
EFDA–JET–CP(04)06/14

L. Di Pace , B. Patel and JET EFDA Contributors

Factors Affecting the Inhalation Dose from Tritiated Dust and Flakes

Factors Affecting the Inhalation Dose from Tritiated Dust and Flakes

L. Di Pace , B. Patel and JET EFDA Contributors*

¹*Associazione Euratom-ENEA sulla Fusione, Via Enrico Fermi 45, 00044 Frascati, ITALY*
²*EURATOM/UKAEA Fusion Association, Culham Science Centre, Abingdon Oxon OX14 3DB, UK*
* See annex of J. Pamela et al, "Overview of Recent JET Results and Future Perspectives",
Fusion Energy 2002 (Proc. 19th IAEA Fusion Energy Conference, Lyon (2002)).

Preprint of Paper to be submitted for publication in Proceedings of the
23rd SOFT Conference,
(Venice, Italy 20-24 September 2004)

“This document is intended for publication in the open literature. It is made available on the understanding that it may not be further circulated and extracts or references may not be published prior to publication of the original when applicable, or without the consent of the Publications Officer, EFDA, Culham Science Centre, Abingdon, Oxon, OX14 3DB, UK.”

“Enquiries about Copyright and reproduction should be addressed to the Publications Officer, EFDA, Culham Science Centre, Abingdon, Oxon, OX14 3DB, UK.”

ABSTRACT

Tritiated dust and flakes were produced in JET during D-T campaigns. Measurements showed some atypical radiological effects; a very large T concentration and off-gas rate. A higher dose per unit intake was foreseen and it is a consequence of the particulate nature of the material and of the expected longer biological retention.

Particle size plays an important role in the deposition pattern in the human respiratory tract. Larger particles are mostly deposited in the anterior nasal passage and remaining airways of the head and neck and removed in less than one day. Smaller particles are mostly retained in the bronchial and bronchiolar regions and alveoli and cleared in longer periods (up to one year). The only clearing mechanism of tritium in the latter case is its absorption into lung fluid, an important phenomenon which can be reproduced by in-vitro tests to determine doses and dose conversion factors. Based on a review of initial in-vitro tests performed on JET dust and from a literature study related to in-vitro dissolution experiments, some recommendations for next in-vitro experiments on JET dust were formulated.

1. INTRODUCTION

The work presented in this paper has dealt with the literature review of radiation hazards due to inhalation of tritiated dust and flakes of JET and other tokamaks. It has been performed in the frame of the EFDA JET Fusion Technology Task JW3-FT-5.12 [1].

2. TRITIATED DUST AND FLAKES IN TOKAMAKS

The generation of dust and of loosely adhered deposits in experimental tokamaks is due to the erosion of C/Be First Wall (FW) by plasma/wall interaction, disruptions and evaporation, combined with deposition on the FW through diffusion, surface saturation and co-deposition with carbon [2]. Co-deposition is the dominant process for tritium uptake by carbon Plasma Facing Components (PFCs). Unlike implantation, which is relevant for all metals, co-deposition has only been found significant for carbon thus far.

Significant quantities of tritiated dust and flakes were produced in JET during D-T campaigns. A quantity of 6.2 g-T was estimated to be still inside the JET vacuum vessel at the end of D-T campaigns. After recovery, that amount has been reduced to about 1.5 g-T [3]. The radiological concern of JET tritiated dust is related to the measured levels of tritium content (up to ~ 1.3TBq/g-dust or 3.5mg-T/g-dust). It has been estimated, by using the dose conversion factor defined for HTO, that 1mg-dust with a tritium specific activity of 1TBq/g might cause a dose up to 20mSv, if no personnel protective suits are worn. The study of factors affecting the inhalation dose of tritiated particles is thus important in order to use the right dose conversion factors both for worker and public and to develop guidance on monitoring for routine or accidental intakes. Due to the different nature and the different residence time of tritiated particles in the human body, much larger doses due to their intake may be foreseen than for a similar HTO intake. In addition to particulate dusts, flakes have

been found in JET [4]. Flaking is a mechanism observed on carbon tiles, leading to a progressive detachment of particles with size greater than $\sim 100 \mu\text{m}$ [5].

3. TRITIATED DUST AND FLAKES PHYSICAL PROPERTIES

Particle size plays an important role in the deposition pattern of particles in the human respiratory tract and it is required to assess the self-absorption factor for beta emission. The particle size distribution of a dust particle population is wide, but in most cases it can be described through a log-normal distribution with a distribution density function. The most significant physical parameters, used to characterize the particle size distribution, are:

Surface area mean diameter (D_{MVS}); it is useful in relating dust specific surface area, defined just further on, to the geometric size distribution:

$$D_{MVS} = \frac{\sum_i n_i \cdot d_i^3}{\sum_i n_i \cdot d_i^2} \quad (1)$$

Specific surface area (S); it is given by the ratio of the total external surface of all particles divided by the sum of their mass:

$$S_{sp} = \frac{6}{\rho \cdot d}; \quad S_{sp} = \frac{6}{\rho \cdot D_{MVS}}; \quad S_{sp} = \frac{k}{\rho \cdot D_{MVS}}; \quad (2)$$

a) b) c)

a) $k=6$ and $D_{MVS} = d$ for mono-disperse spherical powder; b) $k=6$ for poly-disperse spherical powder; c) poly-disperse powder with irregular shape, [k is the surface area shape factor; $k=6$ for a sphere].

Projected area diameter (d_{pa}); it is the diameter of an irregular particle calculated by measuring the projected area corresponding to a circle having an equivalent area.

Volume equivalent diameter (d_{ve}); it is the diameter of a sphere with the same volume of the particle dust.

Aerodynamic equivalent diameter (d_{ae}); it is the diameter of a unit density sphere having the same settling velocity of non-spherical particle.

Activity median diameter (AMD); it is defined such as the half of the aerosol activity is associated with particles having $D < AMD$. [D can be d_{pa} , d_{ve} and d_{ae}].

Activity median aerodynamic diameter (AMAD); it is defined such as 50% of the aerosol activity is associated with particles with aerodynamic equivalent diameter $< AMAD$. It determines the dust mobility and describes the extent for deposition in the respiratory tract and for inhalation intake.

Self absorption factor; it is a size dependent characteristic having influence on the dust radiological properties. Because tritium beta emission is low, there is absorption of beta particles within the dust. SAF_{β} is the fraction of beta rays escaping their absorber, SAF_e is the fraction of beta energy escaping

the particle. The specific surface area can be directly measured through gas adsorption method or indirectly by equation 2 if the inner porosity of the particle is negligible. Some graphitic dusts, like those collected in JET, TFTR, Alcator C-mod and DIII-D, showed measured specific surface areas greater than that predicted by equation 2. That is due mainly to the multi-faceted nature of graphite, and, to a lesser extent, to the suspected surface connected porosity of graphitic dusts. This issue is important as far as the tritium retention/dissolution is concerned and it may play a certain role in the in-vitro tritium dissolution tests.

4. TRITIATED DUST AND FLAKES RADIOLOGICAL PROPERTIES

Among the physical properties, the most important ones from the radiological point of view, are the aerodynamic equivalent diameter d_{ae} and the activity median aerodynamic diameter (AMAD). Measurements performed during the JET 1999 shutdown showed that the value of d_{ae} for airborne dust was $\sim 4 \mu\text{m}$.

It is known from different studies that particle deposition in the different parts of the human respiratory system depends on the aerodynamic equivalent diameter. Material deposited, mostly with $d_{ae} \geq 10 \mu\text{m}$, in the anterior nasal passage is removed in ~ 1 day. The remaining airways of the head and of the neck are covered by a fluid lining, which is cleared in few minutes to the pharynx and swallowed. Much of the material deposited in the bronchial (BB) and bronchiolar (bb) regions is cleared rapidly (few hours) by mucus, while smaller particles than a few μm can stay longer. In the Alveolar Interstitial (AI) region there is about 80% retention at 50 days and 50% at 1 year. Because small dust particles might be retained there for years, the dissolution of tritium adsorbed into the dust particle is an important factor in the assessment of dose and dose conversion factors. According to the new ICRP Model for the Human Respiratory Tract Model (HRTM), the clearance of a material from an organ or a tissue may take place either by particle transport, which moves the material of our interest to the Gastro Intestinal (GI) and lymph nodes, or by absorption into blood which is a two-stage process: dissolution plus uptake. Figure 1 illustrates the fractional deposition of dust vs. AMAD, according to predictions done by the new ICRP Model for the HRTM [6]. The dissolution can be reproduced by in-vitro tests and it is of primary importance to describe phenomena related to dissolution and uptake of tritium into lung serum, being the most important way of tritium clearance. On the other hand, in-vivo experiments can give additional information to complete the global picture related to biological effects from inhalation of tritiated particles. Some in-vivo experiments carried out by intra-tracheal instillation of metal tritides (Zr, Ti) in rats have shown slow clearance rates from the rat lungs. The radiation dose was estimated to be an order of magnitude larger than those calculated for HTO [7].

5. RESULTS OF JET DUST IN-VITRO DISSOLUTION TESTS

In-vitro dissolution tests were conducted by NRPB to better define properties of JET tritiated C/Be dusts [8]. ‘Coarse’ and ‘fine’ dust collected during the 1998 shut-down of JET were subjected to T

dissolution tests in lung serum simulant. The results of these experiments were given as a fraction of the initial particle tritium activity passed into the serum simulant. For each sampling time, the fractional tritium remained in the particle assay was measured. For ease of interpretation and in order to find agreement with the ICRP model, the tritium clearance rate was divided in a 2-stage process: fast dissolution rate of a limited fraction (up to < 10%), very slow one of the rest. The T fractional retention in JET carbon particles is shown in Figure 2. The equation:

$$R(t) = f_r \cdot \exp(-s_r \cdot t) + (1 - f_r) \cdot \exp(-s_s \cdot t) \quad (3)$$

where the parameters f_r , s_r and s_s , which are the dust fraction with fast dissolution rate (f_r), the fast dissolution rate (s_r) [s^{-1}] and the slow dissolution rate (s_s) [s^{-1}], are used to fit experimental data and to get parameters describing the ICRP HRTM. In this model, the gut uptake factor (f_1) describes the fraction of an inhaled element directly absorbed in the body fluids. For HTO $f_1 = 1$, for tritiated dust f_1 is much lower than 1; it was estimated from the analysis of in-vitro experiments a value of 0.1 for ‘coarse’ particles and a value of 0.01 for ‘fine’ particles. The results of JET dust in-vitro tests were compared with other ones carried out on TFTR carbon particles [9], or on titanium tritide [10].

The major conclusion from this comparison is that NRPB experiments have not been conclusive and demonstrated some anomalies when compared to other available data, particularly as particle size was not quantified. Another limitation of the study was the short duration of these experiments (2 weeks), in comparison to the typical time scale of phenomena under investigation (tens of days). TFTR dust and titanium tritide in-vitro dissolution experiments lasted 110 and 30 days respectively. Hence, before performing any in-vivo experiments, required to get material-specific absorption rates and to set biokinetic models of tritium dust inhalation, it would be important to repeat the in-vitro tritium dissolution experiments on JET dust.

6. RECOMMENDATIONS FOR NEW IN-VITRO EXPERIMENTS

Some guidance for the next in-vitro experiments on JET dust can be summarised as it follows.

- 1) To define the dust size class more rigorously providing the count mean diameter value, the standard deviation and the related aerodynamic equivalent diameter.
- 2) To determine the specific surface diameter.
- 3) To perform some beta radiation measurements on dust samples to be compared to those obtained by computation introducing the value of the particles average beta activity, to have an idea of the shift from homogeneity of the actual T distribution in the particle.
- 4) To conduct dissolution trials over more than 14 days. A reasonable time might be 3 months.
- 5) To determine the T dissolution behaviour from dust particles from the very first start of experiments. As the fast dissolution component calculated during the JET dust tests had an half time of ~2 minutes for both ‘fine’ and ‘coarse’ particles, it would be undesirable to get the first measurement 5 minutes after the test start.
- 6) To determine the chemical composition of dust in addition to C and H.

The dust to be sampled for the next in-vitro tests should include two different size classes. The first class should have a value of the count mean diameter (CMD) $\leq 1 \mu\text{m}$, that is in the range of the reference diameter of the ITER dust particles ($0.5 \mu\text{m}$), the second class with CMD around $5 \mu\text{m}$. The complete understanding of the behaviour of inhaled tritiated particles in the human body and their related effects cannot rely on in-vitro experiments only, as other physico-chemical effects should allow for, e.g. the macrophage action occurring in the lung which can be material specific. Complex biokinetics models have been set up to reproduce phenomena related to inhalation of tritide particles in a living being, the parameters of which can be obtained only by in-vivo experiments. These phenomena include (Cheng et al. 1999 [11]):

- particle transport from lung to gastro-intestinal (GI) tract,
- particle transport from lung to bronchial lymph nodes,
- transport of tritium from GI tract to circulatory system and vice versa,
- transport of T from circulatory system to lung air,
- transport of T from lung air to exhaled air,
- transport of T from circulatory system to urine,
- transport of T through the GI tract into faeces.

It is important to outline that, from the radiological point of view, it is of primary importance to describe phenomena related to dissolution and uptake of tritium into lung serum, as this is the most important way of clearance from small dust particles retained in the lungs.

For this purpose, in-vitro experiments can provide important information, while the in-vivo experiments supply additional information to complete the insight related to biological effects from inhalation of tritiated particles.

ACKNOWLEDGMENT

This work, supported by the European Communities under the contracts of Association between EURATOM/ENEA and EURATOM/UKAEA, was carried out within the framework of the European Fusion Development Agreement. The views and opinion expressed herein do not necessarily reflect those of the European Commission.

REFERENCES

- [1]. L. Di Pace, Literature study on radiological hazards of tritiated dust and flakes, ENEA Final Report Task JW3-FT-5.12, FUS-TN-SA-SE-R-86 Rev. 1, December 2003
- [2]. S. J. Piet and G. Federici, ITER White Paper on Integrated Picture of In-Vessel Tritium and Dust, ITER, IDoMS S 81 RI 13 96-06-28 W 1.4, July 1996
- [3]. R. Lässer et al. JET Fusion Technology R&D for Wastes from Fusion Facilities, Workshop W53: Experience in the Management of Wastes from Fusion Facilities, Culham, UK, 25-26 March 2003

- [4]. B. Patel, S. Knipe, P. Macheta, and A. Peacock, Radiological Properties of Tritiated Dusts and Flakes from the JET Tokamak, 18th IEEE/NPSS Symposium on Fusion Engineering, Albuquerque NM USA, October 1999
- [5]. C. H. Skinner, C. A. Gentile, M. M. Menon, R. E. Berry, Flaking of co-deposited hydrogenated carbon layers in the TFTR limiter, Nucl. Fusion **39** (1999) 1081-1085
- [6]. International Commission on Radiological Protection (ICRP), Human Respiratory Tract Model for Radiological Protection, Publication 66, Ann. ICRP. 24(1/3), Oxford Elsevier Science, 1994
- [7]. R. F. Kropf, Y. Wang, and Y. S. Cheng, Self-Absorption of Tritium Betas in Metal Tritide Particles, Health Phys., **75** (1998) 398-404
- [8]. A. Hodgson, E. R. Rance, P. G. D. Pellow and G. N. Stradling, In vitro Dissolution of Tritium Loaded Carbon Particles from the JET Tokamak, NRPB Report, National Radiological Protection Board, Chilton, Didcot, NRPB-DA/1/2002, March 2002
- [9]. Y. S. Cheng, Y. Zhou, C. A. Gentile and C. H. Skinner., Characterization of Carbon Tritide Particles in a Tokamak Fusion Reactor, Fusion Sci. Technol. **41** (2002) 867-871
- [10]. Y. S. Cheng, A. R. Dahl and H. Nian Jow, Dissolution of Metal Tritides in a Simulated Lung Fluid, Health Phys. **73** (1997) 633-638
- [11]. Y. S. Cheng, M. Burton Snipes, Y. Wang and H. Nian Jow, Biokinetics and Dosimetry of Titanium Tritide Particles in the Lung, Health Phys. **76** (1999) 120-128

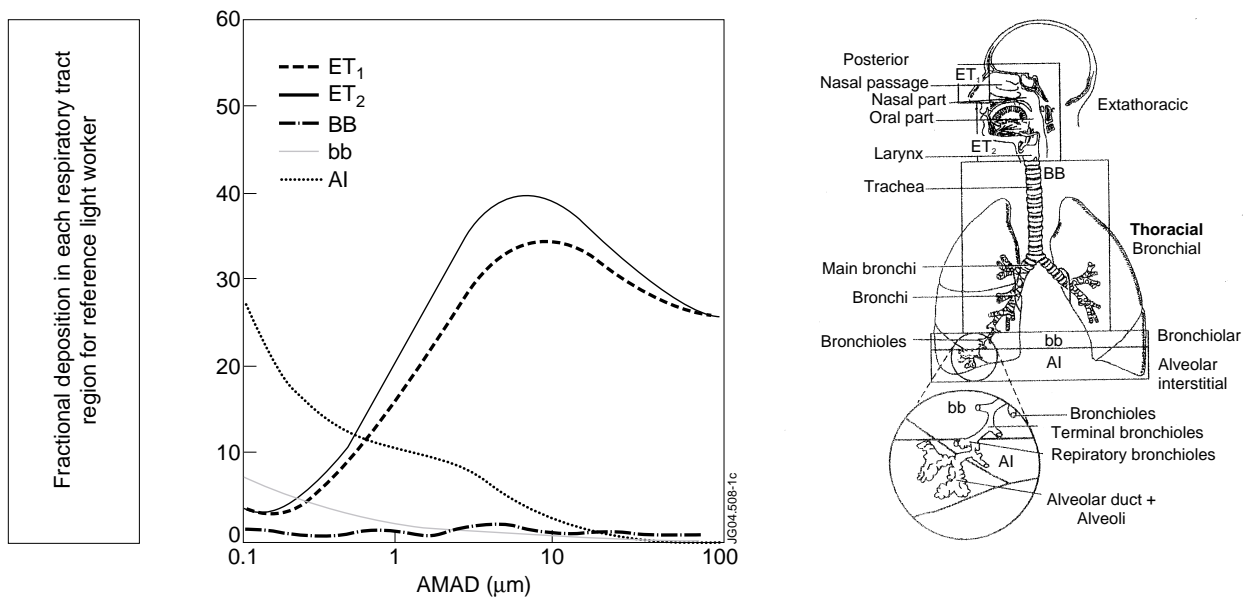


Figure 1 - Left: fractional deposition of particles in the different respiratory tract region for a reference light worker. Right: the respiratory tract model defined in the new ICRP model for the Human Respiratory Tract.

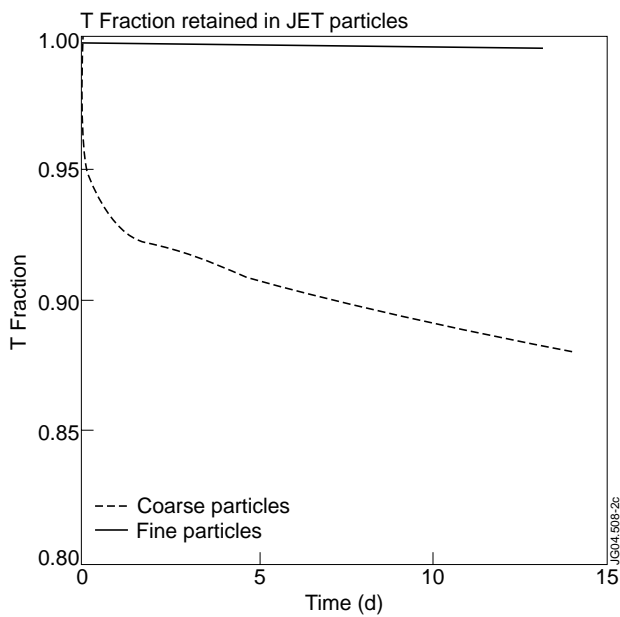


Figure 2 - Fractional retention of tritium in JET carbon particles, NRPB in-vitro tests.