

EXPERIMENTAL TRIALS OF METHODS FOR METAL DETRITIATION FOR JET

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Several methods of decontamination, such as melting, heating with flame, isotopic exchange with gaseous hydrogen, replacement with hydrogen, and thermal desorption under moist gas, were tested on stainless steel, INCONEL[®], beryllium, copper, and aluminum bronze contaminated with tritium. The detritiation methods were assessed with respect to the fraction of the tritium inventory removed, the residual tritium concentration remaining, and the reduction in the rate of tritium outgassing. Potential applications of these decontamination methods include detritiation of the Joint European Torus (JET) vacuum vessel and the tritium plant prior to dismantling for decommissioning and subsequent processing of the intermediate-level waste this has generated.

KEYWORDS: *tritium in metals, metal detritiation, fusion machine*

I. INTRODUCTION

The Joint European Torus (JET) is a large experimental tokamak designed to operate using deuterium and tritium fuel. An intensive campaign of operation with deuterium/tritium plasmas, Deuterium-Tritium Experiment 1 (DTE1) (Ref. 1), was carried out in 1997. In preparing for DTE1 it was recognized that operation with tritium will produce solid radioactive contaminated materials that may require the future building of additional facilities for tritium recovery or detritiation.

While the materials could be disposed of as waste without further processing, the most cost-effective and

best practical environmental option may prove to be processing the materials to recover tritium prior to disposal. This may also permit materials that would otherwise be classified for disposal as intermediate-level waste (ILW) to be reclassified as low-level radioactive waste (LLW) following treatment.² Thus, detritiation of ILW might be a preferable option for dealing with the waste from future JET decommissioning.³ A detritiation procedure would need to be employed at JET to decontaminate the vacuum vessel and the tritium plant [active gas handling system (AGHS)] prior to their disassembly. Dismantling will generate large streams of materials contaminated with tritium at levels beyond the ILW/LLW threshold, and engineering solutions would be required to deal with tritium outgassing from the waste if it is not subjected to any treatment. It is therefore preferred that the materials undergo a treatment to reduce tritium outgassing and, where possible, reclassify the waste into the LLW category, thus reducing the cost of waste disposal substantially.

The largest stream of ILW for the JET decommissioning will be metallic materials. These include INCONEL[®] grades 600 and 625, used in the construction of the JET vacuum vessel walls, and stainless steel, Type 316L, from which the AGHS is constructed. Beryllium, copper, and aluminum bronze are also present in components installed within the torus.

The choice of which detritiation method to apply depends on many factors, among which are the following: the objective of the decontamination; whether decontamination in situ or ex situ is required; the nature, size, and shape of the material; the chemical form in which the tritium exists in the material; and whether the tritium is largely confined to the surface or has permeated into the bulk.

A short review of detritiation studies reported in the literature for metals is given below. Broadly, the following techniques, in various combinations, have been used in the past for solid material detritiation⁴: superficial

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cleaning and washing; vacuuming; purging; thermal desorption; isotopic exchange; chemical or electrochemical etching; plasma discharge methods; and destructive techniques, for example, melting and chemical dissolution. In practice, a combination of washing, purging, thermal desorption, and isotopic exchange with water vapor and/or hydrogen was the method of detritiation most commonly used. While this generally does not provide complete decontamination, it is often all that is necessary to permit equipment maintenance to be carried out safely. Chemical methods tend to produce "mixed" wastes, i.e., both chemically and radiologically hazardous wastes.

Highly tritiated solid components that have to be disposed of as waste may be detritiated and volume-reduced by destructive techniques.⁵ Thermal desorption is usually first to be carried out. Then, the material is melted or destroyed to recover the remaining tritium and to provide a volume-reduced waste form in which the residual tritium corresponds to the limit of solubility at the decontamination parameters. As an example, several tonnes of stainless steel were detritiated initially by thermal desorption at a temperature of 1070 K, which reduced the mean tritium activity from 19 to 0.15 GBq/kg. Melting at 1670 K under vacuum then resulted in a further reduction of mean activity to below 37 MBq/kg and the subsurface activity to 7 kBq/kg (Ref. 4). For example, stainless steel, which was exposed to pure tritium of 2.5 MPa pressure at a temperature of 720 K and absorbed 13.4 TBq/kg, was heated at a temperature of 720 K for 300 h under argon flow. This resulted in a reduction of the surface contamination from the initial level of ~ 1 GBq/m² to the level of 2 MBq/m², which was sufficiently low to undertake welding tests. Heating with flame was also tested as a predetritiation treatment before welding of stainless steel contaminated with tritium.⁴ A study⁶ of tritium removal from stainless steel using heating with flame showed that $>70\%$ of the tritium can be removed very quickly.

Some techniques were tested to decontaminate metals.^{7,8} None of the techniques, such as carbon dioxide ice pellet blasting, high-pressure water jetting, particle impact cleaning, chemical decontamination, electrochemical decontamination, or steam jetting displayed any particular ability to decontaminate steel contaminated with tritium.

The most simple and widely adopted decontamination technique is thermal desorption. A number of studies^{4,7-10} have reported how metals originally contaminated by exposure to dry gaseous tritium were decontaminated by means of thermal desorption. The presence of compounds containing hydrogen (for example, hydrogen, moisture, or ammoniac) in the gas atmosphere during thermal desorption increases the decontamination rate significantly. This is thought to be due to the isotopic exchange of tritium in the contaminated metal with the hydrogen in the hydrogen-containing gases. This decontamination process occurs even at am-

bient temperature, in particular, for tritium located in a subsurface layer of several-micrometers depth.¹¹ A significant effect of pressure of water vapor on the tritium removal rate has been observed even for very small vapor partial pressure⁹: As little as 0.0073 Pa of water vapor in the inert gas was found to be sufficient for effective tritium removal from the surface and the bulk of stainless steel when purged at elevated temperatures.¹² The presence of 5% hydrogen in argon was demonstrated¹¹ to achieve a decontamination factor (DF) of ~ 1000 for thermal desorption in the temperature range 870 to 1770 K in a static gas atmosphere. The maximum DF was observed for temperatures in the range 1070 to 1270 K, decreasing at higher temperatures of up to 1770 K. All these tests were performed with stainless steel with an initial tritium loading in the range of 10 to 100 GBq/kg (Ref. 11). Purging with air or with an inert gas containing ammonia or hydrogen removed a large proportion of tritium within several hours. It was found⁶ that temperatures of ~ 570 K were optimal both in terms of rate of tritium release and fraction of tritium removed when purging with ambient air. While thermal desorption under an air atmosphere is a very simple technique, it does require a long period of treatment to remove substantial amounts of tritium and is likely to produce a metal oxide layer that could then act as a trap for tritium.

While thermal desorption has been shown to be an effective method for processing metals contaminated by contact with gaseous tritium, only a small DF for purging with moist air was measured when decontaminating metals contaminated through exposure to tritiated water.¹³

Another simple technique is the treatment of solid waste with ozone gas. Metals exposed to gaseous tritium at ambient temperature were placed in an atmosphere containing 400 vppm ozone and heated to a temperature of 400 K for 1 h. Approximately 99% of tritium was removed from stainless steel and 80 to 86% from aluminum samples.¹⁴ However, a quite different result was reported in Ref. 15 in which the reduction of tritium surface contamination was merely 15% after exposure of the metallic surface to ozone gas over a period of a few hours.

Melting is widely regarded as a most promising technique for detritiation and volume reduction of metallic waste. As mentioned previously, a reduction of the DF has been observed for increasing the temperature from 1070 K (as used for thermal desorption prior to melting) to the melting point of stainless steel: DF of 10^4 for the thermal desorption of steel at 1070 K compared to 4×10^3 measured during the melting exercise with a circulating gas atmosphere and from 10^3 to 167, respectively, with a static atmosphere.¹¹ This effect of DF decrease is consistent with the increase of hydrogen solubility in metal with temperature rise according to the Sievert's law. Also, the solubility of hydrogen in liquid stainless steel just above the melting point is

about two times above that of solid steel just below melting point and increases further with increasing temperature. The increase of hydrogen solubility with temperature in liquid stainless steel, which is ~ 0.032 wppm/K, is much greater than that in solid stainless steel, which is 0.0034 wppm/K (Ref. 16).

Most of the tritium is likely to be released from the metal as tritiated water vapor at relatively low temperatures early in the heating/melting operation. However, reaching a very small residual tritium concentration in the decontaminated metal (below the ILW/LLW threshold in particular) has proven to be difficult.⁷ There is evidence for stainless steel retaining a noticeable quantity of tritium even at a temperature near the melting point.¹¹

One of the possible ways to improve decontamination by melting is purging the melt with gas to produce small bubbles: This both increases the rate of tritium removal from the melt of metal and reduces the residual tritium concentration.¹⁷ Theoretical consideration shows that the rate of process increases and the residual tritium content decreases with reduction of the gas bubble size.¹⁸

Applying glow discharge cleaning to tritiated stainless steel reduced the activity sorbed on the surface by a factor of 250 or larger.^{10,19} However, subsequent regrowth of tritium at the decontaminated surface has been observed.¹⁹

Employing chemical and electrochemical detritiation of metals allows²⁰⁻²² removing a large portion of the surface's tritium and some tritium from the bulk. However, this treatment causes production of mixed waste, i.e., waste being both chemically and radiologically hazardous.

Most of the studies referred to above were performed with samples of metal loaded with tritium at different conditions and using different methods for tritium analysis. This makes comparative evaluation of the detritiation technique's effectiveness unreliable. In this study, the effectiveness of several detritiation methods was assessed using samples of several metals loaded with tritium under identical conditions and the same techniques for tritium analysis.

II. EXPERIMENT

II.A. Sample Preparation

Coupons of Type 316L stainless steel, INCONEL® 600, beryllium, low-oxygen copper, and aluminum bronze, representative of metals present in the JET vacuum vessel and in the AGHS, were contaminated through exposure to a hydrogen-tritium gas mixture. Prior to exposure, the coupons were cleaned using the procedure adopted at JET for cleaning of in-vessel components and then baked under vacuum at a temperature of 770 K for 12 h. All coupons were then exposed to a gas mixture containing 50 ± 2 vol% tritium in hydrogen at a pressure of 0.05

MPa for a period of 8 h. The coupons were held at 470 K during the tritium-loading process, apart from the beryllium sample, which was at 570 K. The coupons were 100×40 mm with a thickness of ~ 6 mm. A detailed description of the sample preparation is given elsewhere.²³

II.B. Tritium Analysis

Two techniques, namely, radioluminography (RLG) and complete acid dissolution (CAD), were employed to measure the tritium content and distribution in pretest and posttest samples.²³ The RLG method is based on registering photostimulated luminescence (PSL) caused by the interaction of tritium beta particles with the material of an image plate attached to the sample being analyzed. The RLG method is capable of measuring the tritium distribution along the surface analyzed by detecting beta particles escaped from the subsurface layer of depth smaller than the mean free path of the particles. To evaluate tritium depth profiles, the samples were cut transversely, and an image plate was then applied to the newly exposed surface. It was found that the intensity of the PSL rises with the time elapsed after exposing the surface for analysis, so images were obtained within the first 4 h of the sample being cut. The tritium depth profiles were then normalized to the reference time of 4 h as described in Ref. 23. Evaluation of tritium inventories using the RLG measurements was based on integration of the normalized depth profiles. The tritium inventories were also measured by complete dissolution of the metal in an aqueous acid solution. On completion of the dissolution, the acid was neutralized and then distilled to obtain a sample of pure water for the activity to be measured by liquid scintillation counting (LSC). Because the tritium depth profiles were normalized to the reference time of 4 h, the tritium inventories evaluated from those profiles have to present values larger than that measured by the CAD method.²³

II.C. Detritiation Methods

Detritiation methods were tested using samples of 100-mm length, 40-mm width, and ~ 6 -mm thickness, apart from those for the melting tests (which measured 15 mm in length and 7 mm in width) and heating-with-flame samples 50×40 mm.

The following detritiation methods were tested: thermal desorption, isotopic exchange with gaseous hydrogen, isotopic replacement with hydrogen, heating with flame, and melting, as discussed in Secs. II.C.1 through II.C.5.

II.C.1. Thermal Desorption

This is the most commonly employed method for recovering tritium from solid materials. In this study thermal desorption was carried out with air at atmospheric

pressure and at a temperature of 570 K (this was observed⁶ to be an optimal temperature in terms of the tritium removal rate and the fraction of tritium inventory removed under purging with air). The samples were placed in a glass reactor and purged with air for 5 h. Air coming out of the reactor passes through two bubblers connected in series. The first bubbler was filled with demineralized water, and the second was filled with water and a hydrophobic catalyst, which promotes conversion of gaseous hydrogen into water. Therefore, tritium released from the metal both in the form of water vapor and molecular hydrogen was collected in the bubblers as tritiated water, and the activity was measured by LSC. The efficiency of the bubblers for tritium collection was $\sim 99\%$. The air velocity through the reactor was the same for all the thermal desorption tests. The water in the bubblers was replaced at 30-min intervals.

II.C.2. Isotopic Exchange with Gaseous Hydrogen

A quartz reactor was loaded with a sample and evacuated to below 0.1 Pa before heating to the desired temperature. It was then backfilled with hydrogen to a pressure of 0.05 MPa for 1 h at operating temperatures of 570 and 770 K for 0.5 h at temperatures of 870 and 970 K. After this time, the hydrogen was removed for tritium analysis and replaced with fresh tritium-free hydrogen. This was repeated another four times. The residual content of tritium in the decontaminated samples and the tritium depth profiles were measured using the CAD and the RLG techniques.

II.C.3. Isotopic Replacement with Hydrogen

For this process, the sample was sealed between two chambers: one charged with hydrogen at 0.2 MPa and the other continuously vacuum pumped. The sample of metal to be detritiated acts as a membrane for hydrogen permeation. The residual content of tritium in decontaminated samples and the tritium depth profiles were measured using the CAD and the RLG techniques.

II.C.4. Heating with Flame

The sample of metal to be decontaminated was placed inside an enclosure and heated using a hydrogen-oxygen flame until it reached a cherry-red color (this typically took 1.5 to 3 min). Heating was applied first to one face and then to its opposite before both faces were heated together. During heating and for 1 h afterward, the enclosure was purged with air with the exhaust vented through two water bubblers connected in series, as mentioned above. Tritium released from the metal and collected as tritiated water in the bubblers was assayed by LSC.

II.C.5. Melting

Three melting processes were tested: melting under vacuum, melting under a flow of argon, and melting under

a static atmosphere of hydrogen. For vacuum melting, the sample was placed in a graphite crucible within a quartz vacuum chamber of 0.7-dm³ volume. The chamber was vacuum pumped and maintained at a residual gas pressure of ~ 3 Pa. The sample then was heated and melted using an induction heater installed around the vacuum chamber. The sample was kept at melting temperature for 0.5 h and then cooled down under vacuum. The ingot produced was then split into two halves; one-half was analyzed for residual tritium, and the other was subjected to a further melt. Residual tritium was measured by the complete dissolution method.

The procedure for melting the sample under an inert gas flow was similar to that for vacuum melting. The difference included purging the chamber with argon at a flow rate of 0.2 dm³/min and collecting the argon with released tritium in a buffer tank. The sample was maintained at the melting temperature for 1 h. Samples of the collected gas were withdrawn after 30 and 60 min of heating from the buffer tank for tritium analysis using an ionization chamber: The results show that detritiation was nearly complete in the first 30 min. Residual tritium in the decontaminated sample was measured by the method of complete dissolution.

The procedure for melting under a hydrogen atmosphere was also similar to that for vacuum melting; the difference included backfilling the chamber with 0.05 MPa of hydrogen before heating began (only a small fraction of hydrogen around the crucible was heated; the rest remained at approximately ambient temperature). At the end of heating, a sample of hydrogen was withdrawn to analyze the tritium content. Residual tritium was measured by complete dissolution of metal.

The melt chamber was cleaned of residual tritium after each decontamination test by flushing with tritium-free hydrogen, and the background level of tritium was determined before each test by carrying out the test without a sample of tritiated metal.

III. RESULTS AND DISCUSSION

III.A. Testing Detritiation Methods

The tritium concentrations on the surface and the tritium inventories of the pretest metal samples are listed in Table I. The tritium distribution through the thickness of the sample is illustrated as depth profiles; see Figs. 1 through 5 (Ref. 23). All the contaminated metal samples have a thick subsurface layer highly enriched with tritium.

The JET vacuum vessel was designed to be baked out at temperatures up to 770 K and to operate at temperatures up to 620 K. The operating temperature is limited by the maximum temperature allowed for some components of the pumped divertors. In this study the testing of detritiation methods to be employed for

TABLE I
Tritium Inventory and Concentration on the Surface of Original Metals*

Metal	Tritium Inventory (MBq/g)	Tritium Concentration on Surface	
		kBq/cm ²	MBq/g
Stainless steel	46.8	310	1680
INCONEL®	53.6	440	2400
Copper	1.5	990	5000
Beryllium	25.8	340	2740
Aluminum bronze	14.0	680	3580

*Reference 23. Measurements were performed using the RLG method. Surface concentrations are given for subsurface layer of thickness equal to the depth of mean free path of tritium beta particles of 5.6-keV energy. Thickness of these layers is ~0.2 μm for stainless steel, INCONEL®, copper, and aluminum bronze and ~0.7 μm for beryllium.

decontaminating the vacuum vessel was limited in temperature to 770 K.

The following parameters have been used to assess the effectiveness of detritiation techniques: the fraction of the tritium inventory removed, the reduction in the rate of tritium outgassing, and the residual tritium concentration in the decontaminated material. The first two parameters are more important for the in-vessel decontamination. The last parameter is a main factor for the disposal of waste. One of the goals for this study was to determine a technique that could provide a reduction of tritium inventory to below the ILW/LLW threshold, which

is 12 kBq/g in the United Kingdom for gamma and beta emitters.

III.A.1. Thermal Desorption

Decontamination of tritiated metals in thermal desorption under an air atmosphere involves desorption of tritium-containing species from the metal surface and their isotopic exchange with water vapor in the air. The rate of tritium release from stainless steel in thermal desorption under the atmosphere of ambient air is illustrated in Fig. 6, where it can be seen that the rate of release halves after the first 5 h. Analyses of tritium inventories in pretest and posttest samples performed using both the CAD and the RLG techniques (see Table II) show that >65% of the tritium inventory can be removed from stainless steel and INCONEL® by a 5-h thermal desorption in air at a temperature of 570 K.

Figure 7 compares the tritium depth profiles in the pretest and the posttest samples measured using the RLG technique. As a result of heating, tritium was quantitatively removed from the subsurface layer, which was originally highly enriched with tritium. However, tritium was also driven from this layer down to the bulk. For example, the tritium concentration in the bulk of the INCONEL® sample increased from 0.9 MBq/g in the pretest sample to 2.8 MBq/g in the posttest sample. Tritium driven from the surface into the bulk makes a deep detritiation of metals more difficult.

III.A.2. Isotopic Exchange with Gaseous Hydrogen

Gaseous hydrogen isotopically exchanges with tritium to decontaminate the metal's surface. Because of its high mobility,²³ tritium from the bulk diffuses to the

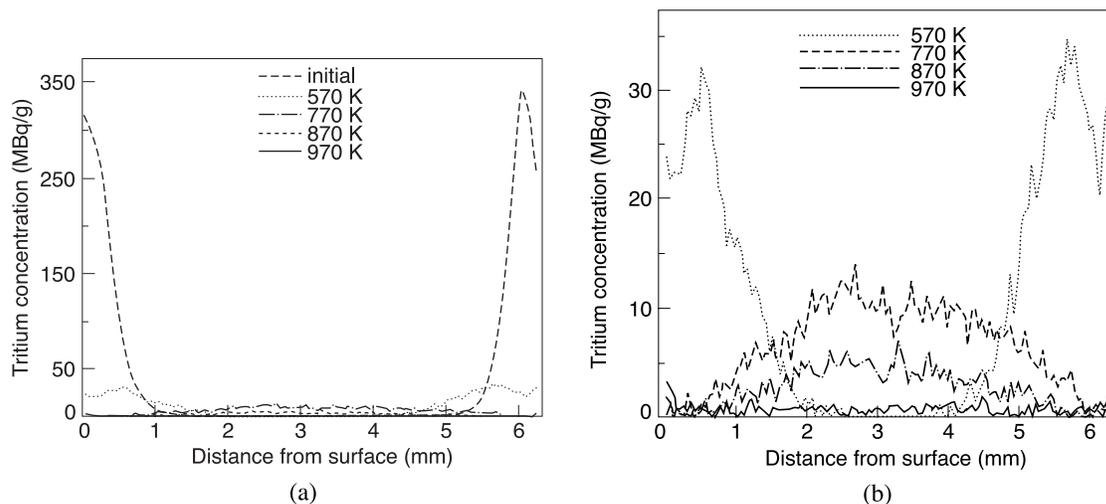


Fig. 1. (a) Comparing tritium depth profiles in original and decontaminated samples of stainless steel and (b) tritium depth profiles in samples decontaminated at different temperatures.

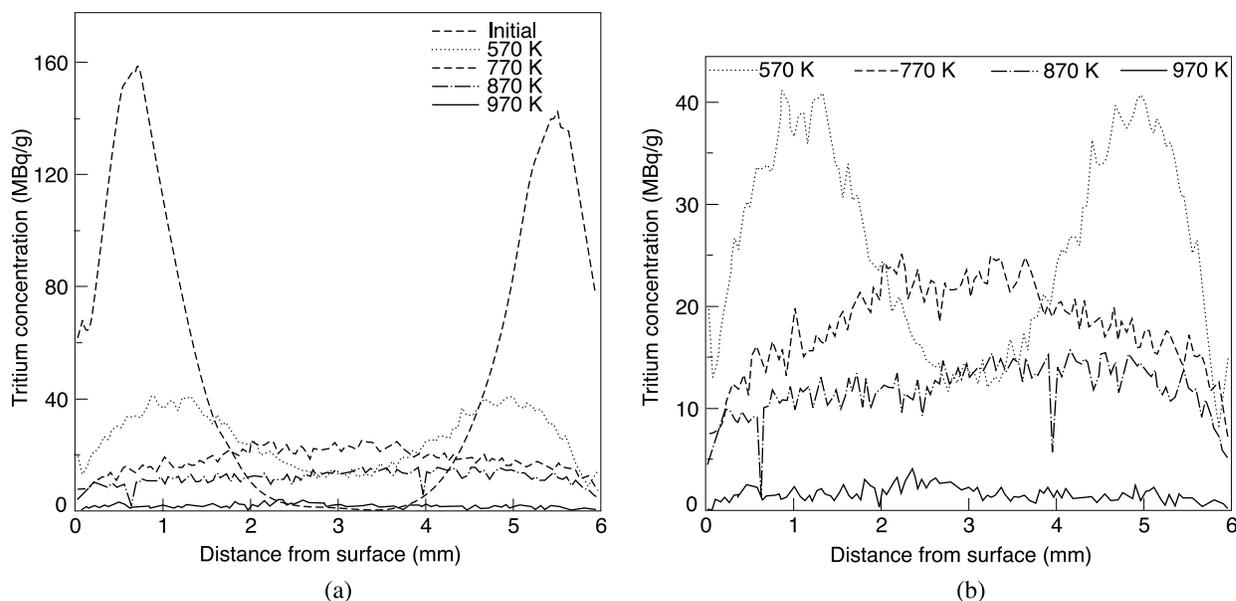


Fig. 2. (a) Comparing tritium depth profiles in original and decontaminated samples of INCONEL® and (b) tritium depth profiles in samples decontaminated at different temperatures.

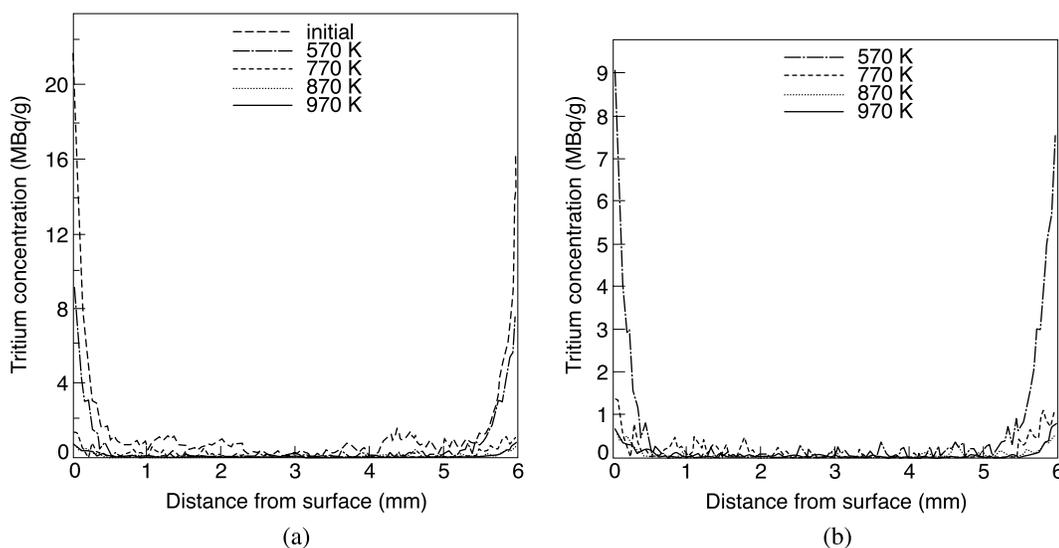


Fig. 3. (a) Comparing tritium depth profiles in original and decontaminated samples of copper and (b) tritium depth profiles in samples decontaminated at different temperatures.

decontaminated surface; therefore, both the surface and the bulk of the material will undergo detrutiation when exposed to gaseous hydrogen. Figure 8 presents the kinetics of INCONEL® detrutiation in five consecutive exposures to hydrogen. Separate, but identically prepared samples were used for tests at each temperature. The concentrations of tritium collected in the hydrogen by the

end of the exposure period are shown in Fig. 8 for different temperatures. The tritium concentrations are quite similar in all exposures at 570 K, whereas at temperatures of 770 K or higher, the first exposure removes more tritium from INCONEL® than those that follow. The quantity of tritium removed with the first exposure rises with a temperature increase, although the rate of tritium

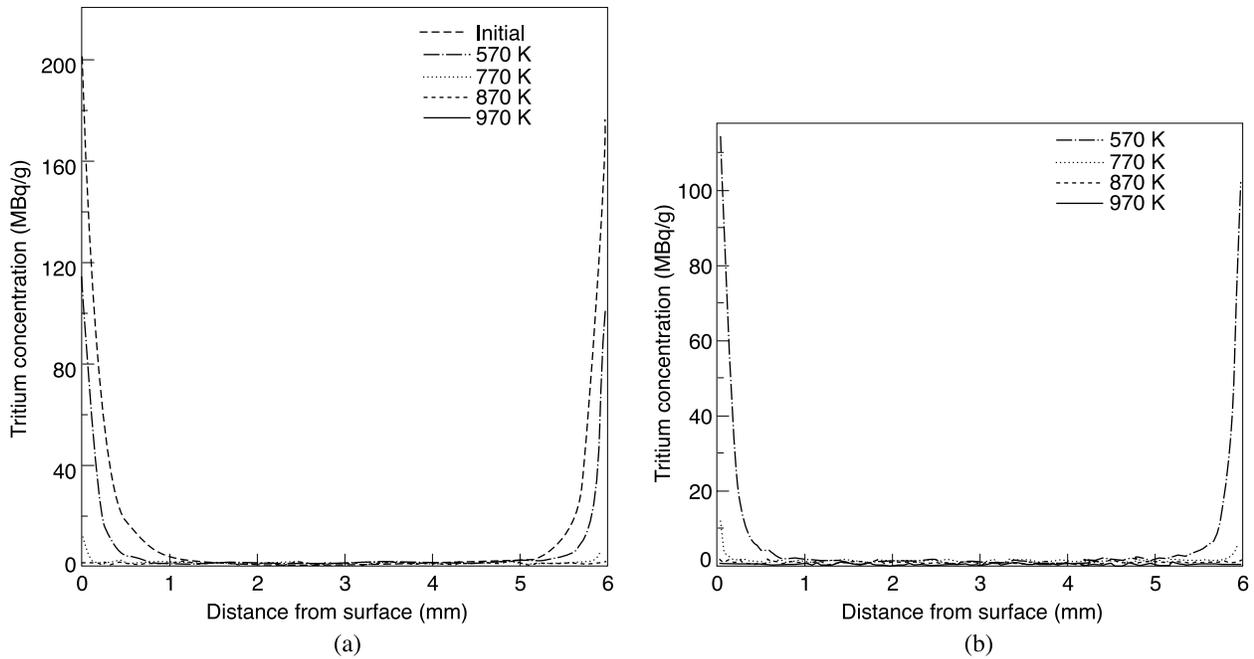


Fig. 4. (a) Comparing tritium depth profiles in original and decontaminated samples of aluminum bronze and (b) tritium depth profiles in samples decontaminated at different temperatures.

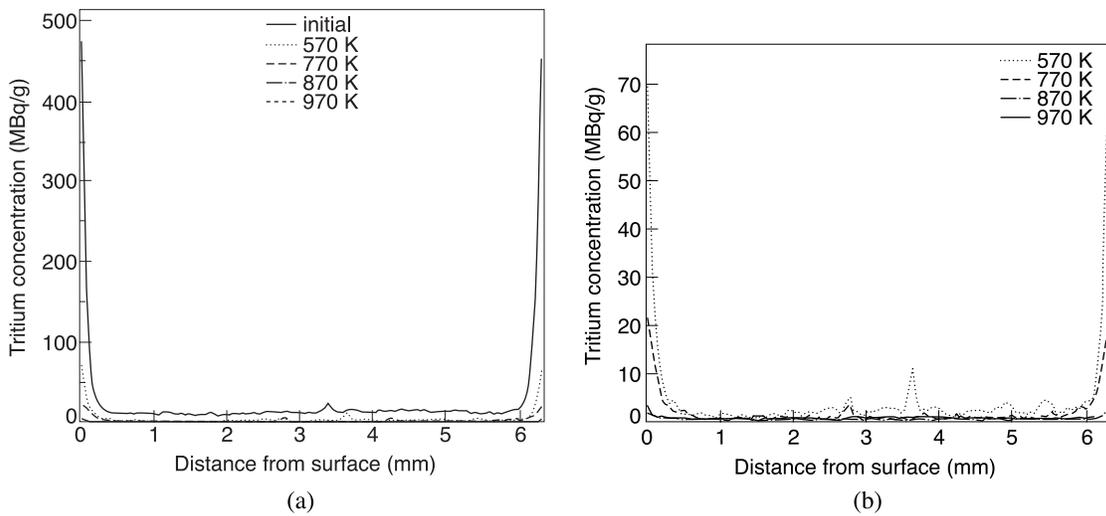


Fig. 5. (a) Comparing tritium depth profiles in original and decontaminated samples of beryllium and (b) tritium depth profiles in samples decontaminated at different temperatures.

removal with second and subsequent exposures is almost independent of temperature. Similar behavior was observed for the other metals studied.

After the completion of decontamination, the samples were analyzed for the tritium concentration on the surface and in the bulk. Table III shows the tritium concentrations on the surface. The temperature dependen-

cies of the residual tritium inventory in decontaminated metals, presented in Figs. 9, 10, and 11, indicate that the temperature limit of 770 K would in most cases be sufficient to achieve low residual tritium inventories.

As illustrated by the tritium depth profiles in decontaminated metals (see Figs. 1 through 5), isotopic exchange with gaseous hydrogen removes tritium from

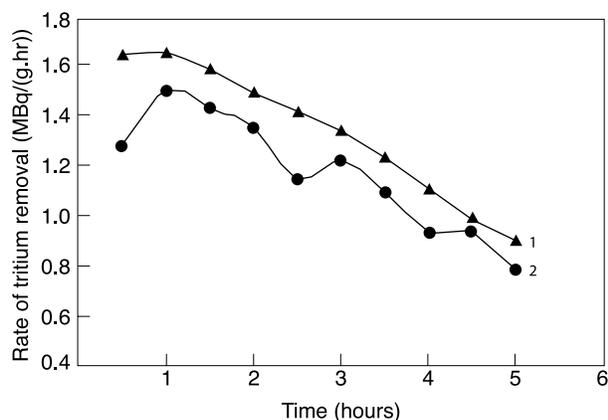


Fig. 6. Rate of tritium release from stainless steel (1) and INCONEL® (2) in thermal desorption under ambient air at temperature of 570 K.

TABLE II

Fraction of Tritium Inventory Removed and DF for Thermal Desorption Under Ambient Air at Temperature of 570 K

Metal	Analysis	Tritium Inventory (MBq/g)		F^a (%)	DF ^b
		Pretest	Posttest		
Stainless steel	CAD	38.1	13.3	65	2.9
	RLG	46.8	10.2	78	4.5
INCONEL®	CAD	40.7	11.5	72	3.5
	RLG	53.6	9.4	82	5.7

^aExtraction factor $F = 1 - C_F/C_O$, where C_O and C_F are the tritium inventories in pretest and posttest samples.

^b $DF = C_O/C_F$; $DF = 1/(1 - F)$.

TABLE III

Tritium Concentration on Surfaces of Metals Prior to and After Decontamination Tests

Metal	Surface Tritium Concentration (MBq/g)				
	Original	Decontaminated at Temperature			
		570 K	770 K	870 K	970 K
Stainless steel	1680	550	66	17	17
INCONEL®	2400	590	54	82	9
Copper	5000	2380	265	4.5	3
Aluminum bronze	3580	4010	14	18	14
Beryllium	2740	1580	176	21	6

the subsurface layer enriched with tritium. To eliminate that layer, a temperature of 770 K or larger was needed. Isotopic exchange at a temperature of 870 K or larger led to a “flat” tritium depth profile in copper, aluminum bronze, and beryllium with a tritium concentration of ~ 24 kBq/g (at distance of ~ 300 μm from the surface).

The fraction of tritium inventory removed F and the DF given in Table IV show a large portion of tritium removed from the metals by the simple procedure of exposure to gaseous hydrogen at moderate temperatures. The measured rate of tritium outgassing from the pretest and the posttest samples at room temperature under air flow does not appear to change as a result of the decontamination process, apart from INCONEL®, which demonstrated an obvious reduction in tritium outgassing from decontaminated samples.

The DFs listed in Table IV and evaluated using the tritium inventories measured by the CAD technique are always larger than those based on the RLG measurements. Because of the phenomenon of tritium concentration rising on the surface analyzed by the RLG technique²³ in time elapsing after the surface production, the DFs derived from the RLG measurements are used here for intercomparison only. The DFs derived from the CAD measurements shall be used as the reference values.

III.A.3. Isotopic Replacement with Hydrogen

Exposing one side of metal contaminated with tritium to gaseous hydrogen and providing a driving force for the hydrogen to diffuse through the metal resulted in tritium being isotopically exchanged with hydrogen and driven out of the metal. Table V demonstrates that the tritium concentrations on the surfaces exposed both to the static hydrogen atmosphere and to vacuum were reduced substantially after 70 h of decontamination at moderate temperatures. The residual tritium inventories in the decontaminated samples (see Table VI) show that 88% or more of tritium can be removed using this method. The tritium depth profile in Fig. 12 clearly confirms that tritium was driven from the side of the metal exposed to hydrogen through the metal to the surface exposed to vacuum. The results of stainless steel decontamination at temperatures of 623 and 723 K for 70 h are given in Tables V and VI.

III.A.4. Heating with Flame

Heating metal using a flame provides two driving forces for detritiation: tritium thermal desorption, which is promoted by elevated temperatures of both heated and unheated surfaces, and isotopic exchange between hydrogen containing ions produced in the flame with tritium on the surface of the metal. The tritium concentrations on the heated and the opposite surfaces of the samples before and after decontamination are given in Table VII.

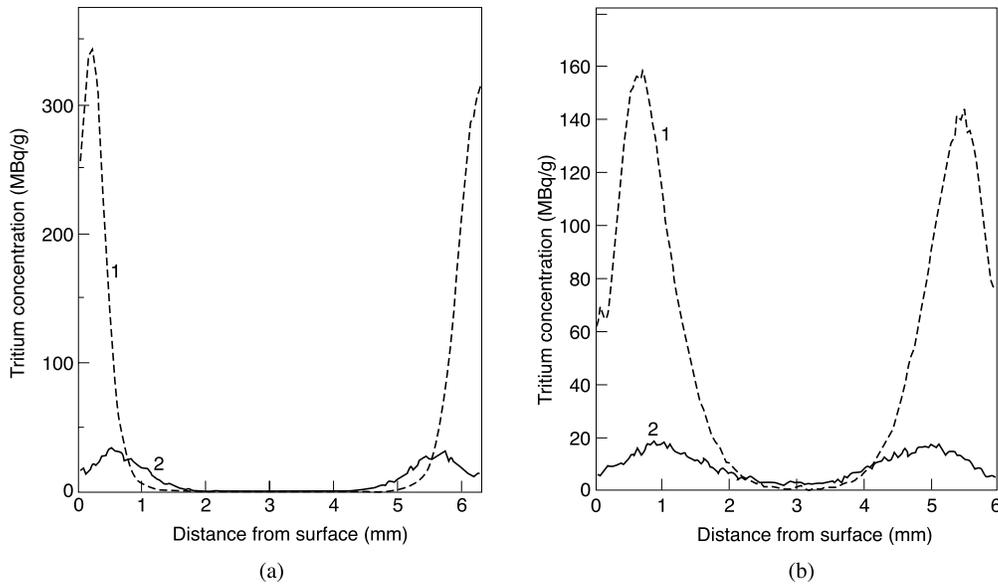


Fig. 7. Tritium depth profiles in pretest (1) and posttest (2) samples of (a) stainless steel and (b) INCONEL®.

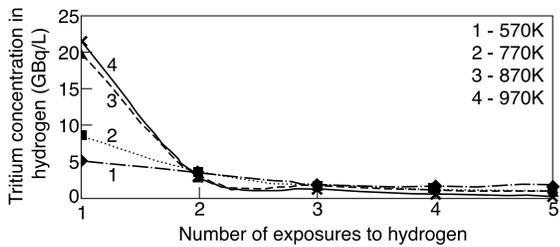


Fig. 8. Tritium concentration in gaseous hydrogen after isotopic exchange with tritium in sample of INCONEL®.

Table VIII presents the tritium inventories in the pretest and the posttest samples measured using the RLG and the CAD techniques.

The results given in Table VII demonstrate that flame heating to obtain high temperatures throughout the metal sample reduces the tritium concentration both on the surface exposed to the flame and on the opposite surface. A single heating cycle was sufficient to achieve a low concentration on the surfaces of decontaminated copper, beryllium, and aluminum bronze. Metals

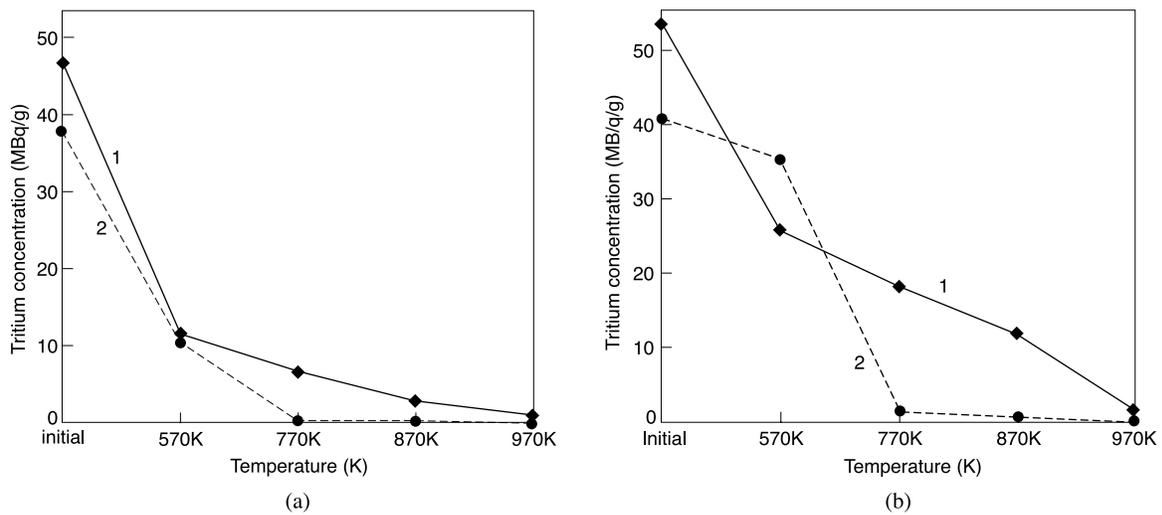


Fig. 9. Temperature dependencies of residual tritium inventory in decontaminated (a) stainless steel and (b) INCONEL® measured using RLG (1) and CAD (2) techniques.

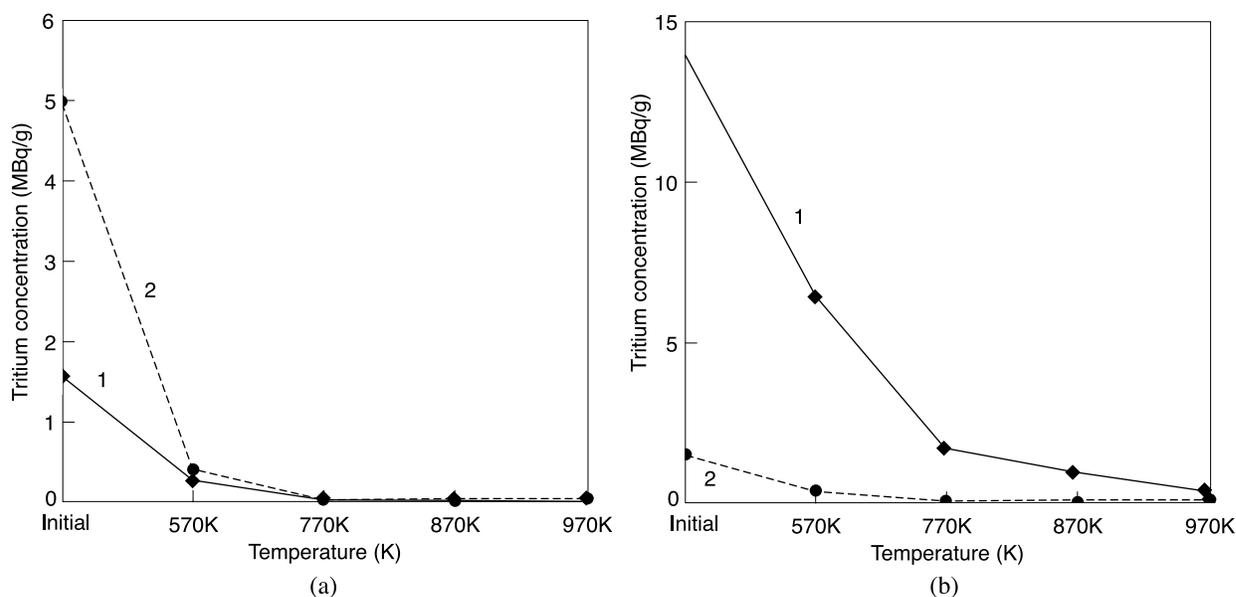


Fig. 10. Temperature dependencies of residual tritium inventory in decontaminated (a) copper and (b) aluminum bronze measured using RLG (1) and CAD (2) techniques.

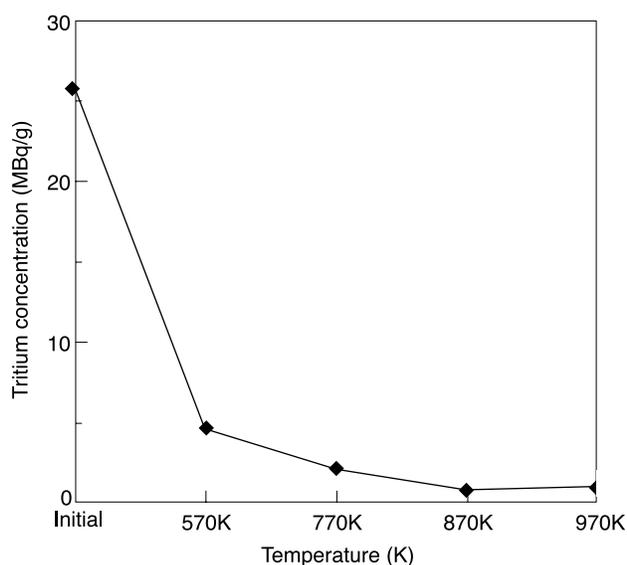


Fig. 11. Temperature dependencies of residual tritium inventory in decontaminated beryllium measured using the RLG technique.

having a large initial tritium inventory, such as stainless steel and INCONEL[®], required additional heating cycles.

Comparing tritium depth profiles in original and decontaminated samples in Fig. 13 shows that heating with flame removes tritium from the tritium-enriched subsurface layer but also drives it into the bulk. Achieving a low

residual tritium inventory using this decontamination method was difficult. However, Table VIII confirms that heating with flame allows removal of a large portion of the tritium inventory in a short period of time.

III.A.5. Melting

Depending on the method of melting employed, decontamination can be driven either by thermal desorption alone (as for melting under vacuum or argon purge) or in combination with tritium isotopic exchange (as for melting under a hydrogen atmosphere). Melting under vacuum and argon purge differ only by means of providing a low hydrogen pressure over the melt. As melting results in a quite uniform distribution of residual tritium throughout the produced ingot, the tritium concentration in the posttest samples was measured by the CAD technique only.

III.A.5.1. Melting Under Vacuum. Table IX presents results of two consecutive vacuum meltings.

III.A.5.2. Melting Under Flow of Argon. The results of the tests are given in Table X.

III.A.5.3. Melting Under Hydrogen Atmosphere. The results are given in Table XI. More than 99% of the tritium inventory was released from all the metals at the first melting, and the residual tritium concentration was considerably smaller than the ILW/LLW threshold.

It should be noted that because of the limited size of the crucibles, melting tests were carried out with samples having a mass an order of magnitude smaller than those

TABLE IV
Effectiveness of Detritiation by Isotopic Exchange with Gaseous Hydrogen

Metal	T (K)	Method of Tritium Measurement								
		Complete Acid Dissolution				RLG				
		C_O (MBq/g)	C_F (MBq/g)	F (%)	DF	C_O (MBq/g)	C_F (MBq/g)	F (%)	DF	
Stainless steel	570	38.1	10.4	73	3.7	46.8	11.5	75	4	
	770			99.5	200			6.4	86	7
	870			99.7	390			2.8	94	16.5
	970			99.8	490			0.95	98	49
INCONEL®	570	40.7	35.2	14	1.2	53.6	25.7	52	2	
	770			96	27			18.1	66	3
	870			98.5	65			11.8	78	4.5
	970			99.7	314			1.6	97	34
Copper	570	5.0	0.41	92	12	1.6	0.265	83	6	
	770			99.4	175			0.04	97.5	40
	870			99.9	900			0.035	97.7	44
	970			99.9	900			0.03	98	49
Aluminum bronze	570	1.5	0.32	79	5	14	6.4	54	2	
	770			94	16			1.65	88	8
	870			96	24			0.95	93	15
	970			97	31			0.37	97	38
Beryllium	570	—	—	—	—	25.8	4.5	82	6	
	770			—	—			2.0	92	13
	870			—	—			0.68	97	38
	970			—	—			0.93	96	28

TABLE V
Tritium Concentration on Surface of Original and Decontaminated Samples of Stainless Steel

Side of Sample	Tritium Concentration on Surface (MBq/g)		
	Original	Decontaminated at Temperature	
		623 K	723 K
Exposed to hydrogen	1920	14	5
Exposed to vacuum	1470	210	33

used for other decontamination trials. Comparison between the melting methods that were tested demonstrated that isotopic exchange with a gaseous hydrogen atmosphere contributes substantially to detritiating the metal.

III.B. Comparative Effectiveness of Detritiation Methods

Table XII compares different detritiation methods in terms of fractions of the tritium inventory removed and the DFs achieved. The residual tritium concentration in decontaminated metals is compared with the ILW/LLW threshold, which is 12 kBq/g in the United Kingdom.

IV. SUMMARY AND CONCLUSIONS

Procedures based on any one, or combination, of the three methods (thermal desorption under flow of ambient air, isotopic exchange with gaseous hydrogen, and isotopic replacement with hydrogen) could be employed to decontaminate the JET vacuum vessel at such times as preparing it for modification or disassembly. The current design of the JET vacuum vessel allows the performing of such decontamination. The methods will remove a large fraction of the tritium inventory (by at least an order of magnitude for stainless steel) and reduce the rate

TABLE VI

Tritium Inventory in Original and Decontaminated Samples of Stainless Steel

Sample	Tritium Inventory (MBq/g)		Fraction of Tritium Inventory Removed, F (%)	
	RLG	CAD	RLG	CAD
Original	46.8	38.1		
Decontaminated at 623 K	5.5	1.1	88	97
Decontaminated at 723 K	0.7	0.03	>98	>99

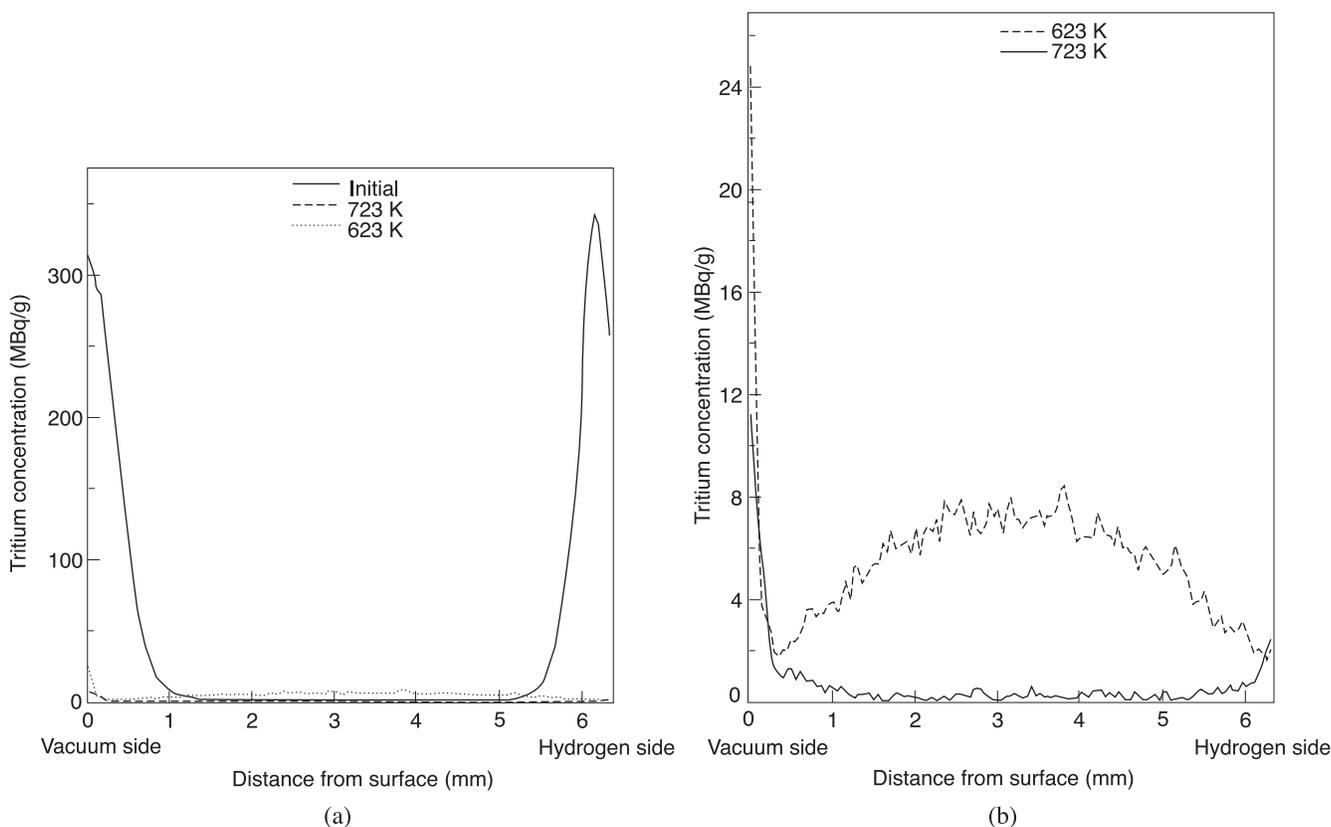


Fig. 12. (a) Comparing tritium depth profiles in original and decontaminated stainless steel and (b) tritium depth profiles in steel decontaminated at 623 and 723 K.

of tritium outgassing. Thermal desorption is the least attractive of the methods because it is likely to produce oxides in the metal's sub-surface layer that then will act as a barrier for tritium migration and as a trap for tritium, making further decontamination more difficult. Isotopic replacement will work mostly for the inner wall of the vessel, and the in-vessel components will also be subjected to decontamination through isotopic exchange with gaseous hydrogen.

Heating with flame could be employed but is less effective and would need an engineering solution to provide a source of heating. Heating with flame might lead to rupture of the vessel wall due to thermally induced stresses resulting from non-uniform heating. However, this method could be a simple and effective solution for decontamination of parts of the tritium plant prior to and after its disassembly.

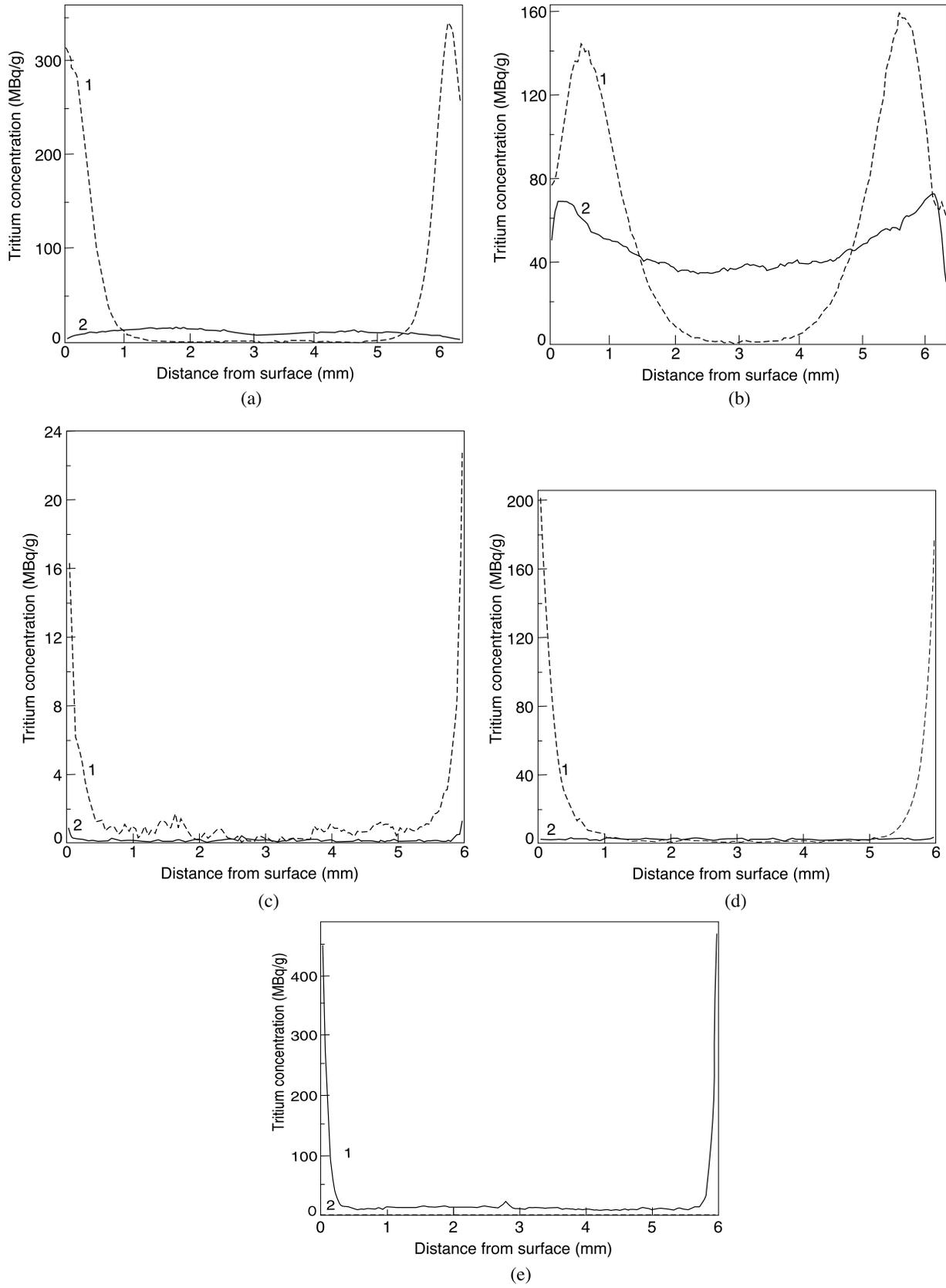


Fig. 13. Comparing tritium depth profiles in pretest (1) and posttest (2) samples of (a) stainless steel, (b) INCONEL®, (c) copper, (d) aluminum bronze, and (e) beryllium decontaminated in three consecutive heatings with flame.

TABLE VII

Tritium Concentrations on Heated and Opposite Surfaces of Metals Before and After Decontamination

Metal	Sample	Tritium Concentration on Surface (kBq/cm ²)	
		Surface 1	Surface 2
Stainless steel	Original	310	
	After heating surface 1	180	200
	After heating surface 2	186	1.5
	After heating both surfaces	1.1	0.5
INCONEL®	Original	440	
	After heating surface 1	47	98
	After heating surface 2	31	23
	After heating both surfaces	13.6	11.4
Copper	Original	990	
	After heating surface 1	0.2	0.1
Aluminum bronze	Original	680	
	After heating surface 1	1.7	1.4
	After heating surface 2	1.6	0.8
	After heating both surfaces	1.3	0.8
Beryllium	Original	340	
	After heating surface 1	0.3	0.8
	After heating surface 2	0.2	0.1
	After heating both surfaces	0.1	0.1

TABLE VIII

Tritium Inventory in Pretest and Posttest Metals Decontaminated in Three Consecutive Heatings with Flame*

Metal	Tritium Inventory (MBq/g)				Fraction of Tritium Removed, <i>F</i> (%)	
	<i>C_o</i>		<i>C_F</i>			
	RLG	CAD	RLG	CAD		
Stainless steel	46.8	38.1	10.8	7.4	77	80
INCONEL®	53.6	40.7	46.3	14.3	14	65
Copper	1.6	5.0	0.12	0.13	92	97
Aluminum bronze	14.0	1.5	2.0	0.1	86	93
Beryllium	25.8	—	0.4	—	98	—

*As indicated in Table VII.

TABLE IX

Results of Detritiation by Vacuum Melting

Metal	<i>C_o</i> (MBq/g)	First Melting			Second Melting		
		<i>C_F</i> (MBq/g)	<i>F</i> (%)	DF	<i>C_F</i> (MBq/g)	<i>F</i> (%) (Total)	DF (Total)
Stainless steel	38.1	0.87	97.7	44	0.26	99.3	147
Stainless steel	38.1	0.63	98.4	61	0.34	99.1	113
INCONEL®	40.7	0.03	99.9	1222	0.013	99.97	3056
Copper ^a	5.0	0.16	96.8	31	0.0046	99.9	1087

^aCopper evaporation and carrying out of the melting crucible were observed during keeping liquid copper at the melting temperature. Tritium concentration in that evaporated copper, which then was condensed and collected, was measured as 0.066 MBq/g.

TABLE X

Results of Detritiation by Melting Under Argon Flow

Metal	C_o (MBq/g)	C_F (kBq/g)	F (%)	DF
Stainless steel	38.1	1.55	$\gg 99.9$	24 600
INCONEL®	40.7	80.3	99.8	510
Copper	5.0	18.4	99.6	270

All tested methods, apart from isotopic replacement with hydrogen, can be employed to decontaminate the dismantled parts of the JET vacuum vessel and the tritium plant. At this stage a higher temperature for decontamination can be used, and only by melting the metal could a DF of $>10^4$ be achieved, giving a residual tritium inventory close to or even below the ILW/LLW

TABLE XI

Results of Decontamination Tests by Melting Under Hydrogen Atmosphere*

Metal	C_o (MBq/g)	C_F^a (kBq/g)	F (%)	DF (Total)
Stainless steel	38.1	0.80	>99.9	38 720
INCONEL®	40.7	1.6	>99.9	25 400
Copper	5.0	<0.3	>99.9	$>1 600$
Aluminum bronze	1.5	<0.3	>99.9	$>5 000$

*Two tests performed for samples of INCONEL®, copper, and aluminum bronze showed good reproducibility.

^aDetection limit of CAD technique was 0.3 kBq/g.

threshold. Melting under hydrogen atmosphere is more effective in this respect than the other tested modifications of the melting technique.

TABLE XII

Comparative Effectiveness of Detritiation Methods*

Method	Metal	F (%)	DF	Residual Tritium Inventory
Thermal desorption (under air at 570 K)	Stainless steel	78	4.5	\gg ILW/LLW
	INCONEL®	82	5.6	\gg ILW/LLW
Isotopic exchange with gaseous hydrogen (570 K)	Stainless steel	75	4	\gg ILW/LLW
	INCONEL®	52	2	\gg ILW/LLW
	Copper	83	6	\gg ILW/LLW
	Aluminum bronze	54	2	\gg ILW/LLW
	Beryllium	82	6	\gg ILW/LLW
Isotopic exchange with gaseous hydrogen (770 K)	Stainless steel	86	7	\gg ILW/LLW
	INCONEL®	66	3	\gg ILW/LLW
	Copper	97	33	\gg ILW/LLW
	Aluminum bronze	88	8	\gg ILW/LLW
	Beryllium	92	12.5	\gg ILW/LLW
Isotopic replacement with hydrogen (623 K)	Stainless steel	88	8	\gg ILW/LLW
Isotopic replacement with hydrogen (723 K)	Stainless steel	>98	>50	\approx ILW/LLW
Three consecutive heatings with flame	Stainless steel	77	4	\gg ILW/LLW
	INCONEL®	14	>1	\gg ILW/LLW
	Copper	92	12.5	\gg ILW/LLW
	Aluminum bronze	86	7	\gg ILW/LLW
	Beryllium	98	50	\gg ILW/LLW
Vacuum melting	Stainless steel	>99	130	\gg ILW/LLW
	INCONEL®	>99.9	3 060	\approx ILW/LLW
	Copper	>99.9	1 090	$<$ ILW/LLW
Melting under argon flow	Stainless steel	>99.9	24 600	$<$ ILW/LLW
	INCONEL®	>99.8	500	\gg ILW/LLW
	Copper	>99.6	300	\approx ILW/LLW
Melting under hydrogen atmosphere	Stainless steel	>99.9	$>38 700$	$<$ ILW/LLW
	INCONEL®	>99.9	$>25 400$	$<$ ILW/LLW
	Copper	>99.9	$>1 600$	$<$ ILW/LLW
	Aluminum bronze	>99.9	$>5 000$	\ll ILW/LLW

*Value F is based on tritium measured using the RLG method, except melting tests, in which the CAD method was used.

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