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Manufacturing Process of Self-Luminous Glass Tube (SLGT) Utilizing Tritium Gas: Design of Tritium Handling Facilities

Kyeongsook Kim†, KwangSin Kim, Soon Hwan Son and Wi-Soo Kim

Nuclear Power Laboratory, Korea Electric Power Research Institute (KEPRI),
103-16 Munji-Dong, Yusung-Gu, Daejeon 305-380, Korea
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Abstract–Tritium, the essential material of self-luminous glass tube (SLGT), is a β-ray emitting radioactive hydrogen isotope that requires a special handling facility. The design basis of a tritium handling facility is to minimize the operator's exposure by tritium uptake and the emission of tritium to the environment. To fulfill the requirements, major tritium handling components are located in the secondary containment such as glove boxes (GBs) and/or fume hoods. Besides a tritium recovery system (TRS) and a tritium monitoring system are included in this facilities. To prevent both tritium release out to the room and air in-leakage into the GB, the GB is designed to maintain equal or slightly lower pressure than room atmosphere. TRS is connected to the GB and process loop (PL) to minimize the release of tritium as well as to remove moisture and oxygen in the glove box. TRS is composed of a molecular sieve adsorption bed (MS bed), a nickel catalyst bed, a metal getter, tritium monitors, and a circulation pump. TRS components are regenerated if needed and tritium is recovered for reuse.

Key words: Tritium, Getter, Zr-Fe Alloy (ST 198), Tritium Recovery System (TRS), Uranium Bed, Nickel Catalyst Bed, Molecular Sieve Adsorption Bed

INTRODUCTION

Unlike other hydrogen isotopes, tritium (^{3}H) is a radioactive hydrogen isotope emitting β-rays. Due to the considerably low energy β-ray emission, a small amount of tritium is being used for general industries without any special precaution. However, a quite large amount of tritium such as hundreds of Curies (1 Curi=3.7×10^{10} Bq) or more should be handled with special cautions [Caffarella et al., 1980]. Since KEPRI started the tritium utilizing project funded by Electric Power Industry Foundation Fund Project (Electric Power Industry Technology Evaluation and Planning), two commercially available self-luminous glass tube (SLGT) products have been chosen for our research [Chung et al., 1994; Kim et al., 2001; McNair, 1991]. To produce SLGTs, high purity tritium gas is injected into the glass tubes and then the tubes are sealed and cut. In this project, as much as one thousand Curies of tritium gas will be used. As tritium from Wolsong Tritium Removal Facility (WTRF) is commercially available, tritium may need to be repackaged into unit amounts from 500 kCi tritium storage vessel. However, no tritium handling facilities to treat large amount of tritium have been available in Korea even though WTRF will have its own tritium gas handling and storage system (TGHSS), which will not be ready to operate until late 2005. Thus, we are developing tritium handling facilities with design basis of minimizing the operator's exposure by tritium uptake and emission of tritium to the environment. The tritium handling facilities including tritium recovery system (TRS), to recover tritium from SLGT specific process components such as a main process loop (PL), a shock test container, and a defective tube container will be also used in other research which require handling large amounts of tritium.

The three design concepts of the facilities are minimization of tritium release, maximum recovery of released tritium, and monitoring of tritium release into the environment. First, to minimize the operator's exposure by tritium uptake and emission of tritium to the environment, tritium release should be kept as low as possible [Sinclair, 1979]. To accomplish this goal, several provisions are adopted. Secondary enclosures such as glove boxes (GBs) and/or fume hoods are used for equipment handling highly concentrated tritium. Therefore, pipes, tubes, containers, valves, and pumps in the main PL to produce SLGTs are not only designed to be leak-tight for primary enclosures of tritium, but also placed in secondary enclosures to prevent accidental tritium release into the room. In addition, the inside pressure of GB will be kept at equal or slightly lower pressure to room atmosphere to minimize both tritium release out of GB and air in-leakage into GB. If tritium concentration in GB becomes higher than a pre-set level, TRS will operate and recover tritium before GB atmosphere is purged to the building exhaust. Second, for maximum recovery of tritium, an efficient TRS is required. TRS consists of a molecular sieve adsorption bed (MS bed) to eliminate moisture, a nickel catalyst bed to decompose organic material including triated water molecule, and a metal getter to recover tritium [Lasser, 1989]. Pipes, tubes, containers, valves, and pumps in TRS should be also designed to be leak-tight. Third, to monitor the release of tritium into the environment, several tritium monitors (TMs) and sampling bubblers are placed in the laboratory or at the stack.

SYSTEM DESIGN

A conceptual design drawing of the tritium handling facilities is shown in Fig. 1. The facility is divided into GB, TRS and the exhaust

†To whom correspondence should be addressed.
E-mail: kskim@kepri.re.kr

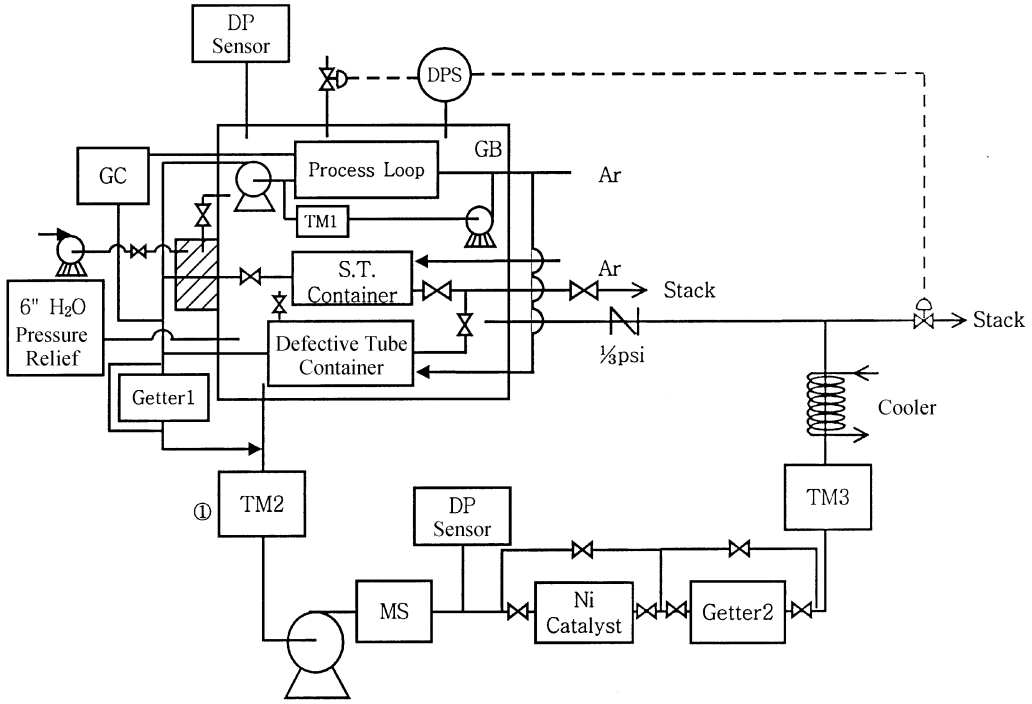

Fig. 1. A conceptual design drawing of the tritium handling facilities.

system, which consists of exhaust blowers and TMs.

1. GB and PL

As shown in Fig. 1, main PL to manufacture SLGT, a high vacuum pump, and quality assurance (QA) equipment including a shock test container and a defective tubes container are placed in GB.

Several inert gases were considered for GB atmosphere: helium, nitrogen, and argon. Of these, argon has been chosen since nitrogen degrades the performance of metal getters and helium is more expensive than argon.

Moisture, which also deteriorates the phosphor, should be eliminated. The oxygen/moisture removal system (not shown) and TRS are connected to GB; ante-chamber (shaded area in Fig. 1) is attached to GB to transfer things such as glass tubes, SLGTs produced, and/or other small items into or out of GB.

To monitor and control GB atmosphere, an argon supply system, a dew point sensor, an oxygen analyzer, a differential pressure switch (DPS) with the adjunctive pneumatic valve were installed. The argon supply system injects argon into the GB when the GB atmosphere becomes below pre-set lower limits of DPS, while the pneumatic valve is opened automatically by DPS. GB atmosphere is purged to the building exhaust when over pre-set upper limits of DPS. The oxygen/moisture removal system should be kept operating while GB is in use to maintain the oxygen and the moisture level of GB atmosphere at a very low level. An H_2O pressure relief device to protect the structural integrity of GB in case of hydrogen explosion is also installed.

During the processing of SLGTs in PL, the exhaust from PL containing residual tritium is sent to TRS via Getter 1 if necessary. And also, the QA check equipment is connected to TRS via Getter 1 to purge exhaust gases.

1-1. Main PL

Fig. 2 is the conceptional design drawing of the main PL. The purchased tritium gas moves into the uranium bed of the tritium storage vessel (TSV) from the special Uranium-bed shipping container (USC), in which the band heater with temperature controller is attached to outer shell of the USC. All tritium in USC is transferred to TSV by heating USC. By heating TSV, the required amount of tritium is supplied to glass tubes through the manifold. TSV is sealed in a secondary container to reduce the leakage of tritium. The space between TSV and the secondary container is evacuated to remove potentially leaked tritium and evacuated gas is sent to TRS via Getter 1. Uranium has been chosen as the tritium storage metal due to the low risk of tritium leakage at room temperature and the high tritium storage capacity. Due to the low equilibrium tritium pressure of uranium tritide at room temperature, uranium can reduce the chance of tritium leakage and sometimes uranium is used as a vacuum pump to remove tritium from manifolds in other applications.

Prior to the first supply of tritium to glass tubes from TSV, tritium is analyzed by gas chromatograph (GC). The effluent from the GC is sent to Getter 1. The remaining tritium gas in PL after gas filling into the glass tubes is sucked into TSV, which has been back to room temperature. When PL pressure comes down to about 5 torr, a circulation pump is activated to accelerate the absorption of tritium in uranium bed of TSV until the tritium concentration is close to equilibrium pressure. Since a circulation pump cannot operate under low pressure, argon is supplied to the loop before the pump is activated. Argon with residual tritium after tritium absorp-

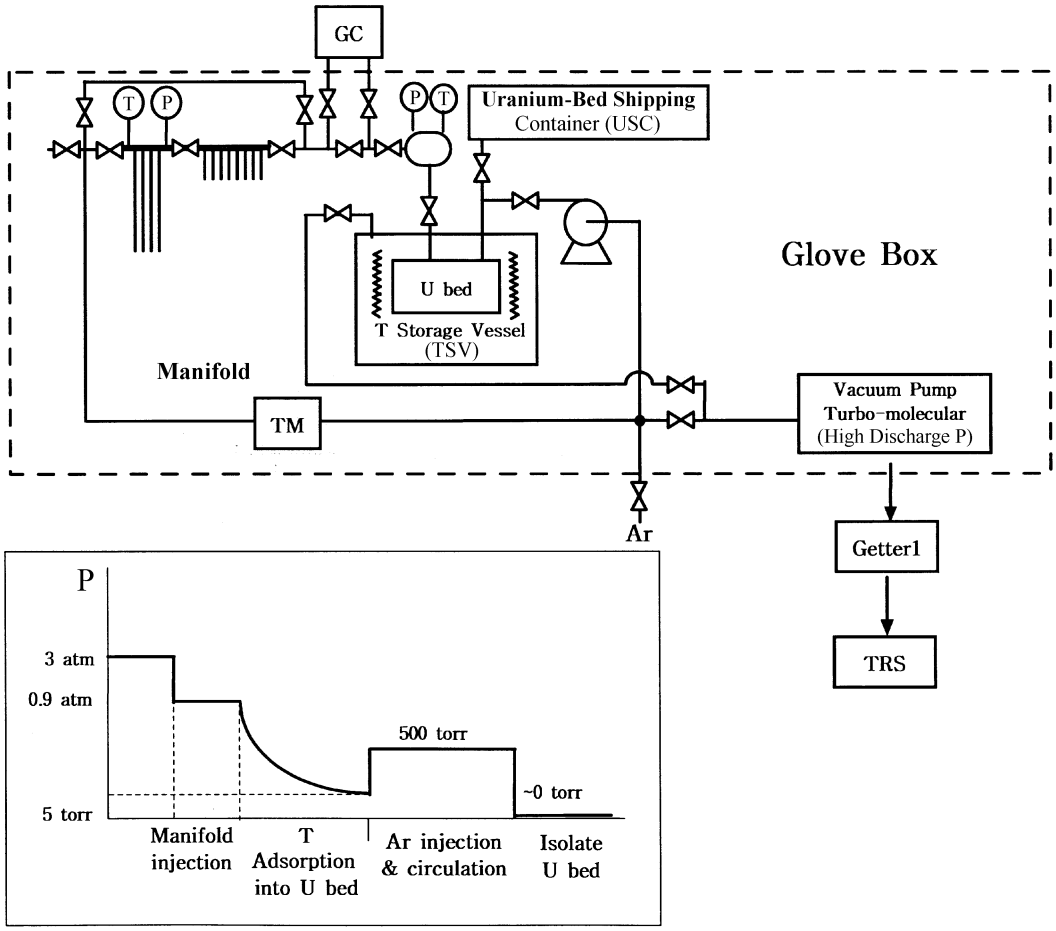

Fig. 2. A conceptual design drawing of the main process loop (PL).

tion is evacuated to TRS via Getter 1 with a turbo-molecular pump. The concentration of tritium in PL is monitored by TM.

1-2. Quality Assurance (QA) Check Equipment

The tritium-filled and sealed SLGTs will be tested for QA of final products. Thermal stress remaining in sealing area can cause the premature fracture of the tubes even by slight shock. The existence of thermal stress is examined by quenching the tubes in a very cold liquid such as liquid nitrogen. Since the gas from shock tests has to go through Getter 1 and TRS to remove potential tritium in the stream, liquid nitrogen may not be used, because nitrogen deteriorates metal getters as discussed above. Thus liquid argon, which has a similar boiling point to nitrogen will be used in shock tests. Defective SLGT rejected in QA tests are destroyed in a TSV and tritium in the tubes is recovered for reuse. TSV is purged with argon. Argon mixed with tritium from the container is sent to TRS via Getter 1.

1-3. Choice of Getter Material

Among metal hydride forming materials, uranium which is adopted in our PL for tritium storage and Zr-Fe alloy (SAES getter, ST 198) getter which is being used in Darlington Tritium Removal Facility and Kinectrics (former research division of Ontario Power Generation in Canada) was finally selected for our getter material. These materials are widely used and their safety and efficiency in tritium handling facilities are well established [Shmayda et al., 1990; Shmayda and Kherani, 1989; Folkers and Singleton, 1975; Kherani et al., 1987; Kherani and Shmayda, 1989; Nobile et al., 1994].

The advantage of uranium is that the reaction between U and tritium at room temperature is so fast that a large amount of tritium can be eliminated effectively without heating the getter. However, the minimum efflux concentration is 2 mCi/m^3, a relatively high value for a tritium getter, although it may vary depending on flow rate and tritium concentration. Furthermore, there are some other disadvantages to using uranium as a tritium getter. Uranium is a nuclear regulated material, so it is required to report its inventory to the government regulatory organization every year. And uranium has a tendency to be fragmented to submicron particles, which makes it difficult to keep uranium particles inside the getter, and uranium particles may spread in PL and cause clogging damage to the components.

On the other hand, a Zr-Fe alloy getter has a moderate tritium storage capacity, and a slower reaction rate than U and tritium, so that the getter should be heated more than 200 °C to remove tritium from gas stream. And unlike U where the equilibrium pressure of tritium remains constant at a temperature, the equilibrium pressure of tritium in Zr-Fe alloy increases with increasing of tritium to alloy ratio at the same temperature. However, tritium concentration in gas stream can be lowered to the order of 10^0 $\mu Ci/m^3$, which is sufficiently low to be released to the environment, if the tritium to getter material ratio is kept low.

The responsibility of metal getters in our system is minimizing the tritium release to the environment. Thus, Zr-Fe alloy ST198 (SAES

Getters, Italy) is selected over U as a getter material for Getter 1 as well as for the getter in TRS for its low equilibrium pressure even though its reaction rate is slow and heating is required for its operation.

2. TRS

As shown in Fig. 1. TRS is composed of a circulation pump, MS bed, Ni catalyst bed, a tritium getter, a couple of TM and a dew point sensor [Hejes and Shmayda, 1995; Shmayda et al., 1992]. MS bed and Ni catalyst bed are placed before the tritium getter (Getter 2) to protect the getter metal by removing moisture and decomposing organic compounds, respectively. Although Ni catalyst bed can eliminate a small amount of oxygen, its major function is to decompose tritiated organic compounds. The tritiated organic compounds are formed by the reaction of tritium in GB atmosphere with organic components of GB such as gloves and rubber gaskets. These hard-to-eliminate tritiated organic compounds not only deteriorate the metal getter but also easily deposit to the laboratory equipment surfaces, which makes them the principal contaminant of the laboratory.

TRS should be operated continuously when GB is in use. TM 2 at the entrance of TRS loop monitors the tritium concentration of GB and lets the inflow bypass the getter when tritium concentration is low. The getter should be in stand-by mode by heating it to operation temperature even though it is bypassed. When tritium concentration in GB is higher than pre-set values, TRS is in a normal operation mode and inflow passes through the beds and the getter. The effluent from the getter is cooled down to room temperature and its tritium concentration is monitored by TM 3. By comparing the readings of TM 2 and TM 3, the efficiency of Getter 2 can be monitored. If Getter 2 does not work properly, it should be regenerated. The operation of the MS bed can be monitored with dew point sensor in TRS and in GB. The regeneration of MS bed, Ni catalyst bed, and getter can be performed in-situ or off-line. A conceptual design drawing of the off-line regeneration system is presented in Fig. 3.

The moisture evaporated from the MS bed is collected in the collector and the dried gas is exhausted to the stack through a drier and a tritium monitor. Ni catalyst bed is regenerated with a mixture of 96% Ar and 4% H_2 for about 2 hours. The moisture from the bed is also sent to the collector. Transferring tritium gas from the heated metal getter directly to the U bed is usually not a proper method. With the valves to U bed closed. Helium gas is injected in the loop and tritium from the metal getter at 550 °C is transferred to the MS bed operating at 77 K. After the transfer is completed, the metal getter is isolated from the loop. Tritium in the MS bed is transferred to the U bed by heating the MS bed to 120 K. When tritium concentration reaches a sufficiently low level, the valves to the U bed are closed and the MS bed is heated up to 350 °C to discharge any tritiated species. The residual gas is sent to the collector through a small Ni catalyst bed.

CONCLUSION

A conceptual design of the tritium handling facilities was completed for handling of several thousands Curies or more of tritium. The design basis of the facilities is to minimize the operator's uptake and the emission of tritium to the environment. The three major components of the facilities are secondary enclosures for tritium processing equipment, a tritium recovery system, and a tritium monitoring system. Argon has been chosen for GB atmosphere in consideration of the risk of hydrogen explosion, cost, and effects on tritium getter materials. In addition to TRS, moisture/oxygen removal equipment is attached to the GB, which is operated at equal or slightly negative pressure than the room atmosphere. TRS is in operation all the time when the GB is in use to reduce the risk of tritium release and to recover the released tritium. Zr-Fe alloy (ST 198) is used as the tritium getter material. The laboratory and the stack are monitored on tritium concentration level with both tritium monitors and compliance bubbling bottles in on-line basis. At present, we are designing the tritium monitoring systems including passive sampling bottles to monitor room atmosphere and stack gas flow.

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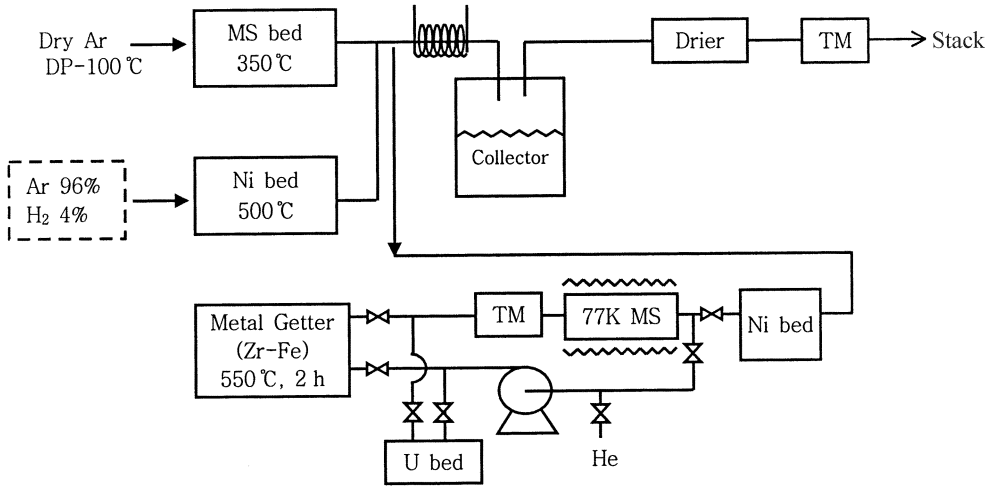

Fig. 3. A conceptual design drawing of the regenerating system.

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REFERENCES

Caffarella, T. E., Radda, G. J. and Dooly, H. H. Jr., "Miniature Radioactive Light Source and Method of its Manufacture," US Patent, 4,213,052 (1980).

Chung, H., Ahn, D. H., Paek, S. W., Koo, J. H., Kuk, I. H., Lee, H. S. and Kim, K. R., "Use of Tritium in the Production of Self-luminous Compounds," KAERI/AR-411 (1994).

Folkers, C. L. and Singleton, M. F., "Collection of Deuterium on a Uranium Getter During Dynamic Flow Conditions," UCRL Report 76734, Lawrence Livemore National Laboratory (1975).

Hejes, A. G. and Shmayda, W. T., "Design of a Second Generation Secondary Enclosure Clean-up System," *Fusion Technology*, **28**, 1509 (1995).

Kherani, N. P., Shmayda, W. T. and Jalbert, R. A., "Tritium Removal from Inert Gases Using Zr_2Fe," 12th, SOFE, IEEE87CH2507-2, 1239-1242 (1987).

Kherani, N. P. and Shmayda, W. T., "Zr_2Fe Tritium Scavenger Beds," Ontario Hydro Research Division Report, # 89-199-k (1989).

Kim, W.-S., Jung, Y.-G., Kim, K., Lee, S.-K. and Song, K.-M., KEPRI Report, TM.00NP34. T2001.175 (2001).

Kim, W.-S., Son, S.-H., Kim, K., Lee, S.-K. and Song, K.-M., "Development of Tritium Removal Technology (I)," 96NJ18 KHNP Report (2002).

Lasser, R., "Tritium and Helium-3 in Metals," Springer-Verlag, New York (1989).

McNair, R. C., "Self-luminous Light Source," US Patent, 4,990,804 (1991).

Nobile, A., Mosley, W. C., Holder, J. S. and Brooks, K. N., "Deuterium Absorption and Material Phase Characteristics of Zr_2Fe," *J. Alloys and Compounds*, **206**, 83 (1994).

Shmayda, W. T., Hejes, A. G. and Kherani, N. P., "Comparison of Uranium and Zirconium Cobalt for Tritium Storage," *Journal of the Less-Common Metals*, **162**, 117 (1990).

Shmayda, W. T. and Kherani, N. P., "Uranium for Hydrogen Isotope Removal from Inert Gas Streams," *Fusion Engineering and Design*, **10**, 359 (1989).

Shmayda, W. T., Kherani, N. P., Wallace, B. and Mazza, F., "Inert Gas Secondary Enclosure Clean-up System," *Fusion Technology*, **21**, 616 (1992).

Sinclair, K. W., "Tritium in the Environment," NCRP Report, 62 (1979).

Son, S.-H., Song, K.-M., Kim, K., Lee, S.-K. and Kim, W.-S., "Development of Tritium Removal Technology (I)," TR 96NJ18.J1998.45, KEPRI (1998).