JET-P(99)02

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Operational Experience with the JET Impurity Processing System During and After DTE1

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Preprint of a Paper to be submitted for publication in Fusion Engineering and Design

March 1999

ABSTRACT

The hydrogen containing gases generated during operation and maintenance of a fusion machine need to be detritiated prior their discharge to the environment. The amounts, chemical and isotopic compositions of gas impurities which were decontaminated in JET Impurity Processing system during and after deuterium- tritium experiment, DTE1, are presented. Two different techniques for impurities detritiation were tested. The first technique is based on hydrogen containing species catalytic oxidation followed by water decomposition in reaction with uranium at elevated temperature. The second technique is direct decomposition of impurities with molecular hydrogen liberation in reactions with uranium at elevated temperatures. The results of the impurities detritiation using both techniques are given.

1. INTRODUCTION

The Impurity Processing system (IP) is an integral part of the JET Active Gas Handling System (AGHS) [1]. IP was designed for tritium removal from hydrogen containing gases, such as hydrocarbons, water, etc., prior their discharge to the environment via the Exhaust Detritiation (ED) system [2]. Gas mixtures containing tritium in a chemically bonded form are generated during both the torus operation for D-T experiments, and during maintenance of the torus and AGHS. The amount and chemical composition of the gas mixtures to be decontaminated in IP can be classified into three general categories (non-hydrogen gases are referred to below as impurities):

Category 1 includes impurities collected on the cryopumps within the torus vacuum vessel and the Neutral Injector Beamlines (NIBs) during DTE1 operations.

Tritium and deuterium introduced into the Neutral Beam (NB) system and into the JET torus were pumped onto the NIB and Pumped Divertor (PD) cryopumps respectively [3], [4]. These cryopumps were periodically regenerated by warming their Helium panels (normally at 4-5K) to a temperature of 77K. This released the tritium and deuterium for reprocessing [1], [3], and also the condensed impurities.

These impurities are separated from molecular hydrogen isotopes in the AGHS Cryogenic Forevacuum (CF) [1] system prior to transfer to IP. Impurities consist mainly of helium, methane, nitrogen and some other gases including residual amount of molecular hydrogen not fully separated in CF.

Category 2 includes impurities collected by the Liquid Nitrogen (LN_2) panels of the NIB and PD cryopumps during torus operation and subsequently released during the regeneration of the cryopumps to room temperature or higher. The regeneration of the LN_2 panels (which are a key factor in maintaining good vacuum conditioning) only occurred very infrequently. The elevated temperature of the regeneration allows release of water and higher hydrocarbons which can not be released from the cryopumps during their regeneration at 77K. The impurities are separated from the molecular hydrogen isotopes in CF prior transfer to IP. **Category 3** includes gas mixtures generated during torus and/or AGHS maintenance. There are also gas mixtures generated during Torus vacuum conditioning, in particular bakeout at elevated temperature (570K) during a restart after a shutdown. Amount, chemical and isotopic composition of such mixtures can vary in a large range.

The existing Impurity Processing system was designed to operate in batch mode and process up to 0.1MPa.m³ of gas in a single batch. Two different techniques can be used for impurity processing. The first technique is a catalytic conversion of hydrogen containing species into water form followed by water collection in a cold trap and then decomposition in reactions with metal at elevated temperature. The second technique is a direct decomposition of hydrogen containing species in reactions with uranium at elevated temperature. In both techniques molecular hydrogen liberated from the impurities is then absorbed in uranium operated at room temperature.

A description of the Impurity Processing system, amount and composition of the gas mixtures of different categories decontaminated during and after DTE1, and the results of IP performance are presented below.

2. GENERAL DESCRIPTION OF IMPURITY PROCESSING SYSTEM

A schematic diagram of the present configuration of IP is shown in Fig.1. The gas processing loop consists of a 2m³ reservoir (RS), a cold trap (CT) cooled by gas nitrogen with a temperature of 77K, seven chemical modules and two Normetex pumps (N) connected in series. The chemical modules include: a catalytic reactor (CR) filled with palladium catalyst; two containers (IB1, IB2) filled with iron powder; and four containers (UB1 - UB4) filled with uranium powder. The gas mixture to be decontaminated is admitted to the reservoir, analysed by the gas chromatograph and circulated through relevant chemical modules. For impurity conversion to water form, gas in the reservoir has to be mixed with oxygen prior to circulation through the catalytic reactor and the cold trap. Water collected in the cold trap has to be released and passed through iron and uranium containers operated at elevated temperature. Uranium is very reactive with water but the reaction leads to irreversible formation of uranium oxide. To reduce the amount of uranium consumed by water decomposition, it was foreseen to use iron for decomposition of the bulk of the water. Iron is less active than uranium towards a reaction with water but this reaction is reversible, ie iron oxide can be regenerated to metal form in reaction with hydrogen gas. Carbon dioxide, which is a product of the reactions of hydrocarbons' oxidation and is collected in the cold trap together with water vapour, can also react with iron and uranium at elevated temperature leading to their additional consumption. To reduce metal consumption in the reaction with CO₂, this carbon dioxide can be selectively released from the cold trap by its controlled warming up to temperature of 200K prior water processing.

An alternative operating mode of the impurity processing system is to decompose the hydrogen containing species in reactions with uranium at elevated temperature. Irreversible reactions occur liberating molecular hydrogen and forming uranium oxide, carbide, etc. The molecular hydrogen is then absorbed by uranium in a uranium container operated at room temperature. The disadvantage of this technique is that the uranium is consumed irreversibly.



Fig.1: Overview of the Impurity Processing system.

3. GAS MIXTURES DECONTAMINATED IN IP

This section gives the amounts and the compositions of gas mixtures of different categories decontaminated in IP. These numbers are based on the results of AGHS operation during and after JET deuterium-tritium experiment, DTE1 [5].

Category 1. The total amount and composition of the gas pumped from the PD and NIB cryopumps as a result of all the regenerations of their helium panels to 77K during DTE1 are shown in Tables 1 and 2.

Doromotor	Cryopump	
ratameter	NIB	PD
Amount of gas pumped (NL)	1682	226
Average tritium concentration in pumped molecular hydrogen isotopes (at.%)	12.3	29
Amount of impurities pumped (NL)	19.6	5.3
Average concentration of impurities (vol.%)	1.2	2.3

Table 1. Gas pumped from the PD and NIB cryopumps regenerated to 77K during DTE1 (second phase of the DTE1, September - November 1997).

Impurity	Concentration (vol.%)		
Не	67.67		
N ₂	10.53		
$O_2 + Ar$	0.92		
$CO + CO_2$	1.62		
CQ ₄ ^a)	16.40		
C ₂ Q ₂	0.15		
C ₂ Q ₄	1.21		
C_2Q_6	1.3		
C ₃ Q ₈	0.2		

Table 2. Average composition of impurities pumped from the PD and NIB cryopumps when regenerated to 77K during DTE1.

^{a)} Q represents T, D, H. Presence of higher hydrocarbons was detected by gas chromatographic analysis, but their composition and concentration were not determined.

Category 2. Total amount of gas released from the LN_2 panels of the NIB and PD cryopumps during their regeneration to ambient temperature after DTE1 was 56.5NL. The impurities collected on these panels had been pumped throughout the entire DTE1 experiment. The impurities included 18.0NL of water vapour released from NIB cryopumps and 5.1NL of water vapour released from PD cryopumps. The chemical composition of the gas is shown in Table 3.

Table 3. Composition (excluding water vapour) of the gas pumped from the LN_2 panels of the PD and NIB cryopumps regenerated to ambient temperature after DTE1.

Gas component	Concentration (vol.%)		
Q2	65.1 (0.9% T, 12.1% D, 87.0% H)		
Не	0.2		
N2	30.3		
$O_2 + Ar$	0.5		
$CO + CO_2$	2.5		
CQ ₄	1.1		
C ₂ Q ₂	0.12		
C ₂ Q ₄	trace		
C ₂ Q ₆	0.18		
C ₃ Q ₈	trace		
Other gases	Higher hydrocarbons ^{a)}		

^{a)} The composition and amount of higher hydrocarbons were not determined.

The isotopic composition of the water released to IP from NIB and PD cryopumps during the regeneration of their LN_2 panels to ambient temperature after DTE1 is given in Table 4.

Cryopump	Isotope concentration (at.%)		
	Т	D	Н
PD	11.6	53.7	34.7
NIB	4.0	19.7	76.3

Table 4. Isotopic composition of water released from NIB and PD cryopumps.

Category 3. Tritium containing gases generated during maintenance of the torus or tritium systems of AGHS are usually discharged directly to Exhaust Detritiation system. The main source of the highly tritiated gas which needs to be decontaminated in IP prior discharge to Exhaust Detritiation system is a gas transferred to IP during the vacuum vessel baking and pumping. The amount of such a gas could be up to several hundreds litres. Water vapour is the dominant component of the gas. For example, 337NL of water vapour with tritium isotopic concentration of 0.14at.% were collected in IP during the JET vacuum vessel baking at temperature of \approx 570K in September 1998. The amount of gases generated in maintenance of tritium systems of AGHS which needed to be decontaminated in IP was negligible in comparison to the amount of gas collected during vacuum vessel baking.

4. RESULTS OF OPERATION OF IMPURITY PROCESSING SYSTEM

4.1 Efficiency of impurities decontamination via tritium transfer to water form.

The efficiency of impurities oxidation into water form was found to be very high for the catalytic reactor operated at temperature of 770K and with 30% surplus oxygen in the gas mixture. Water generated in the catalytic reactor was collected in the cold trap. For a cold trap operating temperature of 160K, a residual water vapour pressure in the 2m³ reservoir as low as 5Pa was achievable but only with a long period of gas circulation.

The highest efficiency of water decomposition in two iron containers connected in series and operated at temperature of 770K was determined during inactive commissioning as 85%. The maximum achievable decontamination factor of iron containers, ie ratio of amount of tritium in water decomposed in the iron containers to that in water generated during the iron oxide regeneration, was found to be less than 200. The efficiency of water decomposition upon expansion from the cold trap to a single uranium container operated at a temperature of 770K was $\approx 100\%$. Because of the above mentioned result of the tests of the iron and uranium containers it was decided to use only uranium containers for water decomposition during DTE1. The maximum achievable factor of gas decontamination in IP via impurities oxidation to water form was limited by residual tritium contamination of the surface of the reservoir and pipes in the gas circulation loop. This residual tritium contamination led to a tritium concentration of ≈ 100 GBq/m³ in the nominally tritium-free helium after its circulation through the IP loop.

4.2 Efficiency of impurities decontamination via their reaction with uranium at elevated temperature.

The efficiency of gas decontamination via reactions of hydrogen-containing gases with uranium at elevated temperature was evaluated using methane, which is one of the most difficult gases to decompose, as a reference gas. The reaction of methane decomposition is reversible, ie

$$x CQ_4 + U \Leftrightarrow UC_x + 2xQ_2 \tag{1}$$

To shift the thermodynamic equilibrium of the reaction (1) in the direction of methane decomposition, liberated molecular hydrogen must be removed from the gas mixture. Decontamination of gas mixtures from methane and other impurities occured in a uranium container operated at a maximum permissible temperature of 800K. Liberated molecular hydrogen was then absorbed by uranium in a second uranium container operated at room temperature. The maximum methane decontamination factor achieved was \approx 300 and required a long period of gas circulation.

4.3 Uranium consumption for impurities processing.

The amount of uranium consumed in IP uranium containers used for impurities processing is illustrated in Table 5 which shows the capacity of the uranium container for hydrogen sorption, before and after DTE1.

	Capacity (mol. H ₂)			
Container	Original	Remaining after tritium commissioning ^{a)}	Remaining after DTE1	Used
UB1	26.0	20.4	4.4	21.6
UB2	27.0	24.7	13.9	13.1
UB3	26.6	26.6	20.9	5.7
UB4 ^{b)}	26.3	26.3	26.1	0.2
Total	105.9	98	65.3	40.6

Table 5. Capacity for hydrogen sorption by uranium containers in IP.

^{a)} 8.6 moles of water were decomposed using UB1 and UB2 during IP tritium commissioning.

^{b)} Container UB4 has been used during impurities processing for hydrogen absorption only.

Table 5 shows that the use of uranium for the decomposition of 8.6 moles of water during IP tritium commissioning [6] caused loss of 7.9 moles H_2 of the hydrogen sorption capacity of the IP uranium containers. This value is much lower than the observed decrease of uranium

capacity, 32.7 moles of H_2 , caused by use of uranium containers for impurities, which included ≈ 1 mole of water, processing during DTE1. This large loss of hydrogen sorption capacity can be only explained by large uranium consumption in runs for gases decontamination performed using reactions of impurities with uranium at elevated temperature.

5. FUTURE DEVELOPMENT

To avoid irreversible uranium consumption for impurities processing in future operation of AGHS, modification of the existing IP system is under way. The modification consists of the replacement of two existing iron containers with two new modules: a catalytic reactor filled with nickel catalyst, and a palladium permeator. The catalytic reactor will be used for decomposition of hydrocarbons and water decomposition in reactions catalysed by a nickel catalyst:

$$CQ_4 = C + 2Q_2 \tag{2}$$

$$2Q_2O + C = CO_2 + 2Q_2 \tag{3}$$

Molecular hydrogen liberated in the reactions 2 and 3 will be removed from the gas mixture due to selective hydrogen penetration through a palladium membrane in the palladium permeator. The use of reaction 2 avoids the conversion of tritium to water form.

Additionally, to minimise tritium release from IP to the Exhaust Detritiation system, it is foreseen to discharge the gas from IP to ED via a PERMCAT reactor developed in Tritium Laboratory Karlsruhe [7].

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