RADIOACTIVE WASTE FORMATION DURING HANDLING TRITIUM AND ITS COMPOUNDS

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ABSTRACT

Regularities of radioactive waste formation during tritium and metal tritides handling were experimentally researched. There were identified: all types of the waste should be considered as radioactive ones, the volume of gaseous waste is greater than that of liquid and solid waste by 10^6 - 10^7 times, and the gas contains about 90% of the total tritium activity of the waste.

Up to 50-70% of the total tritium activity in the exhausted gases are in the tritium oxide form, so tritium oxide in the exhausted gases should be considered as the main factor of radiation hazard for the public and the environment. Technological equipment is the source of tritium and its oxide ingress into the atmosphere.

Tritium and tritium oxide removal systems have been developed at VNIIEF to recover tritium in effluent gases. The systems are based on tritium catalytic oxidation and tritium oxide sorption by synthetic zeolites. The efficiency of tritium recovery of the effluent gases is greater than 99.9%.

Implementation of the process at the RFNC-VNIIEF tritium facility permitted reduction of tritium releases to the environment by 200 times. Technologies for tritium-containing liquid and solid waste reprocessing have been developed and operated in the production practice.

The cost effectiveness of the technologies operated is assessed over $1,000,000 (U.S.) per year.

INTRODUCTION

A simplified flow-chart of tritium (T) through its “life cycle” for all of the technologies is illustrated in Figure 1. It is implied that T can be used (or presented in technological processes) in a gaseous form or as tritium oxides, metal tritides, or organic compounds. Gaseous T is released from the metal tritides as well as organic compounds during technological operations. So, independently of the used T-form, all types of the technological operations should be executed within containment (glove boxes, chambers, containers), filled with different types of gaseous media (air, argon, nitrogen etc.), or in evacuated vessels (Belovodski, Grishmanovski, 1985) (see Figure 2).

A characteristic features of T is its ability to convert to its oxide form (HTO) by self-radiation in the presence of oxygen, hydroxide or oxygen containing compounds (Belovodski et al, 1973, Failor et.al., 1986, Golubev et al., 1994). It is well known that HTO is more toxic than T by »10 000 times. T and its oxide are sorbed rapidly by the structural materials (SM) and permeate through SM due to diffusion and effusion. Thus, for all T handling technologies (Figure 1) a common feature is T interaction with gas media and composite materials (metals, plastics, glasses) of technological equipment. This T-feature results in gaseous, liquid and solid waste formation during T and T-compound handling (Belovodski et. al., 1985, Gaevoy et. al., 1973).

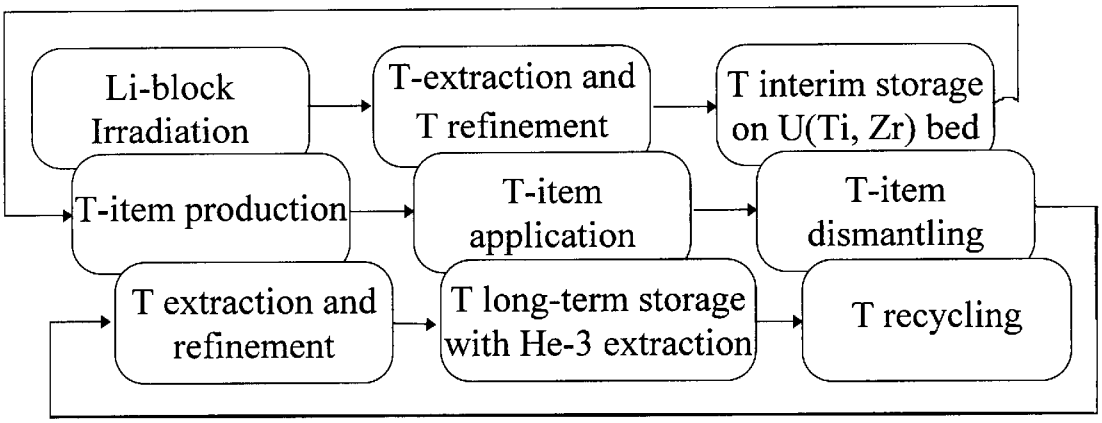

Figure 1 Simplified flow chart of tritium through its "life-cycle"

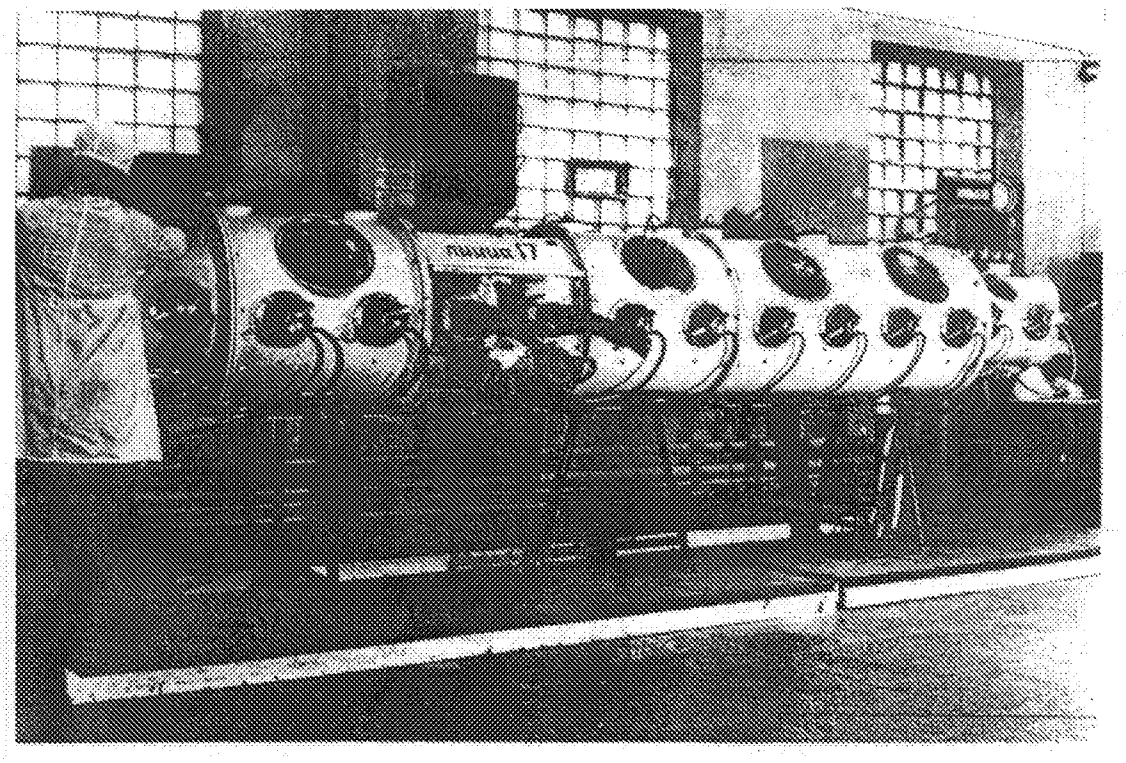

Figure 2 Double-walled glove box to handle tritium

REGULARITIES OF RADIOACTIVE WASTE FORMATION DURING T-HANDLING

According to current Russian Protection Regulations liquid wastes are classified as radioactive at specific activity greater than $4*10^{-6}$Ci L^{-1} (1Ci = $3.7*10^{10}$Bq), solid waste - at specific activity more than $2*10^{-6}$ Ci kg^{-1}. There is no regulations for gaseous waste, however, as well as in the case of liquid waste, gaseous waste can be considered as radioactive at specific activity more than $2*10^{-6}$Ci L^{-1} for T and $3*10^{-10}$ Ci L^{-1} for HTO. Regularities of radioactive waste formation were researched at the RFNC-VNIIEF during processing of a units, containing large amounts of T. The research results are listed in Table 1. From the

data presented it follows that all types of waste of tritium facility should be regarded as radioactive. By the way, the volume of gaseous waste is about 10^6 - 10^7 times greater than those of liquid and solid waste, respectively. It was found experimentally that up to 50-70% of the total tritium activity in exhausted gases are in the HTO form (Belovodski, Gaevoi et. al., 1994). Taking into consideration the much higher toxicity of T in its oxide form, it should be regarded that HTO in the exhaust gases present the main radiological hazard for public and environment.

Table 1 Radioactive waste formation during tritium-containing units production

Waste Type	Waste Volume, relative units	Relative amount of activity %	Specific activity, Ci/l
Gaseous	$3*10^7$	26.4	$3*10^{-8}$
Liquid	51	1.8	0.001
Solid	1	71.8	2.5

Notes: */Tritium release in the atmosphere after T-removal system.

Volume of gaseous waste is ~ 10^6 Ci/year, or over 90% of the total tritium activity in the waste.

The contribution of different sources of T-release to the stack plume gases were researched at VNIIEF during operation of the tritium facility. The research results are presented in Table 2. It can be seen from data of Table 2 that the production equipment (technological effluent gases) is the main source of tritium emissions to the atmosphere. Here it is defined that up to 90% of activity tritium in the effluent gases is presented in the HTO form.

Table 2 Contribution of different sources to gaseous waste

Emission Source	**T-amount released %**
Pumping-out of technological equipment	**15**
Opening of equipment, containers	**42**
Heating of uranium beds	**27**
Incidental release	**10**
General ventilation system	**6**
Total amount	**100**

Liquid radioactive wastes are formed during decontamination, wet cleaning of working rooms and washing of working clothes. The liquid waste specific activity does not exceed 10^{-5} Ci L^{-1}, but the volume of those wastes is about 99% of the total volume of liquid waste. Besides, the T-contaminated oil of technological vacuum pumps should be treated as liquid waste. The properties of vacuum pump oil contamination by tritium were researched experimentally. The oil represented the main part of the tritium activity of the liquid waste, but the oil volume is about 1% of the total waste volume. It was defined that the maximum level of tritium activity may reach to 700 Ci L^{-1}, when the average level is about 10-15 Ci L^{-1}. Up to 50% of the oil activity is dissolved tritium, with the balance being organically bonded. There are presented dissolved oxide form of tritium in the oil (Matveev, 1994).

As for solid waste, it can be considered that it includes all the materials from the internal parts of technological equipment, including composite materials and reusable parts of equipment. Also, significant amount of solid waste are generated in the working place. They are: individual protection means (gloves,

gas-masks, protective-suits etc.), wiper materials (gauze, cotton, wool), construction debris from repair work and different kinds of packages of tritium containing units.

It has been experimentally determined that the tritium solubility in the different materials when they interact with tritium activity of 1Ci L^{-1} (solubility in units of Ci cm^{-3}) are:

- for stainless and carbon steel from $6.6*10^{-6}$ to $2.2*10^{-4}$;
- for aluminum alloys $(1\text{-}2.5)*10^{-4}$;
- for rubbers and plastics $(1.5\text{-}30)*10^{-5}$

In this case the HTO solubility in the rubbers and plastics is of $1.7*10^{-3}$ to $5.5*10^{-1}$Ci cm^{-3}. This means the metals, when interacting with pure T, can be contaminated up to the level of thousands of Ci kg^{-1}, and rubbers when interacting of atmosphere with high HTO content, can be contaminated up to the level of hundreds of Ci kg^{-1}.

TRITIUM-CONTAINING WASTE HANDLING

It is allowed to discharge liquid radioactive waste of $3*10^{-5}$ Ci L^{-1} activity through the common sewerage, if they are diluted by the 10 times in the collector of the facility. Because of this, recovery of liquid waste after decontamination and washing of working clothes is not required. Liquid waste of 1Ci L^{-1} or more T activity should be treated for T extraction (Belovodski, Gaevoi, Grishmanovski, 1985).

Technology and equipment has been developed at VNIIEF to treat T-contaminated vacuum pump oil. The technology includes dissolved T extraction (about of 50% activity) by bubbling gas through oil and subsequent T-extracting of the bubbled gases. Then the oil is cemented with gypsum and converted into solid incombustible waste. Cementing reduces T release from the contaminated oil more than 20 times (Matveev, 1994). Solidified oil is packed in the metal containers and transported for the disposal. Solid metal waste (equipment, pipes, instruments, fitting etc.) are heated in vacuum to the temperature over 1000C for extraction of dissolved T. Released T is captured by the gas cleaning system. Other solid waste (gloves, gasket, individual protection means, wiper materials) are kept in sealed tanks equipped with T and HTO sorption elements. Then, the wastes are pressed, packed in metal containment and delivered to be disposed.

To extract T from the containers storing items and wastes, sorption elements have been designed (Figure 3) which are placed in containers. The operation efficiency of these elements is illustrated with the curves shown in Figure 4. The above sorption elements allow to reduce T release of the waste to the environment by a hundreds times. As the main hazard for public and environment is technological effluent gases, units have been developed at VNIIEF to extract T and HTO from the exhaust gases. The operation principles of T-extraction units is based on the T catalytic oxidation and generated HTO (oxidized and presented in effluent gases) sorption by the molecular sieve NaA (Belovodski et.al., 1975). Exhaust gases from technological equipment (boxes, vacuum systems, containers) fills the receivers (evacuated tanks of 1.2 m^3, equipped with ionization chamber for T-measurements). Removing of the gas mixture from the receivers is provided with the M-2 gas cleaning equipment (Figure 5). After cleaning by circulation regime, the gases are pumped out from the receivers with a vacuum pump and discharged through the stack to atmosphere. In this case at the vacuum pump exhaust, the filters for T trapping (Figure 6) are mounted. The filters include an oil wiper, oil capture element, as well as catalytic and sorption units. T removal efficiency by filters is over 99% (Belovodski et.al., 1995).

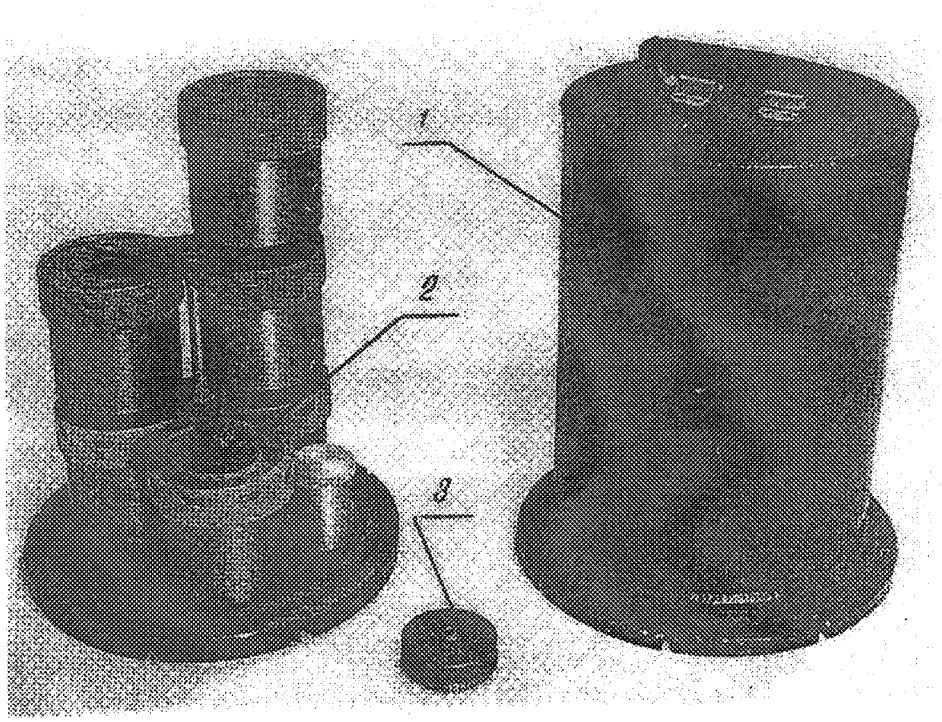

Figure 3 T sorption elements

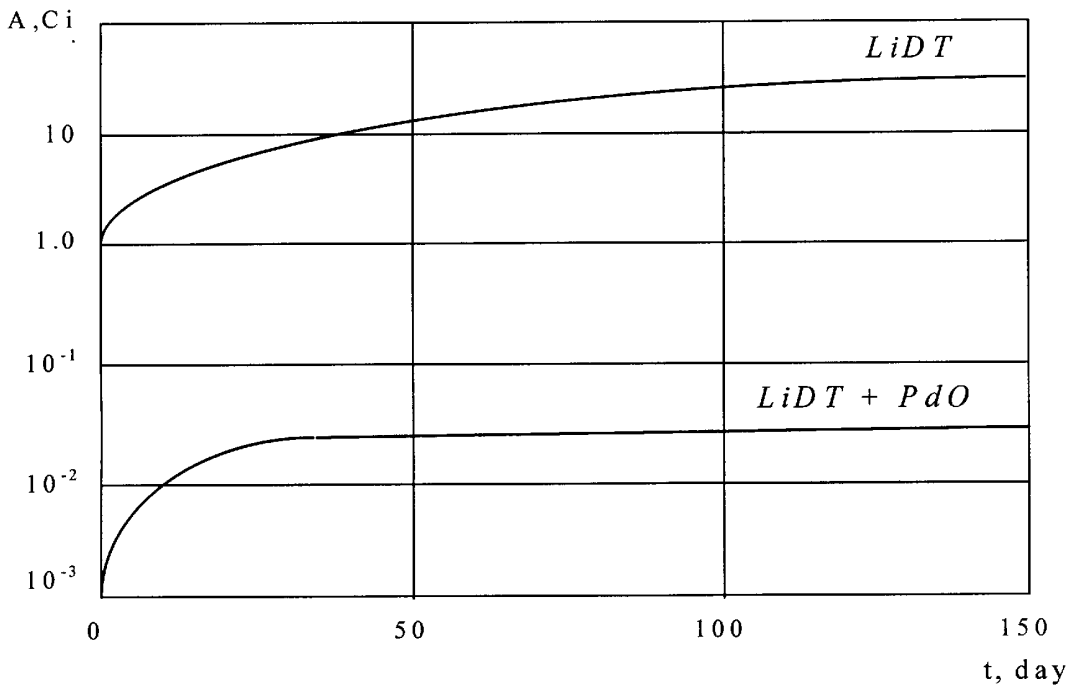

Figure 4 Operation efficiency of tritium sorption elements

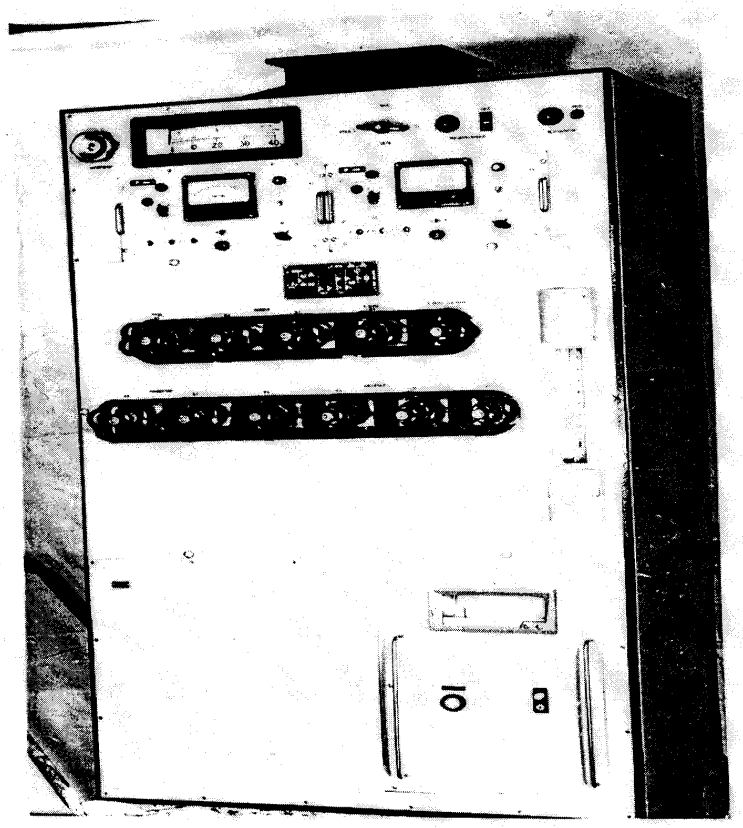

Figure 5 Tritium removal system

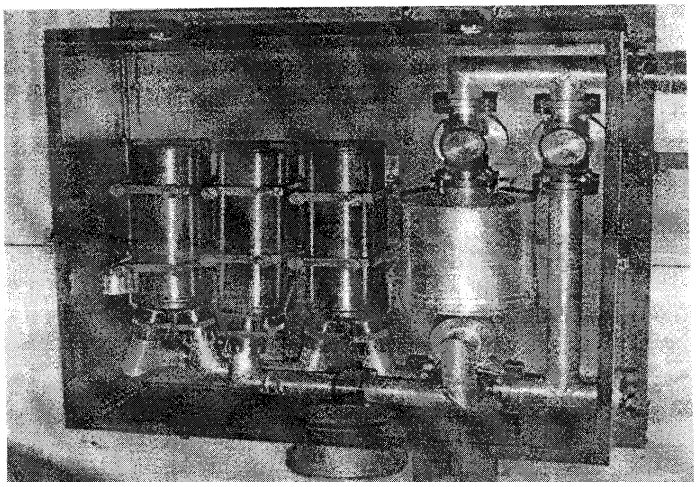

Figure 6 Vacuum pump exhaust filters

To extract T from non-oxygen gas mixtures a three stage filter was designed consisted of titanium - molybdenum, a catalytic converter and molecular sieve bed. These filters are mounted at the inlet of vacuum pumps to exclude vacuum pump oil contamination. The filter efficiency in T extraction from technological stream gases is over 99.99% (Belovodski et.al., 1995). Gases are released from the boxes and facilities in the receivers automatically, as well as the receivers are cleaned and pumped out. An automatic monitoring system - “Vybros” - accounts for T activity released to the atmosphere.

Implementation of the above T and HTO removal equipment has resulted in a reduction of T releases to the environment by more than 200 times and further 10-fold reductions are planned. An almost wasteless tritium technology has been developed at VNIIEF, as extracted T is reprocessed and recycled.

CONCLUSION

The economical efficiency of effluent gases cleaning system implementation into the industrial practice is assessed as $1,000,000 - 1,500,000 (U.S.) due to:

- reduction of technological losses of T;
- decreasing of T-impact for environment;
- reduction of collective exposure dose for population.

With the available unique tritium technologies and high skilled staff, VNIIEF is engaged in the International Thermonuclear Experimental Reactor (ITER Project) Technical Project development. Effluent gases removal systems have been developed at VNIIEF within the ITER project framework.. The system is intended to remove tritium and tritium oxide of the atmosphere of technological equipment, working rooms and facility's discharges. The workbench base for testing of operability of different units of ITER's technological equipment has been created at VNIIEF to duplicate ITER operation conditions in terms of tritium amount. Also, technical requirements for the ITER technological equipment have been developed in regard to the radiation protection and environmental safety.

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KEY WORDS

Tritium, tritium technologies, radiation protection, tritium-containing waste, equipment to handle tritium, tritium extraction from waste, environmental tritium release.