A NEW SUGGESTED SYSTEM DESIGN FOR TRITIUM PRODUCTION

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ABSTRACT

A study has been carried out to put a primary design for tritium production system by radiating one of lithium compounds in the vertical nuclear reactor channels. Pure mono crystal of lithium oxide had been selected as primary substance to produce the tritium which has good physical properties suitable for this purpose. We found that the product of one gram from tritium is needed to use eighteen vertical channels from 14th July, reactor (Baghdad, Iraq) to irradiate about 1437gm from natural lithium oxide. This study deals with the probability to use enriched lithium for this purpose. According to these properties tritium production system was built and its requirements are explained.

1- INTRODUCTION

Hydrogen isotopes (deuterium and tritium) are considered to be the important isotopes used as fuel in the fusion reactors. Therefore, in recent years a technical development has taken place to prepare these two isotopes. Deuterium is considered as a stable isotope and it is found in very rare percentage. It is possible to prepare and store deuterium to be used at any time. However, tritium is unstable isotope which is decayed to ^3He by emitting beta particles with half life of 12.33 year[1]. Other important properties of the tritium are shown in table(1). To prepare a suitable amount of tritium, lithium compounds are generally used. The most important compounds were lithium silicate (Li_4SiO_4),lithium metasilcate (Li_2SiO_3), $LiAl_5O_8$, lithium illuminate ($LiAlO_2$), Li_2 in addition to Li-Al alloy. Each of these compounds has its own thermal and physical properties, which are very important in tritium production operation. The thermal and structural properties[2,3] as a function of temperature are correlated in equations (1-4) as follows:

$$E = 85 \exp(-5.8\,P)[\,1 - 1.8\times10^{-4}(T-293)] \quad (1)$$

$$K = (\,1.98 + 850/T)[(1-P)/(1 + b_p)] \quad (2)$$

$$b = 2.14 - 7\times10^{4}\,T$$

$$C = 9399 + 1.4577\,T - (\,4.011\times10^{7})/(T^2) \quad (3)$$

$$\alpha = 18.8 + 1.66\times10^{-2}\,T \quad (4)$$

Where (P) is the porosity and T represents the temperature in centigrade degrees. The equations are valid in the temperature range of 27 –827 °C. The coefficient (b) in equation (2) is assumed to be the same as that for lithium oxide.
The above mentioned lithium compounds are used as blanket in fusion reactors to produce a suitable quantity of tritium for the work continuity of these fusion reactors. Large quantity of tritium production depends essentially on the neutron flux, the quantity of the lithium and its degree of enrichment (the fraction of ^{6}Li isotope in the radiated model). However, the extraction of tritium from irradiated samples depends on many factors. The most important of these factors are temperature at which the tritium is librated as well as the type and the nature of the irradiated substance [4,5]. The extraction operation is considered to be the essential subject in tritium production. Many foundations extract a large quantity of tritium used as a fuel in fusion reactors [6,7]. For example in France, many specialized reactors produce tritium since 1967. The current study includes the investigation of the primary designs beside the requirements needed for the industrialization of tritium production system.

2-PRIMARY SUBSTANCE SELECTION FOR TRITIUM PRODUCTION

For the purpose of producing tritium, a number of lithium compounds are used; for example ceramic the following compounds; $LiAlO_2$, Li_2O, Li_7Pb_2, Li_4SiO_4, Li_2SiO_3 and Li-Al alloy. In this work, the properties of these compounds have been studied to know the sides which had a relation to produce tritium and their use. Some physical properties for a number of lithium compounds, which are used in the manufacturing of fusion reactors blanket that we have designed specially for tritium production are shown in table (2)[3,2]. However, most of the methods utilized in the extraction of tritium from lithium lead compounds , need more conduction of experiments to verify their competent. The ceramic compounds can be considered as the best compounds used for tritium production especially lithium oxide (Li_2O) which has very well known properties such that it contains a high density of lithium [8,10] as high as 8.2×10^{28} lithium atom/ m^3 [8] with melting point of 1427-1570 °C [8,11] beside its high conductivity [10]. Moreover, tritium is librated with high rate. However, with regard to tritium extraction the compound has some disadvantage namely it is very moisture-sensitive in the powder form reacting with very small quantity of water to give lithium hydroxide[12,13]. Liberation of tritium from lithium hydroxide, at a temperature range of (400-600C^0) as water vapor, leads to the reduction of the tritium concentration in the collected water. However, the highly pure lithium oxide with the mono crystal form has a low capability to react with water to be highly preferable in tritium production[5]. There is no large difference between other lithium compounds and lithium oxide in the way of tritium extraction. Therefore, it is difficult to select a substance among these compounds to be used as a primary material in tritium production. Many references indicate that a noticeable development in the field of lithium oxide and aluminum alloy as two primary substances to produce tritium production. This belong to the completed information

and the performance of many researches in this field that have been used in nuclear reactors to produce tritium. Comparing these compounds, we found that the density of lithium oxide Li_2O was higher than the density of Li-Al. As a result we can use a quantity with large weight and small volume from the oxide compared with the alloy. Moreover, very limited number of reactor channels gives rise to the usage of highly pure lithium oxide in the mono crystal form in the tritium production

3- SAMPLE PREPARATION FOR IRRADIATING

A sample from lithium oxide crystal with purity of about 99% was heated in gloved box to 900 °C for a period of 1-5 hours in vacuum (pressure 10^{-3} pa) or in the presence of dry helium current. This is to remove the impurity which is represented in lithium hydroxide and lithium carbonate. Taking the crystal powder of lithium oxide and compressing to pellet shape by using a pressure of up to approximately 2Mpa in one direction. This is to get one pellet with low density, or pressing it under a temperature of about 900 °C to get a high density pellet. After that, these pellets were aggregated under a temperature of 900 °C for one hour under a dry helium current. Later, we have measured the weight, width and the diameter of the aggregated pellets. There after, these pellets were put in quartz container or stainless steel container isolated with platinum layer to prevent any contact of lithium oxide with the steel[10].

4- IRRADIATION PROCESS

The reactor vertical channels, which were used to irradiate with neutron flux, are shown in figure (1). For each channel in this work we have used neutron flux rate of about $1.67x10^{13}$neutron/cm^2.sec. This is to estimate the necessary quantity of lithium irradiated for one month to get one gram of tritium. For this purpose we have used equation (5)[15]. The obtained results showed that the required quantity equals to 3.2 gm of enriched ^6Li. This quantity is equivalent to about 1437gm of natural lithium oxide supposing that, the percentage of extracted tritium was 100% so that it should use all the vertical channels reactor at the same time. In the above calculation we have taken in our consideration the spontaneous absorption of the radiated samples. Equation(5) shows that the tritium is produced by the reaction of neutron with ^3He obtained by the decay of tritium.

$$He^3 + n \longrightarrow P + T \qquad (5)$$

The cross-section values included in the above equation were found to be 533 barn for thermal neutrons in the reaction with ^3He and 940 barn for thermal neutrons in the reaction with ^6Li [16].

5- TRITIUM EXTRACTION SYSTEMS

Many different systems were used for tritium extraction. They can be classified into the following three types of systems:

1-systems used as blanket for fusion reactors[2,7,13,17]
2-systems used to extract tritium during sample irradiation process in nuclear reactors [4,10].
3-independent systems used to extract tritium from different lithium compounds after being irradiated in nuclear reactors[18-23].

We have studied all the above types of extraction to get an idea about its work nature and the properties of the used materials to put a primary system design for tritium extraction in suitable quantity of about one gram per month.

6- THE SUGGESTED SYSTEM DESCRIPTION

The suggested system; which is shown in figure 2; consists of high purity helium, liquid nitrogen unit needed to purify tritium by removing water, and two ovens. The walls of each oven is composed of three stainless steel layers which allow argon gas to pass through the gap between the second and the third walls. The first oven contains a heater surrounded by a stainless steel vessel in which the irradiated sample was put. However, the second oven contains copper oxide, heated to 500 °C, which oxidized the passing tritium to tritium vapor water. In addition, the first oven was situated in lead cell with a width of about 10 cm. The inlet and outlet of this oven were provided with glass wool stoppers to prevent the escape of the copper particles from the sample with the helium gas current. However, the inlet and outlet of the second oven were provided with stainless steel stoppers.
Moreover, the system contains a trap cooled at liquid nitrogen temperature of (- 196 °C) needed to freeze tritium water passing through it and a vacuum pump required for the continuity of out flow of helium gas. The remaining and leaking tritium gas in side the glove box can be collected and passed through oxidation and retention tube[24] .

7- STEPS OF WORK

A- The sample was irradiated in the first oven; cooled for a suitable period, which depends on the type of impurities; and put in a stainless steel container, where the sample was in touch with thermocouple probe. This have led us to know the temperature of the sample through the experimental steps. Knowing the temperature is important because the rate of tritium liberation depends highly on it. Easley removed stainless steel blanket was used to protect the oven and the probe from corrosion.

B- The oven was then closed and the system has been evacuated until the pressure reached about 5 pa. The vacuum pump was stopped and the helium valve has been closed during this process.

C- The cold trap was filled with liquid nitrogen with increase of copper oxide temperature to about 527 °C in the second oven.

D- The pure helium gas was passed through the system with a constant rate. After that, the sample temperature inside the first oven was regularly

increased until the tritium was liberated in the form of tritium water vapor (T_2 + T_2O) and tritium oxide gases. This took place when the temperature became of range 297-397 °C.

E- The helium valve and the other two valves which are situated at the end of copper oxide oven were closed. There after, the collected tritium is removed and the vessel containing the frozen tritium water was replaced by un empty one. The system is then closed and after that, the helium was pumped again.

F- The temperature of the sample was increased to about 597 C^0 for a period of about one hour to remove all the tritium in the sample.

8- DISCUSSIONS AND CONCLUSIONS

The limited quantity which can be irradiated in the 14th July reactor, needs to select a compound containing on a high density in lithium.

Consequently, we have selected lithium oxide. In addition, according to the primary calculation, we needed 1437 gm of lithium oxide, as a natural lithium to produce one gram of tritium and 130.2 gm of 90% enriched lithium as shown in table 3. Both quantities were irradiated in eighteen channels reactor giving one gram of tritium, whereas, the same quantity of Li-Al alloy of 3% lithium produced only 64 mgm of tritium. Many difficulties were encountered in designing the system required for tritium extraction during the irradiation process using lithium oxide as a primary substance. This belongs to absence of the suitable place in the core of reactor, required for the irradiation of the 1437 gm of lithium oxide. However, such system can be used only with lithium of at least 90% enrichment, specially when we have a pure lithium metal. More over, this system can also be used to extract tritium from lithium metal.

The irradiation period was chosen to be only one month to get tritium during a short period. This is because the irradiation of the substance for long time will increase the cooling period needed for the irradiation of samples. This is specially significant in the presence of impurities such as ^{59}Co which are converting to radioactive isotopes of relatively long half life.

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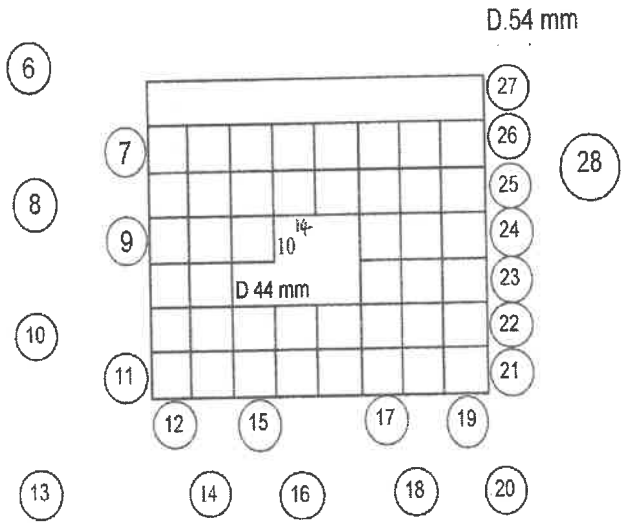
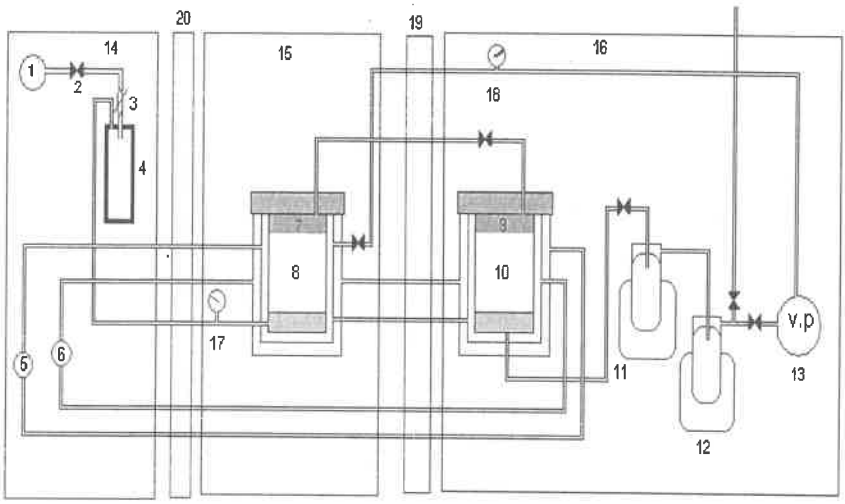
Fixers	Natural lithium	90% Enriched lithium
1. neutron flux rate ($n/cm^2.sec$)	1.67×10^{13}	1.67×10^{13}
2. container dimensions (cylinder) (cm)	40 length and 3.5diameter	40 length and 3.5 diameter
3. total lithium oxide mass required to generate 1 gm from tritium (gm)	1437	130.2
4. number of reactor channels used	18	2
5. irradiating period (month)	1	1
6. irradiate sample cooling period	According to the impurities presented in the sample	According to the impurities presented in the sample

Table (3): Designed fixers in state of enriched and natural lithium.



Channel number (long and dry)	Neutron flux $x10^{13}$
7	0.8
9	1.4
1	0.89
11	0.97
13	0.985
14	1.84
15	2.25
16	2.3
17	2.75
18	1.86
20	1.09
21	1.96
22	2.16
23	2.5
24	2.2
25	1.92
26	1.42
27	0.8

Figure (1) Core cross section of the reactor shows the vertical channels and the neutron flux in the channels.



1- liquid helium container	2- valve	3- flowing measurement
4- purification trap for helium gas	5- circulating pump for argon gas	6- circulating pump for water
7- glass wool stopper	8- sample heating oven	9- steel wool stopper
10- oven contain CuO	11-12- nitrogen trap	13- vacuum pump
14-16- gloves boxes	17,18- pressure scale	19-20- lead protectors

figure (2) Schematic diagram for the suggested system

Physical properties		Values
1	Isotope mass	3.0160492 a.m.u
2	Spin	$1/2^+$
3	Excess energy	14949.794 keV
4	Binding energy	8481.821± 0.004 keV
5	Decay energy	18.59 keV

Table (1) Some of the Nuclide Data of Tritium

Some lithium compounds						
Properties	Li_4SiO_4	Li_2SiO_3	$LiAlO_2$	Li_7pb_2	Li_2O	Li
Melting point C°	1255	1201	1610	726	1570	180
Density (gm/cm^3)	2.39	2.53	2.55	4.6	2.013	0.484
Molecular weight	119.84	89.96	65.91	463.36	29.881	6.94
Density of lithium in compound (gm/cm^3)	0.55	0.39	0.268	0.482	0.931	0.484
Heat conductivity (W/m.k)	2-5.5	2-5.5	-	20	1.7	50

Table(2):Physical properties for lithium and some of its compounds[2,3]