithium main loop. All the curves are saturated within about 10 days after cold trap operation. Also from Fig. 2, it requires about 3 appm/day for M to restrict tritium to within about 3 g (= 0.65 appm) in the main lithium loop.

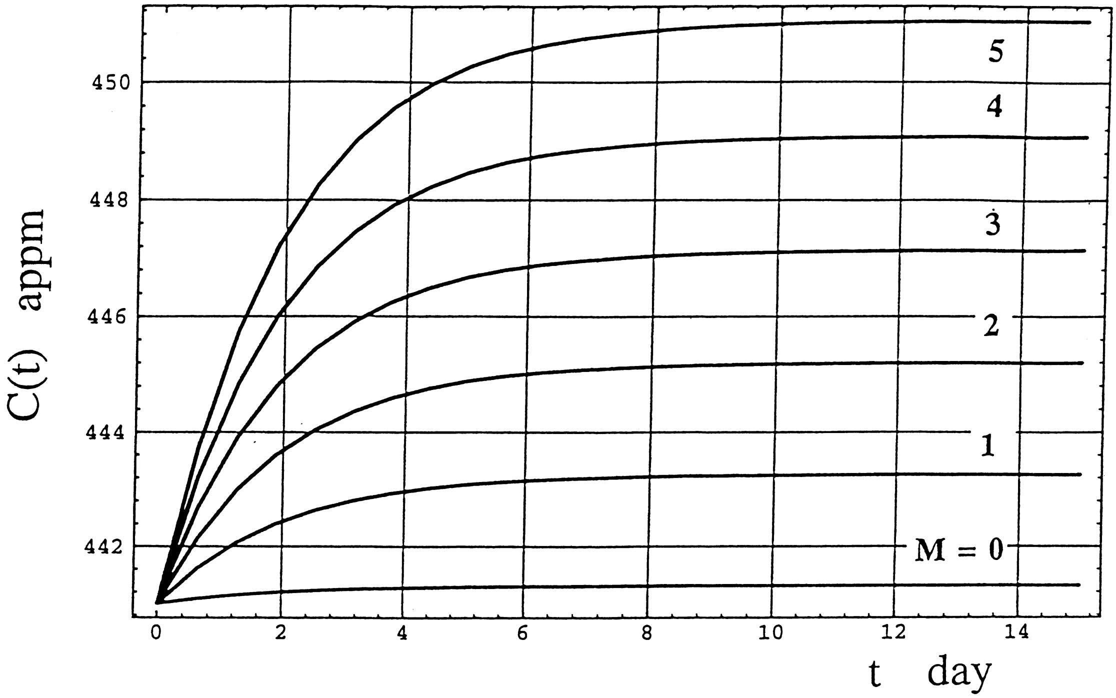

Fig. 1. Concentration of hydrogen isotopes, $C(t)$.

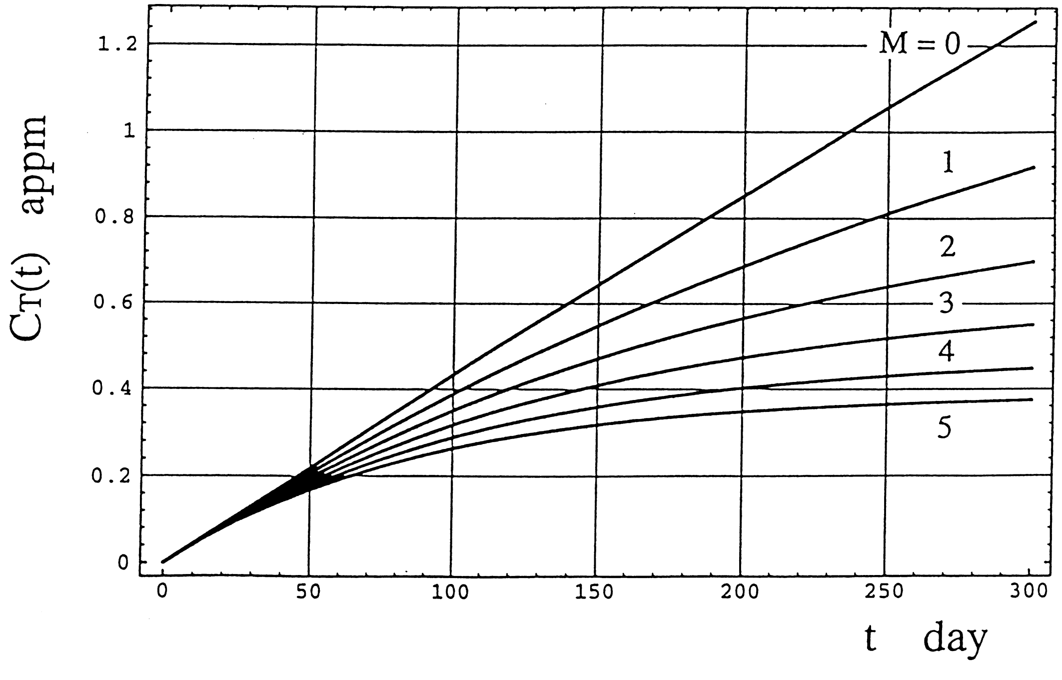

Fig. 2. Tritium concentration, $C_T(t)$.

4. Designs of IFMIF cold trap and hot trap

4.1. Cold trap

In the IFMIF-CDA, the cold trap with swamping is planned to be exchanged once a year. The total amount of hydrogen capture, C_h, is given as

$$C_h = S_m - (\mathrm{Co} + T\,S_m/e) = 1.08\ \mathrm{kg}. \tag{6}$$

The chemical form of the hydrogen deposited is LiH, so the total deposit weight W will be about $W = 8.6$ kg.

To keep the lithium flowing in the cold trap, the liquid lithium volume in the lithium system (mesh volume) should be designed to have more than 75% after 1 year of operation. The mesh volume V_m which contains a steel mesh and lithium is obtained as

$$\begin{aligned} V_m &= (f \cdot \mathrm{Lt}/\beta)(1 - \rho(\mathrm{li})/\rho(\mathrm{sus})) \\ &\approx (f \cdot \mathrm{Lt}/\beta), \end{aligned} \tag{7}$$

where f: the lithium flow rate in the cold trap; Lt: the time of lithium stay in cold trap; $\beta = 1 - \rho(\mathrm{m})/\rho(\mathrm{sus})$; $\rho(\mathrm{li})$, $\rho(\mathrm{m})$, $\rho(\mathrm{sus})$: the densities of lithium, mesh volume and mesh metal, respectively.

Therefore,

$$W/\rho(\mathrm{li}) \leqslant 0.25\beta V_m, \tag{8}$$

so

$$V_m \geqslant W/(0.25\beta\rho(\mathrm{li})) \approx 0.007\ \mathrm{m}^3. \tag{9}$$

We also consider the amount of oxygen trapped in the cold trap. The solubility of lithium oxide at 473 K is about 7 wppm. Even if the total oxygen in the lithium is trapped, and the densities of lithium hydride and lithium oxide are assumed to be the same as lithium at 523 K for the simple and conservative evaluation,

$$V_m = (W + 1.2)/(0.25\beta\rho(\mathrm{li})) \approx 0.1\ \mathrm{m}^3. \tag{10}$$

Then it is clear that the design of the cold trap capacity is dominated by the oxygen concentration.

The equilibrium inventory of Be-7 is estimated to be 0.33 g with an activity of 4.5×10^{15} Bq (half life is 53 days) as noted. Most production will be trapped in the cold trap. The production rate of Be-7 atom is about 1/350 of the production rate for hydrogen isotopes and it will give not effect design of the mesh volume of the cold trap. However, the radiation shielding on the cold trap will have to be designed carefully.

The solubility of carbon in the lithium is about 2 wppm at 473 K. It will be easy to control to this concentration and the initial contamination should be small.

4.2. Hot trap

The solubility of the chemical compound of nitrogen in lithium at 473 K is about 1460 wppm. The hot trap is designed to control the nitrogen and other corrosion products that cannot be removed with a cold trap. In the IFMIF-CDA, titanium is chosen as a getter material for the hot trap. The attainable nitrogen concentration in lithium $C(\mathrm{N,Li})$ is determined by the following equation [2,3]

$$C(\mathrm{N,Li}) = C(\mathrm{N,Ti})/K, \tag{11}$$

where $C(\mathrm{N,Ti})$: Saturation concentration of nitrogen in titanium, K: partition coefficient.

Using the following experimental equations:

$$\ln K = 21.68 + 13526/T\ (\mathrm{wppm}), \tag{12}$$

$$\ln C(\mathrm{N,Ti}) = 4.68 - 3215/T\ (\mathrm{wppm}), \tag{13}$$

we obtained

$$\ln C(\mathrm{N,Li}) = -17.00\text{–}16741/T\ (\mathrm{wppm}). \tag{14}$$

The adequate operation temperature is estimated to be 873 K, and the attainable concentration C(N,Li) is the order of about 10^{-16} wppm on calculation. The result of this small value seems to depend on the model, in any way, the value of $C(\mathrm{N,Li})$ will be small enough.

The maintenance is planned 44 times within the full life time (20 years) of the hot trap. The total amount of about 45 kg of nitrogen should be trapped. In the titanium getter, nitrogen exceeding the saturation concentration is trapped as TiN. The required weight of titanium is determined to be about 155 kg. The design weight of titanium getter is then obtained to be 230 kg with 50% margin. We take about 2 kg/l for a filling density and 4000 $\mathrm{m}^2/\mathrm{m}^3$ for a configuration coefficient. Then the titanium getter volume of 115 l and its effective surface area of 460 m^2 are obtained.

The cold trap and hot trap designed for the IFMIF is shown in Fig. 3. The impurity control sub-system in the lithium target system is also shown in Fig. 4. Each equipment has a spare in this sub-system to get redundancy [4–6].

5. Impurity monitoring sub-system

The following impurity detectors are planned to be set up in the IFMIF impurity monitoring sub-system. Most of them are required to make R&D by real lithium loop experiments in the next CDA-phase.

Hydrogen meter: The concentration of hydrogen isotopes in Lithium are detected by measuring their partial pressure of gas which comes through a Niobium (Nb) or Nb–Zr membrance. To distinguish each isotopes from the other, a quadruple mass spectrometer will be installed.

Oxygen meter: The detector of an yttrium doped solid electrolyte cell of ThO_2 have been developed for

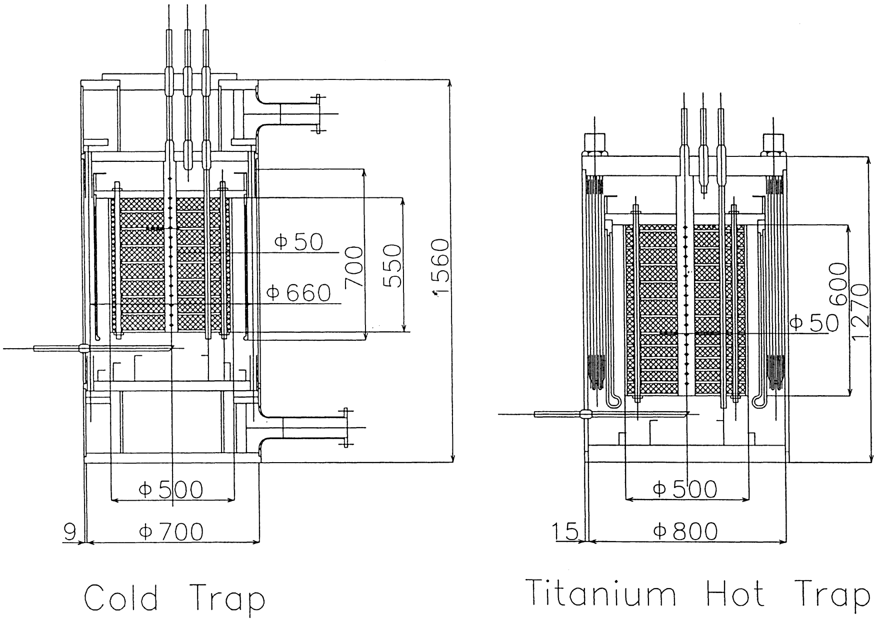

Fig. 3. Cold trap and hot trap.

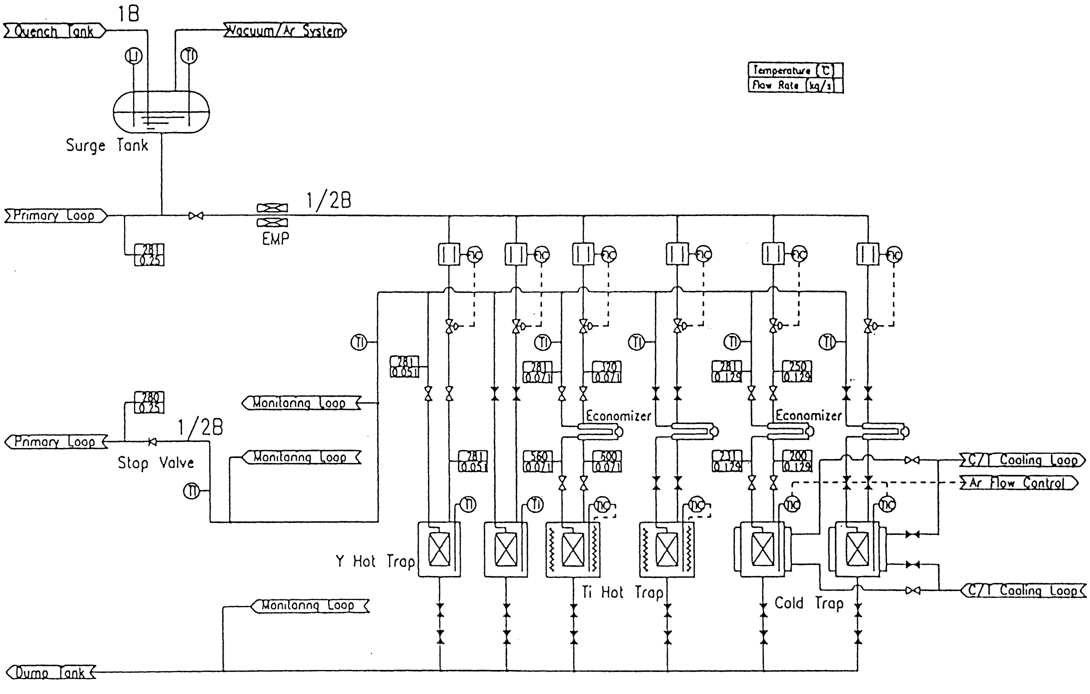

Fig. 4. Impurity control sub-system in target lithium system.

a sodium system. The corresponding detector of this type will be developed for the lithium system. It has to be confirmed the compatibility with the liquid lithium.

Nitrogen meter: This is the electrochemical device applying a molten-salt electrolyte cell. As a molten-salt, LiCl–LiF–Li_3N system will be promising but further study will be required to develop.

Electro-resistivity meter: Electrical conductivity measurement of lithium will be available to detect the high level nitrogen concentration in lithium. R&D is required to get the feasibility.

Off-line sampling system: This is a lithium sampling system to analyze impurity elements in the off-line analytical chemistry system. Practical remote operation system should be developed.

6. Concluding remarks

In the IFMIF target facility, the control of the concentration of tritium and beryllium is especially important for the safety. There have been many design experiences of these equipment in sodium and lithium facilities but the experiments for hydrogen isotopes with swamping and beryllium trapping are very few, so it is important to get these experimental data with a basic lithium loop. Most of the impurity monitoring devices require more basic study and developing efforts.

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