# TRITIUM HANDLING AND RECOVERY SYSTEM FOR ACCELERATOR BASED 14-MEV NEUTRON GENERATOR
## *Inventory Calculation and Tritium Processing Technology selection*

Deepti Dubey
*ITER-India, Institute For Plasma Research*
*Bhat-Motera, Ahmedabad-380005, India*
Email: deepti.dubey@iter-india.org

*Sudhirsinh Vala, and Shishir P. Deshpande*
*Institute for Plasma Research*
*Bhat, Gandhinagar-382428, India*

**Abstract**

An accelerator based 14 MeV neutron source is under development to study the fusion neutronics for Indian fusion programs. The neutrons are generated by impinging 10 mA deuterium beam accelerated up to 340 keV energy over a 140 Curie tritium target. Being a system handling tritium-radio-active material, a recovery system is to be designed to minimize airborne tritium effluent releases to well below the permitted limit. In addition, the system should minimize tritium exposure to staff by maintaining low levels of tritium in the Rotating Tritium Target Holder (RTTH). This paper estimates value of tritium coming out into the exhaust of the Target chamber of the accelerator system. A mathematical model is developed to calculate the amount of tritium getting sputtered out of the target. The calculated result is then successfully simulated using SRIM software, and validated using the experimental results available in the literature. As per our calculation method, tritium release is calculated to be 40 GBq/h which is in very close conformance with the claimed value. Based on this primary calculated data a conceptual design of the Tritium Handling and Recovery System (THRS) is also presented. There are a number of technologies available for THR system like, metal membrane reactors, cryogenic adsorption on molecular sieve beds, getter beds, cryogenic freezing, high temperature electrolysis, and catalytic oxidation. Among all, getter bed technology for the tritium separation is not in frequent practice. This paper elaborates the specific selection technique for choosing the Getter bed system for recovery of tritium from the system.

## 1. INTRODUCTION

Energy production via nuclear fusion requires Deuterium and Tritium (D-T plasma) as fuels. The D-T plasma is produced in the core of the fusion reactor thereby generating 14 MeV neutrons and 3.5 MeV helium. This D-T fusion reaction is very much similar to naturally occurring fusion reaction in the stars with only difference in the temperature of artificially produced D-T reaction. The intensively hot D-T reaction inside the tokamak, may result in different types of mechanical, material and chemical changes during power generation. Therefore, to plan a future fusion reactor, there is a strong need for the development of compatible material to tolerate such high energy neutron induced damages. In order to study the material response in the typical fusion reactor like severe environment due to neutron irradiation, the 'accelerator-based neutron generator' is being developed at IPR, Ahmedabad India. The facility is capable of generating 14 MeV neutron. These neutrons are generated by impinging 10 mA deuterium beam accelerate up to 340 keV energy over a tritium target.[1]

Generally the target used in such experiment is thin film of metals such as Titanium, Scandium, or Zirconium, deposited over Silver, Copper, or Molybdenum substrate. These metals form stable metal hydrides when combined with hydrogen or its isotopes. In fusion related studies we requisite tritium targets, which can be made up of two tritium atoms per metal atom to obtain extremely high density of hydrogen isotope for neutron generation. So, in our neutron accelerator laboratory, we will be handling tritium targets with the activity of 140 Ci each (~ 250 targets) with the ratio of 1.5 Tritium atoms per Titanium atom.

Use of tritium make the work place as activity prone area and demands special care. Exceptional preventive and safety measures are required to keep the shop floor clean. International Commission on Radiological Protection (ICRP) has declared the limit for the maximum permissible tritium concentration in the air, averaged over one year can be $4.8\times10^{-6}$ Ci/m$^3$ [2]. ICRP-30 recommended annual limit of tritium intake to be 0.075 Ci for ingestion as well as for inhalation in the case of radiation-exposed workers. The amount which is being handled at the laboratory of IPR, has a far greater amount compared to the permissible limit inscribed in the radiation safety manuals of the International Commission on Radiological Protection. Conclusively, the laboratory demands a

robust and economical tritium handling and recovery system (THRS) to minimize the air-born tritium effluent release to well below the permissible limit.

## 2. TRITIUM HANDLING AND RECOVERY SYSTEM REQUIREMENT

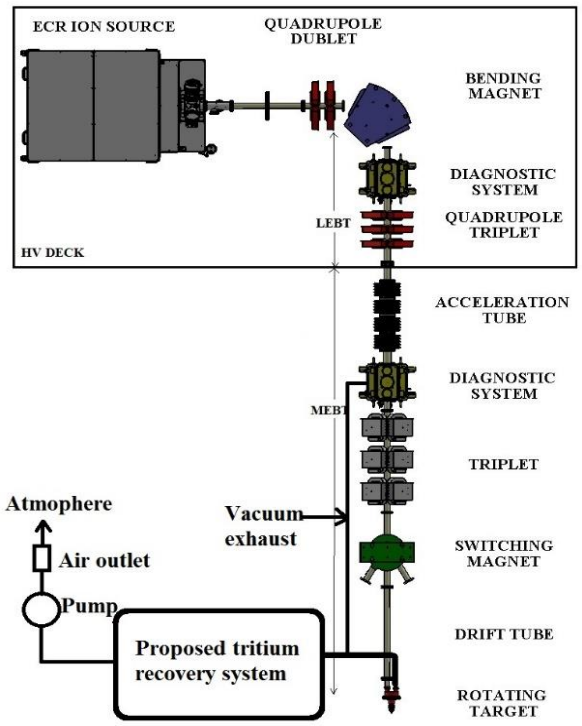



Figure 1: Block Diagram of the system to show the location of the THRS [1].

The accelerator based neutron generator (14 MeV) majorly at gross level consists of, a source from where the deuterium beam originates, a diagnostic system, an accelerator tube to provide high acceleration to the deuterium beam and the rotating target chamber consists of the tritium target. We decided to design THRS at the exhaust of the target chamber (Fig.1) because this region has the high amount of activity due to beam incidence over the metal target. When highly energetic and accelerated beam smashes the surface, two probable events can occur; i) D-T fusion (the main reason to design the project), ii) backward sputtering of the particle from the target plate (Tritium particle is of interest). (i) is required for the experiment to be successful and is the main aim of the whole project. But (ii) is the expected consequence due to (i). Due to this reason there is strong need of the THR system development. Also, this system will play an important role in the case of accidental pressure break-down. To prevent the tritium to come out of the vacuum chamber and enter into the work place (Human exposed area) this system proves to be mandatory. As discussed above there are two likelihoods accountable for the presence of tritium in the gas coming out of the target chamber, i.e. i) during neutron irradiation experiments called, "continuous release" and ii) during an accidental scenario, meaning, due to sudden increment in temperature and pressure of target chamber called "Puff release".

### 2.1. Mathematical Model

For backward sputtering yield of Tritium particles from the target plate due to Deuterium beam bombardment and with the neglect of inelastic losses, Sigmund's results may be written,

$$Y(E) = \frac{0.042}{Uo} \times f\ [\frac{M2}{M1} \times Sn(E)] \quad \text{----------} (1)$$

Where, Y is the total sputtering yield at normal incidence, E an energy of primary particle, U, the surface binding energy, f, the energy independent function of the mass ratio between target (M2,) and projectile (M1) and Sn, the nuclear stopping cross section[3]. The results generated using the Sigmund's equation involves modelling and simulation. But to get the first estimation, hand calculation is performed, which is one of the objective of this report.

When the target plate of the specifications mentioned in the Table-1 below is bombarded with ion beam of Deuterium,

Table 1 –Tritium Target related data used in the calculation

| Parameters | Value |
|---|---|
| Target | Ti-T |
| Ratio of tritium/titanium | >= 1.5 |
| Mass of titanium in the target | 0.01265 gram |
| Half life of tritium | $3.88524\times10^{8}$ seconds |
| Decay constant of Tritium | $1.784\times10^{-9}$ $s^{-1}$ |
| Mass of tritium | $20.747\times10^{-3}$ gram |
| Molar mass of Ti | 47.867 gram |
| Molar mass of tritium | 3.016 gram |
| Target thickness | 1mm |

Table 2 – Deuterium beam

| Parameters | Value |
|---|---|
| Beam type | D+ |
| Beam Energy | 340 keV |
| Beam Current | 10 mA |
| Beam density | 5.65 x $10^{-3}$ A.$cm^{-2}$, 4.493 ×$10^{20}$ atom.$cm^{-2}$ |
| Beam dia | 15 mm |

having a beam energy of 340 keV, and current of 10 m-Amp, it is being assumed that after the generation of the 14 MeV neutrons, some tritium particles are also expected to get sputtered out based on the following calculations.

Using, above data of Table-2, we calculated total number of particle in the incident beam by using,

$I_{Beam} = (q \times N_D \times e) \div t$ ---------- (2)

Where, $I_{Beam}$ = Beam Current, 10mA, q =Charge on the Ion, $N_D$ = Number of the ions in the Beam (Deuterium ions), e = Charge on electron, 1.6 ×$10^{-19}$ coulombs, t = Time, 1 sec.

Calculated, number of the ions in the Beam (Deuterium ions) is to be, $N_D$=6.250×$10^{16}$ particles per sec. The projected beam diameter is given as 0.015 m, with the beam area of 1.766×$10^{-4}$ $m^2$. Thus, estimated bombarded flux will be, Φ =3.539×$10^{20}$ particles. $(sec.m^2)^{-1}$.

To get the total number of possible reactions for the generation of 14 MeV neutron, we need to know the tritium availability on the target source, so using the data from the Table-1 we get the , The number of tritium atoms in the lattice (tritium in target density), NT= 4.143 × $10^{21}$.
Total D-T reaction possible by the Deuterium bombardment on the Tritium Target plate can be calculated using,

No: of reactions, $N_r = N_T \times$ (D-T cross-section at 340 keV) × Φ × t ---------- (3)

D-T cross-section at 340 keV= 5×$10^{-29}$. (Fig-2)

$N_r$ = 7.951×$10^{12}$ = beam particles which are responsible for the generation of 14 MeV neutron.[1]

Out of total ions impinging ($N_D$=6.250×$10^{16}$) the target, only $N_r$ (7.951×$10^{12}$) number of particles are utilised for the generation of neutrons. So, now we know the number of ions which can possibly hit the target and result in the Tritium sputtering out of it.

$N_D - N_r = N_{available}$= 6.250×$10^{16}$ ---------- (4)

Now, the available D-ions in the beam which can result into sputtering of tritium from target lattice will be, $N_{available}$ = 6.25×$10^{16}$ ions. This number corresponds to the amount of Tritium that can sputter out of the target plate.

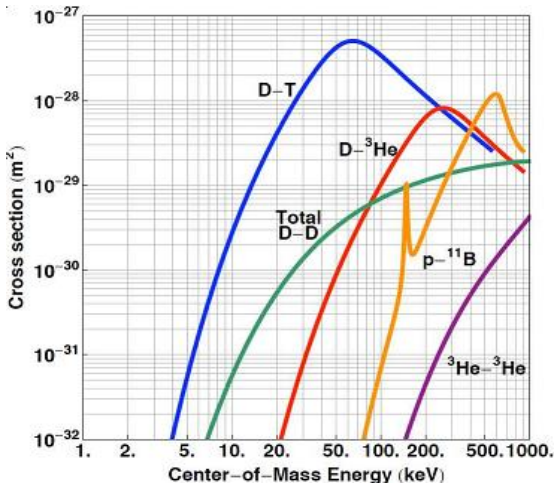



Figure 2. Deuterium –Tritium Cross-section Data. Source: Internet(researchgate.net)

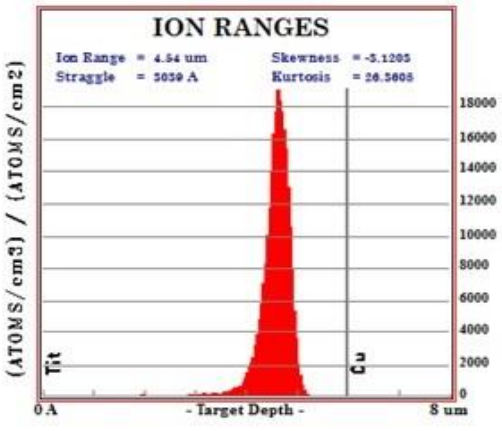



Figure 3-TRIM calculated deuterium ion range in TiT target.[1]

2.1.1. Assumptions

We have considered the tritium activity sputtering out of the target plate corresponding to the beam particles available after the generation of the 14 MeV neutrons, based on the following assumptions,

It is assumed that the beam energy corresponding to the $N_{available}$ particles will be 340 keV. As found in a paper [4] bond energy of the Ti-T molecule is 0.00212 keV/molecule. Bond energy is much less than the beam energy, so it is assumed that almost all the particles in the beam hitting the lattice are capable of breaking the Ti-T bond, thereby resulting in the tritium sputtering.

The target thickness is 1 mm as given in the Table-1, and as interpreted from the graph below, maximum number of beam particles are hitting the target in the range of micro-meter, having almost the same energy intensity. So, it is assumed that as all the Ti-T bond present in this range of 16 micron depth are impacted by the energy of $2.125 \times 10^{19}$ KeV has the equal probability to sputter out.

**2.2. Safety Limit and Internal Exposure Calculations**

To estimate the air activity and internal exposure due to tritium from the 14 MeV accelerator based neutron generator facility it is assumed the humidity of the room is such that all the tritium atoms released into the room atmosphere is exchanged with the hydrogen atom of the moisture present in the room. The evaluation of the airborne tritium concentration and potential internal exposure is carried out for the above mentioned two cases as explained above.

2.2.2. Case-I: Continues release

If the release rate of activity is S (Ci/s) and air removal rate is λ ($s^{-1}$), then the instantaneous airborne concentration ($C_1$) inside a room of volume V is given by Eq- 5.

$C_1(t) = \frac{s}{V\lambda}(1)$ ---------- (5)

The steady state concertation at t = ∞

$C = \frac{s}{V\lambda}$ ---------- (6)

When there is no ventilation or air change the time variation of concentration $C_2$ is given by,

$C_2(t) = \frac{s \cdot t}{V\lambda}$ ---------- (7)

Table- 3 Variation of Concentration with time for different air-change rates

| Time Elapsed (h) | Concentration in DAC | | |
|---|---|---|---|
| | λ=1/ h | λ=2/ h | no air change |
| 0 | 0 | 0 | 0 |
| 0.25 | 147 | 131 | 166 |
| 0.5 | 263 | 212 | 333 |
| 1 | 425 | 293 | 666 |
| 2 | 587 | 335 | 1332 |
| 3 | 642 | 341 | 1998 |
| 4 | 671 | 342 | 2664 |
| 5 | 680 | 342 | 3330 |
| 5.5 | 680 | 342 | 3663 |

Now considering the S = 0.003 Ci/s, V = 2000 $m^3$, the change in concentration is obtained for the two values of λ (λ=1/ h and 2/h) using Eq. 5, and for no ventilation rate the concentration is calculated from Eq. 7.

If there is no ventilation in the room the concentration increases linearly with time. Whereas the periodic removal of air will reduce the concentration and will reach the steady state value after some time. This scenario is clearly inferred from the Table-3. The conversion factor used in the calculation is 1 DAC= $3 \times 10^5$ Bq/$m^3$.

Now we will have a look to the internal exposure, E due to air ventilation and also in air circulation zone. If we integrate Eq- 5 we get,

$$E_1(t) = \frac{s}{V\lambda}\left(T + \frac{e^{\wedge}(-\lambda T)-1}{\lambda}\right) \quad \text{---------- (8)}$$

Also integrating Eq-7, w.r.t time varying from t=0 to T (time of exposure),

$$E_2(t) = \frac{sT^{\wedge}2}{V} \quad \text{---------- (9)}$$

Similar to the above table if we use 1 DAC-h = 0.01mSv of exposure. We can conclude from the calculation that with no air-change as well as with the timely removal of air, the tritium exposure increases with the increase in time. But the difference is that in no air change the value increases with very high magnitudes whereas in the with the air change it increase reasonably.

The exposure can be significantly be reduced by appropriate factor with the usage of proper personal protective equipment.

2.2.3. Case-II: Puff release

Puff release of the total available tritium, i.e. 200 Ci into the room of 2000 $m^3$ volume and surface deposition of the 10% of the activity, the air concentration works out to be 0.09Ci/$m^3$ (1.1 × $10^4$ DAC). Then the internal exposure for puff release will be,

The calculation predicts that the maximum probable internal exposure after 1 h of release will be 6.66 mSv for continuous release (0.003 Ci/s) and for puff release (200 Ci) it is 110 mSv.

Table – 4 Internal Exposure for Puff Release

| Time, h | Exposure(mSv) |
|---|---|
| 0 | 0 |
| 0.25 | 27.5 |
| 0.5 | 55 |
| 1 | 110 |

**2.3. Validation of the Analytical Approach**

*2.3.1. Validation from the experimental data available in the literature*

According to an available literature [2], tritium release from the target at maximum power has been evaluated to be 37 GBq/h experimentally. As per our calculation method detailed out in section, the tritium release is calculated to be ~40 GBq/h which is in very close conformance with the claimed value.

2.3.2. *Validation from the simulation SRIM results*.

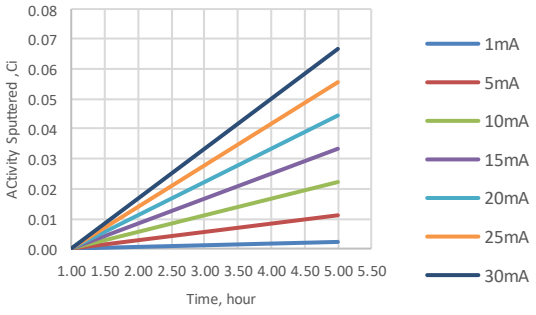



Figure: 5- SRIM Results, Activity Sputtered Vs Beam Current at different time.

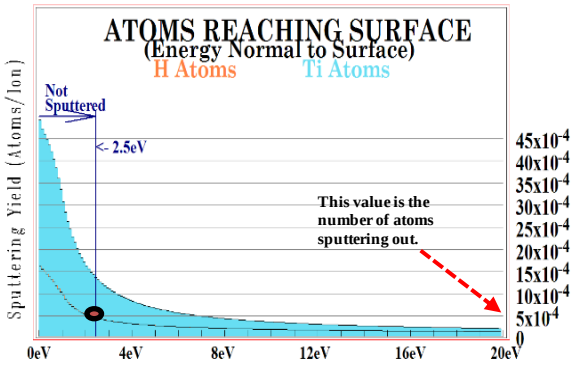



Figure: 4- SRIM, generated data, Energy vs Sputtering yield

We validated the result using SRIM, the sputtering yield is coming out to be as shown in the figure-2 below. We concluded sputtering atoms as $5\times10^{4}$ (Fig-4), and plotted a graph showing sputtered tritium activity with varying time and current (Fig-5). So, Tritium activity corresponding to the value is $1.54\times10^{-6}$ Ci/sec.

Table-5 Calculated and Simulated results

| **Mode of calculation** | **Sputtering rate as per our calculation** | **To cross verify with the published data** |
|---|---|---|
| Mathematical model | 0.003 Ci/sec(DT, Ti, DD, $T_2$) | 40 Gbq/hr |
| SRIM Results | $1.54\times10^{-6}$ Ci/sec (only tritium) | -------- |
| Experimental Results(From Literature) | ---------- | 37 GBq/hr |

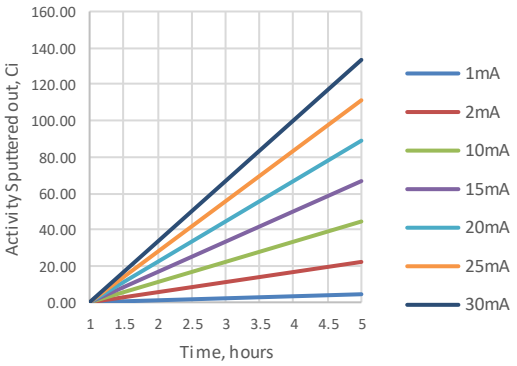



Figure 6- T-Sputtering estimation through Analytical Calculation

## 2.4. Results

It is been assumed, that tritium activity coming out of the lattice will be corresponding to the $N_{available}= 6.25\times10^{16}$ ions, i.e. 0.003 Ci/sec. From Fig-6, one can easily interpret the relation between Sputtered T-activity with variable time and current. This result in close conformance with the data published in the [5] and as shown in Table-5 below.

## 3. TECHNOLOGY SELECTION FOR THE TRITIUM HANDLING AND RECOVERY SYSTEM

There are number of candidate technologies available for the tritium separation like Metal Membrane Reactors, Molecular Sieve, Getter Bed and Cryogenic Freezing. To select the best among them there is a need of a fair enough method. As discussed in a ref. [6], we will be using Pairwise Comparison approach to find the most important design criteria to choose the best separation technology for THRS.

Table -6 Pairwise comparison matrix to select the technology selection criteria.

| Techniques | Capital Expenditure | Operational Expenditure | Safety and Enviornment | Continuous Processing Possible | Easily Modify Purity | Throughput | Technology Readiness | High Purity | Low Tritium Inventory | Separation Factor of a Single Step |
|---|---|---|---|---|---|---|---|---|---|---|
| Capital Expenditure | | 2 | 2 | 0 | 2 | 2 | 0 | 2 | 2 | 2 |
| Operational Expenditure | 0 | | 2 | 0 | 1 | 2 | 2 | 2 | 2 | 2 |
| Safety and Enviornment | 0 | 0 | | 0 | 0 | 0 | 0 | 0 | 1 | 0 |
| Continuous Processing Possible | 2 | 1 | 2 | | 2 | 2 | 2 | 2 | 1 | 2 |
| Easily Modify Purity | 1 | 1 | 2 | 0 | | 2 | 2 | 2 | 1 | 2 |
| Throughput | 1 | 0 | 2 | 0 | 0 | | 2 | 2 | 1 | 2 |
| Technology Readiness | 1 | 1 | 2 | 1 | 1 | 1 | | 0 | 0 | 1 |
| High Purity | 2 | 1 | 2 | 0 | 1 | 1 | 1 | | 0 | 1 |
| Low Tritium Inventory | 0 | 0 | 2 | 0 | 1 | 2 | 2 | 1 | | 2 |
| Separation Factor of a Single Step | 0 | 0 | 1 | 0 | 1 | 1 | 0 | 1 | 0 | |
| **Rank weighting** | 7 | 6 | 17 | 1 | 9 | 13 | 11 | 12 | 8 | 14 |

After selecting the weightage of different selection criteria, we made a Pugh Matrix, to rank different technologies and make a selection of the best out of it.

As it is clear from the above Table-7 Getter Bed technology gained the highest score out of the others. So based on the above results we select getter bed technology to develop the THR system. Although this technology is not commonly used for hydrogen isotope separation.

Chemical Getter bed is nothing but a deposit/ coating of reactive material that is placed inside a vacuum system. When gas molecules strike these reactive getter material, they combine with it chemically or by absorption. The getter material selected for the operation should necessarily have a large gas-solid mass transfer coefficient for

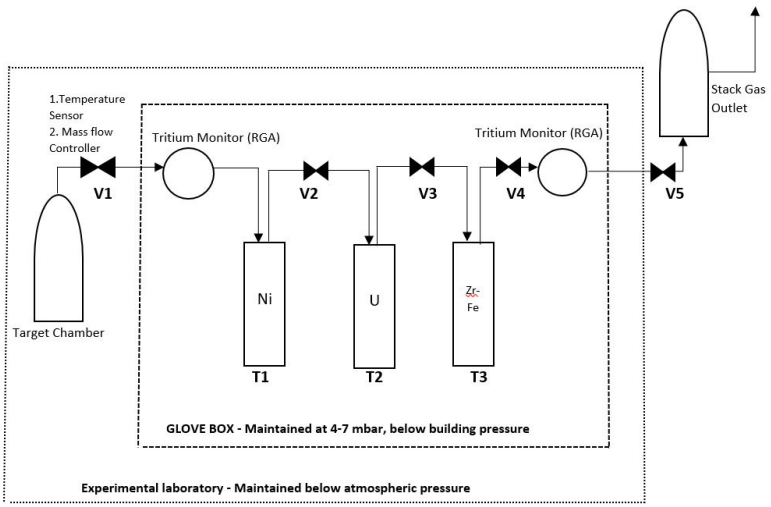


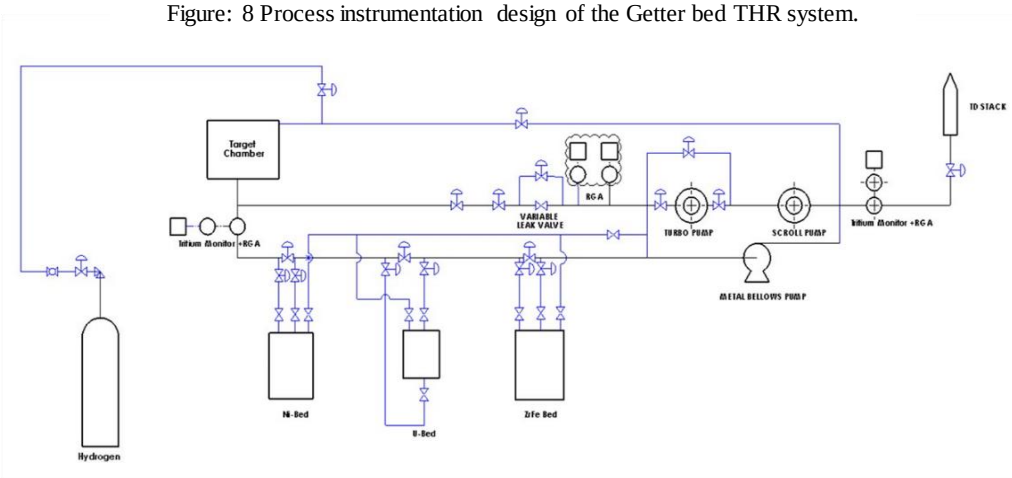

Figure 7: Block Diagram of Getter Bed system

scavenging hydrogen isotopes from gas mixture. The outline of the process implied for removal of tritium from the gas mixture is as shown in Fig-.7 The block diagram of the proposed model of the THRs system and Tritium flow pattern in the THRS is detailed out in the figure. Also we have started the fabrication and procurement process. It is expected to be completed at the early of 2020. A P&ID of the complete getter bed system is shown Fig-8 showing Process instrumentation.

Table- 7 Pugh Matrix for the technology selection.

| Techniques | | Pd-Ag Permeator | Cryogenic Adsorption on Molecular Sieve | Getter Bed | Cryogenic Freezing | |
|---|---|---|---|---|---|---|
| Criteria | **Weightage** | | | | | **good --> 1** |
| Capital Expenditure | 7 | 1 | 4 | 2 | 2 | **better --> 2** |
| | | 7 | 28 | 14 | 14 | |
| Operational Expenditure | 6 | 1 | 2 | 4 | 2 | **best --> 3** |
| | | 6 | 12 | 24 | 12 | |
| Safety and Enviornment | 17 | 3 | 3 | 4 | 2 | **Excellent --> 4** |
| | | 51 | 51 | 68 | 34 | |
| Continuous Processing Possible | 1 | 1 | 1 | 3 | 4 | |
| | | 1 | 1 | 3 | 4 | |
| Easily Modify Purity | 9 | 1 | 3 | 4 | 4 | |
| | | 9 | 27 | 36 | 36 | |
| Throughput | 13 | 2 | 3 | 3 | 3 | |
| | | 26 | 39 | 39 | 39 | |
| Technology Readiness | 11 | 4 | 3 | 3 | 3 | |
| | | 44 | 33 | 33 | 33 | |
| High Purity | 12 | 4 | 2 | 3 | 2 | |
| | | 48 | 24 | 36 | 24 | |
| Separation Factor of a Single Step | 14 | 3 | 2 | 3 | 2 | |
| | | 42 | 28 | 42 | 28 | |
| **Rank weighting** | 360 | 234 | 243 | 295 | 224 | |
| **Percentage** | | **65** | **68** | **82** | **62** | |

Figure: 8 Process instrumentation design of the Getter bed THR system.



## 4. CONCLUSION AND DISCUSSION

The value calculated using the mathematical model is in conformity with the literature available, but the SRIM resulted into very low value of sputtering per sec (As shown in Table-5). One of the reason may be that in the analytical calculation we are talking about the total sputtered atoms in the exhaust, whereas the SRIM is specifically stating the Sputtered tritium atoms, which is expected to be low logically. But to get more specific and correct results we requires further research and experiments.

In the planned activities, we will performing certain experiments with system to get the actual sputtering rate and checking the number by using Residual Gas Analyser. We have planned to use the target plates with known density and tritium activity. The results to this experiment will be further discussed in the next paper which will specifically explain the, experiment results and exhaust gas composition and Getter bed design and process parameters.

At present Getter bed technology is being used for storing the Tritium in the applications related to fusion community as well as in Fission. But they could have more utility. If we inject tritium and $N_2$ to these beds at room temperature, they could collect the tritium and leave the $N_2$ in the gas phase for disposal theoretically. This would be a nice, simple, fast separation system. TSTA actually used this method, but did not quantify its performance. One worry is that uranium will nitride. So, through this experimental facility the idea is to show that the bed capacity will not degrade over time. Also, the separation efficiency would be characterized.

## ACKNOWLEDGEMENTS

We would like to acknowledge for the valuable inputs of Mr. S.C. Parida, Head, PDS, RC&I Group, BARC. We are grateful to him for taking time to review the some part of work presented in this particular paper. All your valuable comments and suggestions added value to our work. We also like to thank IPR research scholar Ms. Priti Kanth and Mr. Mayank Rajput for their valuable suggestion and discussions in the sputtering calculation and to infer the results from the SRIM Simulation software respectively.

## REFERENCES

[1] S. Vala, M. Abhangi, R. Kumar, B. Sarkar, and M. Bandopadhyay, "Rotating tritium target for intense 14-MeV neutron source Rotating tritium target for intense 14-MeV neutron source," *Fusion Eng. Des.*, vol. 123, no. June, pp. 77–81, 2017.

[2] R. Lässer, "Tritium in metals," *Zeitschrift fur Phys. Chemie*, vol. 143, no. 143, pp. 23–49, 1985.

[3] J. Bohdansky, "A universal relation for the sputtering yield of monatomic solids at normal ion incidence," *Nucl. Inst. Methods Phys. Res. B*, vol. 2, no. 1–3, pp. 587–591, 1984.

[4] Y.-R. Luo, "Bond Dissociation Energies," *Q. Rev. Chem. Soc.*, vol. 9, pp. 65–97, 2009.

[5] M. Martone, M. Angelone, and M. Pillon, "The 14 MeV Frascati neutron generator," vol. 215, pp. 1661–1664, 1994.

[6] C. Day, B. Butler, T. Giegerich, P. T. Lang, R. Lawless, and B. Meszaros, "Consequences of the technology survey and gap analysis on the EU DEMO R&D programme in tritium, matter injection and vacuum," *Fusion Eng. Des.*, vol. 109–111, pp. 299–308, 2016.