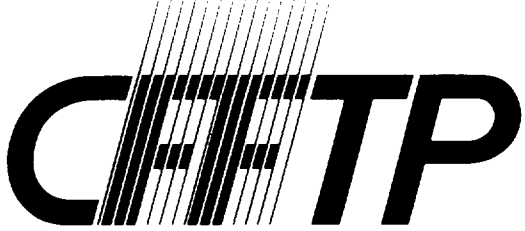
Canadian Fusion Fuels
Technology Project



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SURVEY OF TRITIATED OIL SOURCES AND HANDLING PRACTICES

CFFTP G-9310
AECL-11179
August 1994

J.M. Miller
AECL Research
Chalk River Laboratories

CFFTP GENERAL

The Canadian Fusion Fuels Technology Project represents part of Canada's overall effort in fusion energy research. The focus for CFFTP is tritium technology and remote handling. The Project is funded by the Government of Canada and Ontario Hydro Technologies. Ontario Hydro Technologies administers the Project.

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EXECUTIVE SUMMARY

Tritium interactions with oil sources (primarily associated with pumps) in tritium-handling facilities can lead to the incorporation of tritium in the oil and the production of tritiated hydrocarbons. This results in a source of radiological hazard and the need for special handling considerations during maintenance, decontamination, decommissioning and waste packaging and storage. The results of a general survey of tritiated-oil sources and their associated characteristics, handling practices, analysis techniques and waste treatment/storage methods are summarized here. Information was obtained from various tritium-handling laboratories, fusion devices, and CANDU[1] plants.

The knowledge of tritium interactions with organic materials and the increased maintenance and operational hazards of oil-containing pumps has meant that only dry pumps are recommended for use in tritium systems. Many facilities, however, have a historic waste inventory that must be dealt with, and some use is still made of oil-containing pumps. Within the broad range of tritium-handling facilities and their specific operations, there is a wide variation in the tritium-contamination levels of these oil-sources, as well as in other contaminants and the type of oil used. General handling practices, however, are quite standard throughout all the facilities.

Some detailed analysis data of the type and quantity of the various tritium species (HT, HTO or tritiated hydrocarbons) present in tritium-contaminated oil are available. This information is necessary for the development of any effective pre-treatment steps prior to waste storage. Generally, tritium-handling laboratories have handled the waste by absorbing the oil on a sorbent material and packaging it in a multiple-walled container for storage. The waste package must be shown to meet the acceptance criteria of the waste-handling facility. For tritium-contaminated oil, the pressure buildup in the waste package due to radiolysis and the leach rate of the various tritiated species from the package during the storage period are important considerations. Experimental test data are available for both of these considerations, demonstrating the acceptability of this general approach to waste storage. Incineration is another treatment option and is currently used by the JET facility and others for the treatment of suitable low-level oil sources.

To achieve complete tritium recycle within a facility, waste processing cycles must include recovery from any tritiated oil that may be produced. Data from many facilities indicate that HTO is a major component in the oil; techniques to remove the HTO component, such as vacuum degassing, can be carried out. Tritiated hydrocarbons are also present; the activity level dependent on the oil source (type, age and quantity of tritium) and techniques to remove this component of the tritium activity have also been investigated. Treatment of any secondary waste stream produced during processing must also be considered.

Opportunities exist to develop optimized handling and waste management strategies for tritiated waste oil in the facilities that continue to handle it, and in facilities that need to deal with a historic waste inventory.

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[1]CANDU: CANada Deuterium Uranium; registered trademark.

1. INTRODUCTION

Water and hydrogen are the main tritium-bearing compounds in tritium-handling facilities. However, many other tritium-bearing compounds may be formed during the operation of tritium-handling equipment, or within a facility, through tritium interactions with the various materials present. Metal tritides and organic forms of tritium are often present, formed through the intended process operation or by the interaction of the tritium with the system material(s). Tritium behaves physically and chemically very similarly to hydrogen, but the presence of the ionizing ß-particle can radiolytically catalyze many chemical reactions. Reviews by the International Atomic Energy Agency (IAEA) [1991] and Gill and Coffin [1982] provide a good summary of the interactions and compatibility of tritium with various materials. In addition to tritiated-compound production itself, tritium-materials interactions are also important considerations for containment integrity and contamination control within the tritium-handling facility. In particular, tritium interactions with oil sources (primarily associated with pumps) can lead to the production of tritiated hydrocarbons, resulting in additional radiological hazards and special consideration for maintenance, decontamination, decommissioning and waste handling.

Although the generally accepted design requirements for tritium facilities and tritium-handling equipment counsel against the use of oil-containing pumping systems, in some cases it is unavoidable. A general literature survey was undertaken to assess the following items associated with tritiated oil sources in tritium facilities, including tritium research laboratories, fusion facilities, CANDU stations and tritium removal facilities:

- type and quantities of tritiated oil,
- source of oil,
- tritium concentrations,
- assay of tritiated components,
- facility handling procedures,
- waste management practices, and
- personnel dosimetry.

In addition, many facilities were contacted for details of their operating and handling experience with tritiated oil. With the development of dry pumps to meet a wider range of pumping requirements, many new tritium-handling facilities do not or will not include oil-containing pumps in their facilities. However, tritiated oil is still a current waste source for some facilities. Acceptable handling and waste management strategies for tritiated-oil waste are also being determined and/or put into place for decontamination and decommissioning of some facilities, and for the storage/disposal of historic waste-oil inventories. This report identifies the sources of tritiated oil being produced in specific facilities, reviews some of the development work that has been carried out on characterization, and summarizes the past and present practices of handling tritiated oil.

2. PRODUCTION/SOURCES OF TRITIATED OILS

2.1 Fusion Facilities

The Joint European Torus (JET, Culham, England) and the Tokamak Fusion Test Reactor (TFTR, Princeton, USA) are two fusion devices that include oil-containing pumps and have operated with tritium. Information available to date from these facilities, through both published reports and private communication, is summarized below.

2.1.1 JET

Tritium-contaminated oil waste was produced at JET during the clean-up and maintenance phase [Winkel et al., 1993] following the first series of plasma discharges fuelled with a mixture of deuterium and tritium in 1991 November [JET Team, 1992]. The normal backing-line pumps, a series of roots blower/rotary pump combinations, were isolated for this Preliminary Tritium Experiment (PTE) and replaced by a gas collection system that included a liquid helium-cooled cold finger cryopump. These normal roots/rotary pumps were reconnected for final system cleanup after completion of the experiment and when overall stack releases had been reduced to a minimum level. Thus, this final part of the clean-up and maintenance phase required handling of tritiated-oil sources in the exhaust-pumping system.

A slight contamination level was measured in the rotary (roughing) pumps prior to the PTE, as a result of the small amount of tritium produced in the D-D reaction (see TFTR experience, Section 2.1.2). Levels were in the order of 10 kBq/L oil. Following their use in the clean-up phase after the PTE, tritium concentrations reached in the order of 10-500 kBq/L, depending on the location of the pump, except for one pump in which the concentration was measured at 1.09 MBq/L. JET personnel estimate that this pump had approximately 250 GBq of tritium pass through it as HTO over a three-month period, giving a pick-up rate of <0.01%. The roughing pumps have 20 L of oil each.

Similarly, a significant increase was noted in the tritium-contamination level of the turbomolecular pumps after their use in the clean-up phase. The measured contamination levels showed a clear relationship to the pump operation. Oil in the pumps of the neutral-beam injector systems ranged from 20-120 MBq/L, while only up to the 2.5 MBq/L was found in those pumps mainly pumping the torus. The highest level, 120 MBq/L, was measured in the pump that was used to pump ~40 TBq as HT over 12 h and about 500 GBq as HTO or methane over three months. The presence of tritium-contaminated oil was also reflected in the surface-contamination measurements made in the recovery system. The surfaces that consistently showed elevated levels of tritium contamination were those contaminated with oils.

In the upcoming D-T experiments planned for JET, the Active Gas Handling System (AGHS) [Haange et al., 1988] will be used for backing the torus, therefore replacing the roughing pumps. However, contaminated oil in the turbomolecular pumps will still be an issue. JET have extrapolated the results based on the known throughput and estimate that with the foreseen throughput of 1 kg of tritium, a contamination level of 10 TBq/L will be produced. Anecdotal evidence from the Chalk River Laboratories (CRL) Tritium Laboratory operations indicates that the pick-up is not a linear relationship; the predicted value is also twice that of the maximum level reported in the IAEA "Safe Handling of Tritium" report [1991], which reviews tritium handling experience. However, while the projected maximum concentration may be high, the contamination level will almost certainly be of a much higher concentration (10^4 -10^5 times) than JET have had to deal with to date.

JET are planning to develop a strategy for handling the tritiated oil produced in their next phase(s) of tritium operation. Until now, the contaminated oil (~500 L, ~<150 Bq/g) has been sent to Harwell for incineration. Provided they can maintain the oil beryllium-free, Harwell will continue to accept it. Currently, there is not an upper limit in tritium concentration that Harwell can accept. With the expected much higher tritium concentrations following the planned D-T operating phase, it is an important issue for any maintenance work required during this phase and during the subsequent decommissioning phase.

If JET chose to do some pre-treatment of the oil to reduce the contamination level and/or recover the tritium prior to waste processing, there is the possibility of linking a pre-treatment procedure with the AGHS. Although no detailed analysis has been done on the tritium species in the oil, JET have noted an initial burst of HTO into the surrounding atmosphere followed by HT while changing the oil. The observation that there is a substantial fraction of the tritium in the oil as HTO is consistent with observations in other laboratories (see Section 3). The AGHS is already in place to recover tritium from other tritiated water sources, so potentially it could be used to process this waste stream as well. The presence of any other contaminants (e.g., Be) would also have to be considered in the development of any treatment option.

2.1.2 TFTR

Exhaust-gas pumping from the TFTR machine is provided by oil sealed/lubricated pumps. Tritium contamination was measured in the oil during D-D operation; however, no data have yet been obtained on contamination levels reached during D-T operation. Detailed handling and packaging procedures have been developed and incorporated into the maintenance activities during this operating period.

A brief description and the results of an investigation of the general contamination of the vacuum pumping line in the TFTR prior to the D-T operating phase is given by LaMarche et al. [1993]. In the TFTR, the Torus Vacuum Pumping System consists of eight turbomolecular pumps, four at the end of each of two large pumping ducts, that are fore-pumped by two mechanical pumping packages: a roots blower and a rotary piston pump in each package. Four neutral-beam enclosures (each 50 m^3) are attached to the vacuum vessel. These enclosures contain boiling liquid helium cryopanels shielded by liquid N_2 panels. Ninety percent of the plasma-exhaust gas is pumped by the neutral-beam enclosures. Each enclosure is pumped by a turbomolecular pump, and is regenerated periodically by a roots/rotary piston pump package. The turbomolecular pump is fore-pumped by a mechanical-pumping package. Other vacuum appendages have their own dedicated pumping systems.

During D-D operations, approximately 74 GBq of tritium were produced. This amount was measurable in the vacuum exhaust lines and in the oil of the vacuum pumps. As well as analysing oil samples from the vacuum pumps, samples were taken from areas wetted with oil in the foreline. Tritium contamination was strongly correlated with the presence of oil (similar to the JET experience), which has implications for maintenance and decontamination procedures. Tritium contamination was measured to be at a level of kBq-MBq/L, with the highest value (2.9 kBq/g (~2 MBq/L)) being found in the pumps used to regenerate the cryopanel in the neutral-beam enclosures. This would be expected, as the majority of the plasma exhaust gas is pumped by them.

D-T operations are expected to produce tritium concentrations in pump oil of up to 1 TBq/L [Stencel, 1994]. This extrapolation is based on the reported concentrations from other tritium-handling facilities. Approval has been given to package the oil as a radioactive waste and ship it for disposal to the US Department of Energy's Hanford site. The oil is stabilized using Petroset II^{TM} (granular) solidification media in an approved container prior to shipping. An important issue associated with disposal in containers is the generation of pressure within the disposal package due to radiolysis. The package proposed to be used by TFTR is the Enduropak^{TM} (US Department of Transportation specification Type A container), which has been designed to contain hydrogen that may be generated from radiolysis without the use of a catalyst. To help avoid contact with tritiated oil during pump

maintenance, TFTR staff have engineered and fabricated an oil change-out-cart that allows the direct transfer of oil from the pump into a closed-top, 30-gallon waste container.

2.1.3 Future Fusion Devices

Future devices, such as ITER (International Tokamak Engineering Reactor) and NET (Next European Torus), intend to use oil-free pumping systems, reducing or eliminating this source of waste.

2.2 Tritium-Handling Laboratories

Published reports from tritium-handling laboratories make reference to using or not using oil-containing pumps, but very few details are provided in the literature on any tritiated-oil sources or their treatment if they are produced. The IAEA "Safe Handling of Tritium" report [1991] summarizes the standard practice of handling and disposing of tritiated oil produced in tritium-handling laboratories up to that report's time of preparation (1986-1989). Most of the new tritium-handling laboratories put into place in the 1980's completely banned or tried to limit the use of tritium-containing pumps, and many operating laboratories have changed, or are in the process of changing, to "dry" (lubricant-free) pumps. In the past, for some applications the selection of an oil-containing pump was unavoidable, as dry pumps with the appropriate pumping characteristics were not available or were prohibitively expensive. As a general rule, the recent laboratories put in operation to support fusion programs (e.g., the Tritium Systems Test Assembly (TSTA), Los Alamos, USA [Anderson et al., 1985]; the JAERI Tritium Processing Laboratory (TPL), Tokai-Mura, Japan [Naruse, 1986]; the Tritium Laboratory Karlsruhe (TLK), Karlsruhe Germany [Ache et al., 1990]; and the European Tritium Handling Experimental Laboratory (ETHEL), Ispra, Italy [Vassallo et al., 1988]) do not use oil-containing pumps in their process systems, and have specifically selected lubricant-free pumps or compressors for auxiliary systems, as well. Many of the older operating laboratories are converting to dry pumps where practical, so that the volume and handling of tritiated oil from these laboratories is being limited. No detailed information on the amount of tritiated oil in historic waste inventory has been obtained. In addition to oil- or lubricant-containing pumps, mercury pumps have been used in the past in some tritium facilities. This also has implications on the development of decommissioning and waste-handling procedures for such facilities.

The IAEA "Safe Handling of Tritium" report lists vacuum pump oil as a tritium waste with a typical tritium concentration of 5 Bq/L to <50 GBq/L. Current information obtained from laboratories indicates that this range is still valid, with only limited quantities of tritiated oil at the higher concentration being produced as a waste source. The concentration produced depends on the throughput of tritium and the type of oil used, with anecdotal information from operation in the CRL Tritium Laboratory indicating that a plateau level of the tritium-contamination level is reached. As for the interactions of tritium with organic materials in general, the uptake of tritium by the oil can lead to disruption of their bulk chemistry. This then can impact on pump performance, dictating special servicing and maintenance requirements. Saturated hydrocarbon mineral oils require frequent changes in T_2 service, because of vapour pressure increases (off-gassing) and liquid viscosity increases. Silicone oils are rapidly polymerized or solidified. Polyphenyl ether oils are quite stable in tritium service, although they can also absorb significant amounts of tritium.

The IAEA review of tritium-handling experience also discusses packaging and conditioning of tritiated-oil waste for storage. Generally, it is suggested that the oil be absorbed on a sorbent material in a tight container with single or double containment, depending on the specific activity and the volume. Out-gassing caused by radiolysis must be taken into account in any

conditioning and packaging of the waste oil of a high specific activity (~0.4 TBq/L). Local regulations will usually stipulate the quantities and packaging requirements for storage and transportation.

At the 1993 EuroCourse on Safety in Tritium Handling Technology, Giroux [1993] indicated that oil-containing pumps were no longer being used at the CEA Tritium Laboratory at Valduc, France. However, they do have a historic inventory of tritiated oils which must be dealt with as part of their decommissioning program at the facility. No details have been obtained on the treatment procedure, but an oxidation procedure was being put in place to deal with the inventory. Figure 1 shows a schematic of the tritiated waste processing system proposed by Giroux.

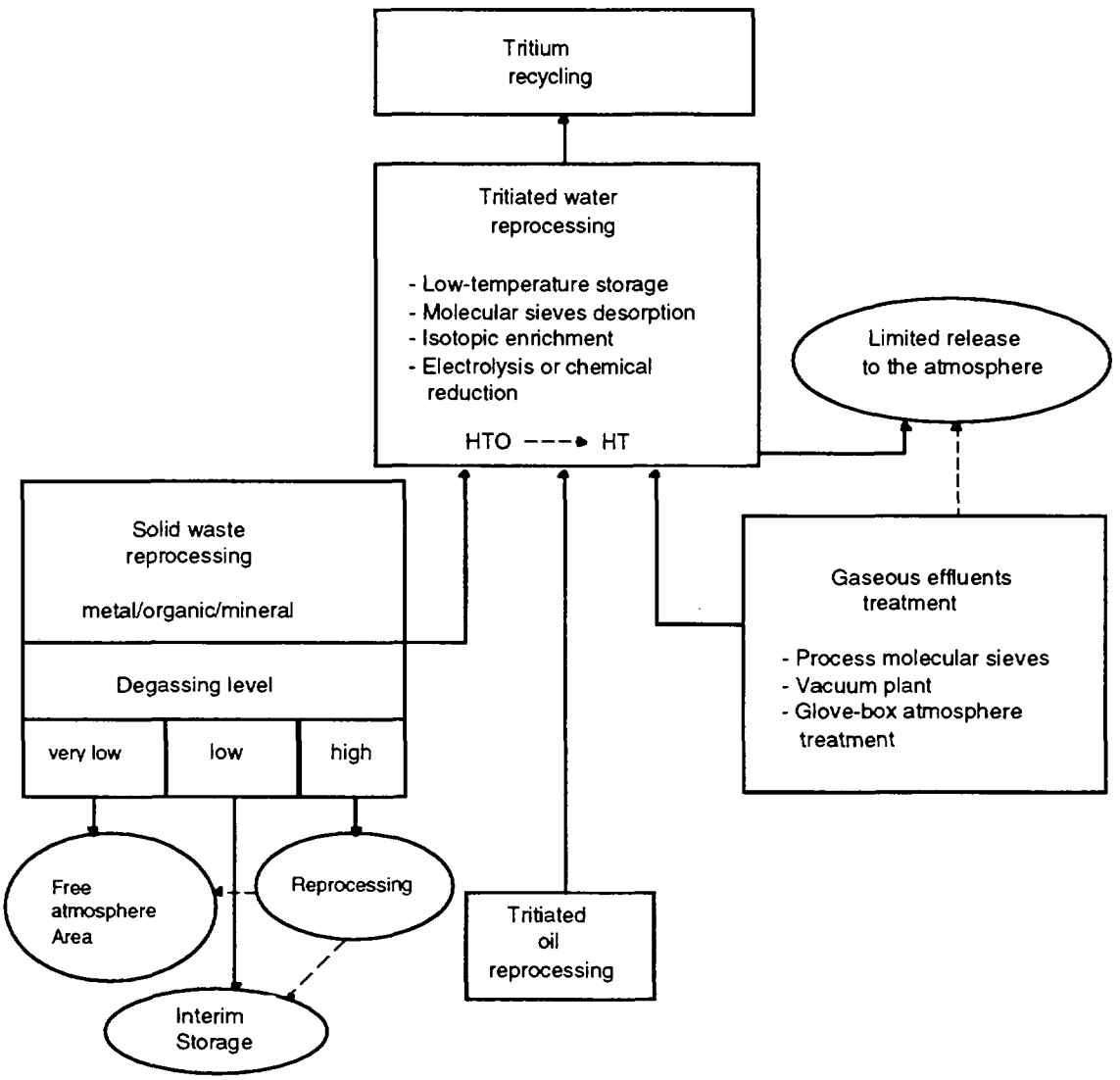

Figure 1: Reprocessing Cycle of Tritiated Wastes at CEA - VALDUC [Giroux, 1993]

Although no specific reference is made to the production of tritiated oil, in defining the waste-management strategy for the ETHEL, Mannone [1992] proposes that any tritiated organic liquid wastes be solidified by sorption on vermiculite or an equivalent. A capacity of 2 GBq/kg is the maximum tritium/solid ratio recommended by the Italian Regulatory Authority. The resulting solid composite would be processed in the facility as a solid tritiated waste. Only dry pumps have been installed in the ETHEL process systems, so any wastes of this type should be minimal.

Both the Tritium Systems Test Assembly (TSTA) and the Tritium Salt Facility (TSF) at Los Alamos use dry, hermetic, and packless tritium-handling pumps [Anderson et al., 1985]. Problems were encountered initially with tritium contamination of their vacuum pass boxes by oil vapour backstreaming from the house-vacuum system. They modified the house-vacuum system by separating it into two systems. A system served by an oil-free scroll pump is used exclusively for evacuation of tritium-contaminated components. However, tritiated oil was produced as a waste source in the past at Los Alamos and concerns have been raised recently [Bartlit, 1993] that radiolysis may produce significant gas pressure, causing the waste drums to rupture and tritium to escape. The Los Alamos Waste Group has been analyzing this problem to determine whether the waste packages are satisfactory. The standard method to prepare the waste package was to absorb the oil on vermiculite (or corn-cob fractions) and doubly contain this waste form in drums.

The CRL Tritium Laboratory continues to use oil-containing pumps in its main Tritium Handling Apparatus. A rotary vane pump with magnetic drives (Nova Magnetics, Halifax, Nova Scotia), lubricated with tritium-compatible, low-vapour pressure polyphenyl ether oil (~20 mL), is used to transfer tritium in the Tritium Handling Apparatus. An oil-containing diffusion pump and a mechanical rotary pump are used as the vacuum-pump set for this apparatus. Operating procedures minimize the amount of tritium pumped through this vacuum-pump set, with the measured tritium concentration in the oil being typically less than 20 GBq/L, depending on the previous operations (tritium throughput and time since oil change). Tritium concentrations in the rotary vane pump have ranged up to 2.2 TBq/L. Special procedures have been developed to minimize contact with the oil during change-out. The oil is aspirated directly into an evacuated storage container.

The operation of heavy-water research reactors at CRL also results in low-level tritium-contaminated oil waste. Approximately 400 L of vacuum pump oil (10W30 and DuoSeal) containing tritium is put into storage each year. The tritium concentration is in the range of MBq-GBq/L. The oil is currently being stored in drums for future disposal. Methods of solidifying this and other contaminated oil sources are being investigated in an R&D program for long-term waste storage or disposal.

The Ontario Hydro Technologies Tritium Laboratory is in the process of replacing the oil-containing mechanical rotary/turbomolecular pumping systems on tritium-handling equipment with dry pump sets (diaphragm and molecular drag pumps). The tritiated oil produced typically has a tritium concentration in the order of 0.1 GBq/L. It is immobilized and sent to the Ontario Hydro Bruce site for waste storage. A long-term tritium compatibility test of a turbomolecular pump with T_2 was carried out. In this test program, oil concentrations in the order of 6 GBq/L were produced and handled.

Some tritium light manufacturers also use an oil-containing rotary pump, to evacuate their tritium-labelling process system. The standard practice of absorbing the oil on a sorbent material and packaging it for waste storage is followed.

2.3 CANDU Plants

Most of the documented information on the contamination characteristics and waste-handling practices for tritium-contaminated oil in CANDU plants is available from Ontario Hydro. Many different types and quantities of oil are used in the different plants, and various contaminants (both radioactive and chemical) are introduced; different utility and station requirements dictate the handling, packaging and storage of this waste source. A significant amount of work on characterization and storage options has been done by Ontario Hydro. This work is summarized here, to illustrate some of the oil characteristics and handling options to be considered for tritium-contaminated oil produced in CANDU reactors. Much of the information that follows has been obtained from various reports by Krasznai et al. [1989-1993], with some additional information being obtained directly from staff at the Darlington Tritium Removal Facility (DTRF).

Ontario Hydro generates 5500 L of radioactive waste oil per month by operating and maintaining its nuclear generating stations. In addition, there is a significant historical source of waste oil (~180 000 L) in storage awaiting selection of a disposal option. A variety of decontamination methods have been investigated for removing the contaminants for a number of treatment options or scenarios. The oils in inventory have been well characterized and contain a number of active contaminants (primarily tritium, Cs^{137} and Co^{60}), inactive contaminants (such as lead and cadmium) and volatile aliphatic and aromatic solvents, that all affect the selection of a disposal option. A source of historic waste oil stored at the Pickering Nuclear Generating Station (PNGS) has the following characteristics:

Radioactive (kBq/L)	Inactive Contaminants (mg/kg)
Beta/Gamma: avg 92, min-max 0.74-629	Solvent: few %
Tritium: avg 30 710, min-max 259-368 150	Lead: avg 24, min-max <2-220
Carbon-14: <148	Cadmium: avg 1.6, min-max <0.2-4.6

Lower levels of contamination were measured in oil samples from the Hydro-Quebec Gentilly 2 station, indicating that the above contamination levels could be considered an upper bounding case for waste oil generated in CANDU reactors.

Any selected decontamination method must consider all of the contaminants. Laboratory tests by Krasznai [1994] on some of the historic inventory have shown that the tritium is associated primarily with suspended and dissolved water in the oil, and to a lesser degree with polar additives in the oil. Efficient tritium decontamination can therefore be achieved by the combination of a water-removal step and an oil-additive removal step. Vacuum degassing was demonstrated to be an effective tritium removal process, but it has limitations. If the tritium contamination results from a long exposure, vacuum degassing may not result in achievement of the required decontamination level, because some of the tritium may be incorporated into the oil additives. If there are water-soluble radionuclides, such as Cs^{134} or Cs^{137}, vacuum degassing concentrates these radionuclides in the vacuum chamber, raising concern over another contamination source. Another drawback is that vacuum degassing generates another secondary liquid waste stream that also must be managed as a mixed waste (low-volatile solvent plus water). As an alternative, testing using an acid-wash technique was successful in reducing the tritium level, but this also produces a secondary liquid waste. Some development work was carried out to generate a solid rather than a liquid secondary waste stream. Development and testing of a dry process using filtration was successful in reducing the tritium levels in waste oil samples to less than the defined study limit of 74 kBq/kg. The choice of one decontamination technique over another

is very dependent on the disposal options and the limitations of those options in consideration of both radioactive and inactive contaminants.

2.4 Ontario Hydro — Darlington Tritium Removal Facility

Vacuum pumps are a source of tritiated oil waste in the DTRF [Krasznai, Chew and Hudson, 1992; Smith, 1993]. Total quantity in the pumps is 42 L. It is changed infrequently, on no more than a semi-annual basis. Tritium concentrations are in the range of 1 TBq/L. Satisfactory operating procedures have been developed to minimize the tritium hazard during service work on the pumps. Analysis of the type and concentration of tritium species present in the surrounding atmosphere during the execution of this change-out procedure is discussed further in Section 3. Waste-package design and leakage criteria were developed and tested [Krasznai, 1989] prior to operation of the facility. Various types of absorbents and the demonstrated effectiveness of selected package barriers are given. The current procedure for handling this tritiated waste source is to drain the oil into a polyethylene container charged with an absorbent. The container is sealed and placed in another polyethylene overpack for transportation to and storage in Ontario Hydro's waste storage facilities.

3. ANALYSIS

Two types of analyses of tritium-contaminated oil are important for its safe handling within a facility, and for the development of appropriate waste storage or waste handling methods. One type of analysis provides the total tritium activity in the oil, while a more detailed compositional analysis provides information on the type of tritiated species present and the relative amount of each (e.g., HT, HTO, tritiated organics). This latter measurement is also important, to ensure that appropriate radiation protection practices are followed during handling of the oil in the facility, as the committed dose due to tritium is very dependent on the tritiated species.

The total tritium concentration (or specific activity) is usually determined by liquid scintillation counting. Care must be taken during this technique, to ensure that the colour and luminescent nature of the oil samples do not interfere with the counting procedure. Sood and Kos [1990] have developed a procedure that demonstrates that the reliable determination of tritium in oil samples can be carried out using liquid scintillation counting. This procedure requires that an acceptable luminescence and counting efficiency be obtained for the sample, or the sample be combusted in a Tricarb sample oxidizer. A minimum detectable activity of ~0.4 kBq/kg was obtained, but higher levels would be expected if very dark or dirty oil samples were analyzed.

While liquid scintillation counting does confirm the total tritium contamination level, it does not give any information on the tritiated species present in the oil sample, or identify what might be outgassing from it. Tritiated hydrocarbons are produced as a radiolysis by-product of tritium-contaminated oil. The amount of tritiated hydrocarbons produced is related to the tritium concentration, exposure time and the interaction of tritium with the specific oil formulation. Reliable dose assessments cannot be made for tritiated hydrocarbons at this time; therefore, exposure to these tritiated species must be carefully controlled. If some decontamination is to be carried out before waste storage, the relative concentration of the tritiated species present must be known, so that the required level of decontamination can be achieved. From the oil being handled and characterized by the various facilities, the general observation is that the predominant form of tritium in the oil is HTO. Data from Krasznai et al. [1990] showed that >99.9% of the volatile fraction released from a tritiated oil sample was exchangeable with H_2O, with a significant fraction of this suspected to be HTO. They also concluded that non-volatile, polar species can represent a significant fraction of the

tritium-contamination level in aged oils. JET have observed during change-out of their pump oils that HTO is released initially into the surrounding atmosphere, followed by HT [Bell, 1994]. TFTR also found a high concentration of tritium in the water phase of a water:oil mixture of 660:1 [Stencel et al., 1988]. In the CRL Tritium Laboratory, ballasting of the mechanical rotary pump used on the Tritium Handling Apparatus with air reduces the tritium concentration in the oil significantly, implying that a large fraction of the tritium is present as tritiated water.

The formation and presence of tritiated hydrocarbons in oil increases with tritium concentration and exposure time. Methods of determining the tritiated hydrocarbon species present have been developed at both AECL and Ontario Hydro, primarily to determine what tritium species are released in out-gassing from oil sources that had been exposed to elemental tritium. Radio-gas chromatography techniques were developed using a flow-proportional-counter detector (FPCD) in series with a thermal-conductivity detector [Krasznai, Massey and Agg, 1992] or duplicate analysis of a sample in a gas chromatograph fitted with a FPCD and a flame ionization detector [Rodrigo and MacDonald, 1993], to identify both the organic species in the sample and those that were tritiated. Although different areas were sampled, a comparison of the two sets of results illustrates the variability in relative concentrations of the various tritiated species that may be expected from different oil sources.

In Krasznai et al.'s analysis [Krasznai, Massey and Agg, 1992] of the work area atmosphere above a 0.39 TBq/L oil source, the dominant volatile tritiated species detected above the oil was HT, with smaller quantities of C_2 and C_3 hydrocarbons being detected (5 to 100 times less, depending on the air sample location). The technique did not allow detection of tritiated polar species (tritiated water or tritiated formaldehyde), because they do not elute from the gas chromatograph column used. However, a comparison of total tritium concentrations measured with a portable ionization chamber-based monitor with the total tritium concentration measured using the GC-FPCD system agreed very well, indicating that most of the tritium in the off-gas was HT. This is somewhat surprising, as a measurable amount of HTO would be expected; however, it may have been released in the off-gas in the initial time of the transfer. From a waste-handling viewpoint, the measurements demonstrated that the concentration of tritium gas and tritiated hydrocarbons released from the oil were significant, and due consideration needs to be given to work procedures for any handling or treatment of the source.

Similarly, the analysis carried out at CRL [Rodrigo and MacDonald, 1993] identified a variety of C_2, C_3 and C_4 hydrocarbons in the head-space gas of a sealed container containing tritiated oil with an ~37 GBq/L tritium content. The total concentration of elemental tritium plus tritiated hydrocarbons was found to be ~15.9 GBq/m^3 in the head-space gas, with the dominant species being $C_2H_4(T)$ ~60%. No tritiated water was detected, but, although polar compounds are not strongly retained by the column used, the HTO could have been retained on the dry system walls and not eluted through to the detector. The large relative amount of $C_2H_4(T)$ contrasts with the results of Krasznai et al., but the relative content of HT and tritiated hydrocarbons may differ, because of the age of the oil (>3 years in this case, cf. ~0.5 years) and the radiation characteristics of the oil.

4. WASTE TREATMENT/STORAGE

The review of tritium-contaminated oil sources in Section 2 identified some of the handling and waste-treatment methods practiced by the various facilities. Standard practice in the past has been to absorb the waste oils on an absorbent (resulting in a solid, rather than

liquid, waste form) and packaging it in a multiple-walled container [IAEA, 1981]. This practice is still continued by some facilities (e.g., TFTR, OH). Approved transportation and storage packages are an integral part of this method, and must comply with the waste management strategy for the operating facility. The long-term integrity of the package is a concern that has been raised recently. At some facilities (e.g., JET, China Institute of Atomic Energy), if the tritium concentration is within acceptable limits, the tritiated-oil waste is incinerated [Nanchang et al., 1993; Bell, 1994].

If a decontamination method is to be put in place prior to storage or treatment, it is important that the production of any secondary waste streams be considered, as well as the increased handling required. For example, to implement techniques like vacuum degassing or oxidation, the secondary waste streams formed must be considered. Figure 1 illustrates the decontamination method proposed by the CEA-Valduc tritium laboratory to allow the recovery and recycle of tritium from all of their waste sources within their facilities. As can be seen, tritiated oil reprocessing is only one part of the tritiated waste reprocessing cycle. Reprocessing the tritiated oil produces a secondary waste stream, which is integrated with another, now similar, waste stream for further treatment.

In some cases, the oil must be considered as a mixed waste (other contaminants are present; e.g., radioactive, heavy metal). All of the contaminants for which restrictions apply must be considered in the selection of an acceptable waste treatment or storage method.

5. CONCLUSIONS AND RECOMMENDATIONS

Following the recommended design practice of eliminating the use of organic materials in tritium-handling system components and processes, oil-containing pumps are being limited in their use in tritium-handling facilities. However, some laboratories and the two fusion devices using tritium (TFTR and JET) continue to handle tritium-contaminated oil and produce this waste stream. Significant quantities of low-level tritium-contaminated waste oil also continue to be produced in CANDU plants. In some facilities, management of historic inventories of tritium-contaminated oil is an integral part of the facilities' waste management strategy. Operational activities in the various tritium-handling facilities have resulted in a wide variation in the quantities of tritiated oil produced within these facilities, the degree of tritium contamination of the oil, and also in the presence of other contaminants within the oil. While the same basic philosophies are followed in many of the facilities, facility regulations and local authorities permit some variation in the handling procedures and waste treatment options that are practiced.

In review of some of the practices followed within the various facilities producing tritiated oil, the following guidelines were developed to summarize a suggested approach to dealing with this waste:

- identify the contaminants (radioactive and non-radioactive), including the tritium species contributing to the tritium-contamination level;
- determine the options for waste storage or treatment to meet institutional and regulatory requirements;
- if pre-treatment is necessary, determine options; demonstrate decontamination levels and define secondary waste streams;
- select final option and develop overall handling, treatment and/or storage procedures, and put in place the necessary quality assurance program to demonstrate knowledge of

the contamination (radioactive and non-radioactive) level and the characteristics of this contamination and the waste itself.

Complete tritium recycle is the ideal within a tritium-handling facility. From the reported development work carried out to date, various approaches to developing pre-treatment or tritium recycle options can be suggested. If a facility can further process a stream containing tritiated water, removal of a large fraction of the tritium (as HTO) from the waste oil inventory in a pre-treatment step may be simple and cost-effective (e.g., vacuum degassing). Removal of the HTO and other polar compounds onto an absorbent material may also be acceptable, depending on the waste oil characteristics and the total tritium activity present (a solid waste would then have to be dealt with). Oxidation methods to reduce the tritiated hydrocarbon concentration would further decrease the waste inventory. Even if complete recycle is not possible, there may be some benefit in pre-treatment to minimize the packaging requirements for transportation and/or storage, and/or to make more options available to the facility operator. The oil must be well characterized before any processing is carried out. If it is a mixed waste, additional treatment steps to reduce the other contaminants may also be required. The current state of knowledge, based on handling practices and the development/characterization work carried out to date, is sufficient to develop optimized handling and waste management practices for tritiated oil within individual facilities.

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