ELSEVIER

Journal of Nuclear Materials 212–215 (1994) 1003–1009

journal of nuclear materials

An assessment of models for tritium release from ceramic breeders for blanket analysis applications

G. Federici [a], A.R. Raffray [b], M.C. Billone [c], C.H. Wu [a], S. Cho [b], M.A. Abdou [b]

[a] *The NET Team, Max-Planck-Institut für Plasmaphysik, Boltzmannstrasse 2, 85748 Garching bei München, Germany*

[b] *Mechanical, Aerospace, and Nuclear Engineering Department, University of California, Los Angeles, Los Angeles, CA 90024-1597, USA*

[c] *Energy Technology Division, Argonne National Laboratory, Argonne, IL 60439-4838, USA*

Abstract

Several models have been proposed and used to analyse the tritium release from ceramic breeder blankets. The transient models range from single-mechanism types of models to comprehensive, fully integrated models over a blanket spatial location, including all the key transport mechanisms, as well as local spatial temperature gradients.

This paper considers different models available for analysis of tritium transport in ceramic breeders and performs a critical assessment of their applicability for blanket analysis and of their limits. Based on the analysis, the type of model which would be best suited for blanket analysis is described. It combines the possibility of including to a reasonable extent the effect of key transport mechanisms with the ease of integrating the computations over the whole blanket space, and provides the capability for both steady state and transient analyses.

1. Introduction

An important aspect of the design and analysis of ceramic fusion reactor blankets is the ability to predict the phenomenological behaviour of tritium in the different blanket materials (e.g., lithium-based solid breeder and beryllium multiplier) under operating reactor conditions. By understanding the behaviour of tritium in such materials, analysis and accurate predictions can be made regarding the blanket tritium release and inventory which are key design issues based on safety and economic considerations. This paper focuses on modelling tritium behaviour in lithium based ceramic materials. The interested reader is directed to the Refs. [1,2] for latest progress on modelling tritium transport in beryllium.

As is evident by the abundant literature existing on this subject (see for example Refs. [3–8]), during the past 10 years, substantial theoretical and experimental efforts have been dedicated world-wide to improve the understanding, to identify the underlying transport mechanisms, and to develop verified and validated methodologies and models for predicting tritium transport. In general, models that are available today seem to cover an adequate selection of physical mechanisms and numerical methods. They have allowed for reasonable interpretation and reproduction of experimental data and have helped in pointing out deficiencies in material property data base and in providing guidance for future experiments.

This paper briefly describes some of the existing models and critically reviews the transport mechanisms included in these models in light of recent experimental data analysis. Desired characteristics of a tritium blanket design code (TBDC) are discussed including the need to account for the blanket geometry details, temperature, tritium generation and purge concentration profiles and histories. Shortcomings of existing models based on these characteristics are pointed out and an example of an attractive TBDC is described

2. Tritium transport characteristics and models

The mechanisms for tritium transport in ceramic breeders have been quite well characterised.

0022-3115/94/$07.00
SSDI 0022-3115(93)E0303-Q

A number of models of varying degree of complexity are available to analyse the tritium release. For transient analyses, models range from those based on simple diffusion and/or desorption control [9–11] to the comprehensive MISTRAL code [12–15] which includes all key transport mechanisms.

Clearly, the use of simple models for which analytical solutions are available over a range of conditions, is very appealing. At the other end of the spectrum, comprehensive codes provide a complete simulation of the tritium processes at one location in the blanket but would have to be run over a number of blanket locations to account for spatial variations in generation rate, temperature and purge flow conditions throughout the blanket. The computing requirements then become substantial making their use cumbersome and complex for blanket parametric design calculations.

The concern with the simple models is that by assuming only one or two major transport mechanisms and, thereby, neglecting the others, they could give erroneous results as conditions change in the blanket. In addition, these simple models would have to be run over multiple blanket locations to account for 2-D temperature and tritium generation profiles. Otherwise, results based on average values might be inaccurate. For example, for the layered blanket configuration of the ITER CDA design [16], the ceramic breeder is sandwiched between two Be layers. The resulting radial temperature across the ceramic breeder at a given poloidal location was about 453°C at the outer surfaces, and following a characteristic parabolic profile, peaked at the centreline at about 537°C. Conversely, the tritium generation rate had a characteristic U-profile, with a maximum at the outer surfaces of about 1.5×10^5 appm/s and dropped sharply across the breeder region to a level of about 0.35×10^5 appm/s. For this case, the steady state inventory was estimated based on the diffusion control and diffusion/desorption control models for typical $LiAlO_2$ diffusion and desorption coefficients [15] equal to $D(m^2/s) = 8.82 \times 10^{-8} \exp(-144.5 \text{ kJ/mol}/RT)$, $K_d(m/s) = 7.7 \times 10^{-4} \exp(-83.7 \text{ kJ/mol}/RT)$, respectively.

In both cases, the inventory at the ceramic breeder region surface, where the generation rate is the highest and the temperature is the lowest, is about 50 times higher than that at the mid-region where the generation rate is lowest and the temperature highest. Thus, to properly account for spatial variation in the inventory and release, the models would have to be run over several radial and poloidal locations and the results integrated.

3. Key transport processes based on recent experimental data analysis

Over the last few years, several analyses have been performed to help interpret and understand tritium release experiments. Some of these were quite illuminating in showing the importance of specific mechanisms on the tritium release behaviour of ceramic breeders under certain conditions. Some of the information emerging from these analyses is summarised in

Table 1
Importance of tritium transport mechanisms in reproducing experimental data

Experiment: Material:	BEATRIX II (Li_2O)	Single crystal ($LiAlO_2$)	TEQUILA ($LiAlO_2$)	MOZART ($LiAlO_2$)	LISA (Li_2SiO_3)
Major transport mechanisms required for data reproduction					
Bulk diffusion		×	×	×	×
Surface processes					
Bulk-to-surface	×	×			
Dissolution	×	×			
Desorption	×	×	×	×	×
Adsorption	×	×	×	×	×
Bulk trapping (chemical or irradiation-induced)	×				
Pore diffusion			×	×	×

Table 1, in terms of the key tritium transport mechanisms that need to be considered in order to reproduce the experimental tritium release and/or inventory.

Analysis of the tritium behaviour in the Li_2O samples of the BEATRIX-II experiment indicated the need to include bulk trapping processes, hypothesised to be chemical trapping due to LiOT formation [15]. Inventory calculations assuming no chemical trapping were substantially lower than experimental values (by a factor of ~ 50). Modelling results including bulk trapping reproduced reasonably well experimental results for the end-of-life inventory and tritium release under temperature transients. For Li_2O in particular, surface-driven bulk inventory (solubility) can be appreciable depending on the partial pressure of the tritium species in the pores (following a Sievert's type relation).

Analyses of tritium release from $LiAlO_2$ single crystal [17] and from the TEQUILA experiment [15] indicated the importance of bulk diffusion in combination with surface processes in modelling $LiAlO_2$ tritium behaviour.

Analysis of the LISA (Li_2SiO_3) and MOZART ($LiAlO_2$) experiments showed the need to properly account for the possible slow-down of pore diffusion due to non-uniform pore size distribution [13].

4. Desired characteristics of a tritium blanket design code

The development of a TBDC is important for design analysis of both blanket and associated tritium processing units. Such a code will also represent an important part of a total tritium system model which will be needed to evaluate the movement of tritium and the resulting inventories in the different parts of the fusion reactor system. The desired outputs of a TBDC include the tritium concentration distributions in the different materials composing the blanket for any given reactor power history, as well as the tritium concentration in the purge outlet, i.e., the tritium release.

The level of confidence of most of the blanket tritium inventory assessments performed in the past (see, for example, Refs. [11,18–20]) depends strongly on the adequacy of the type of models mentioned in section 2 to simulate the real blanket design conditions.

It is clear that, for improved and more accurate tritium blanket analyses, current mechanistic models need to be extended and integrated into a TBDC to account for geometric details of the blanket layout as well as breeder microstructure characteristics (e.g., pore/grain sizes and distributions, and specific surface area) and to include temperature, tritium generation, purge flow paths, and purge concentration profiles and histories. Ideally, a TBDC should be integrated in a calculation methodology which incorporates neutronics, thermal, and purge thermal-hydraulic calculations.

However, a balance must be maintained between the desire for a fully integrated detailed computer modelling using sophisticated methodology and the practicality of using such a code for parametric studies. Simple one-mechanism models are easy to use but could easily overlook effects associated with excluded mechanisms which can be important in time and space as conditions change. Fully comprehensive models provide the right tool for a thorough analysis of tritium transport in the blanket. However, the computational requirements tend to limit these type of models to the analysis of tritium release at specific locations in the blanket and for providing benchmark blanket cases to test and calibrate a simpler model which would be easier to use and to integrate as part of a TBDC. Codes which use correlations based on experimental set of data or on parametric versions of more complicated models rather than the fundamental model themselves, could be attractive for applications that require reduced detail and accuracy for fast running times. For example, phenomena such as breeder densification and other effects induced by neutron irradiation could be accounted for by temperature and burn-up-dependent empirical correlations. However, verification of whether or not a selected correlation has the capability to simulate a particular transient of interest in the blanket remains critical.

5. Example of proposed tritium blanket design code

A schematic of a proposed TBDC is illustrated in Fig. 1 based on the solution of transport equations along the purge flow path. In its conception it allows for steady-state and transient capabilities. Most of the analytical formulation has been previously published (see in particular Refs. [12–15]) and will not be repeated here.

The model is used for the analysis of different sections of the blanket breeding elements. The blanket (inboard and outboard) is divided into a certain number of zones radially and poloidally and/or toroidally depending on the purge stream layout and based on steepest tritium generation and temperature gradients typically in radial and poloidal directions. The local average breeder temperature, calculated for each time step at each blanket contour node, is assumed as input in the control volume which defines the microstructural model unit cell surrounding each node, together with the average tritium generation rate and the composition of the purge at the previous adjacent node. The model allows for 1 or $1\frac{1}{2}$ D analysis at each blanket node. The latter includes the solution of the

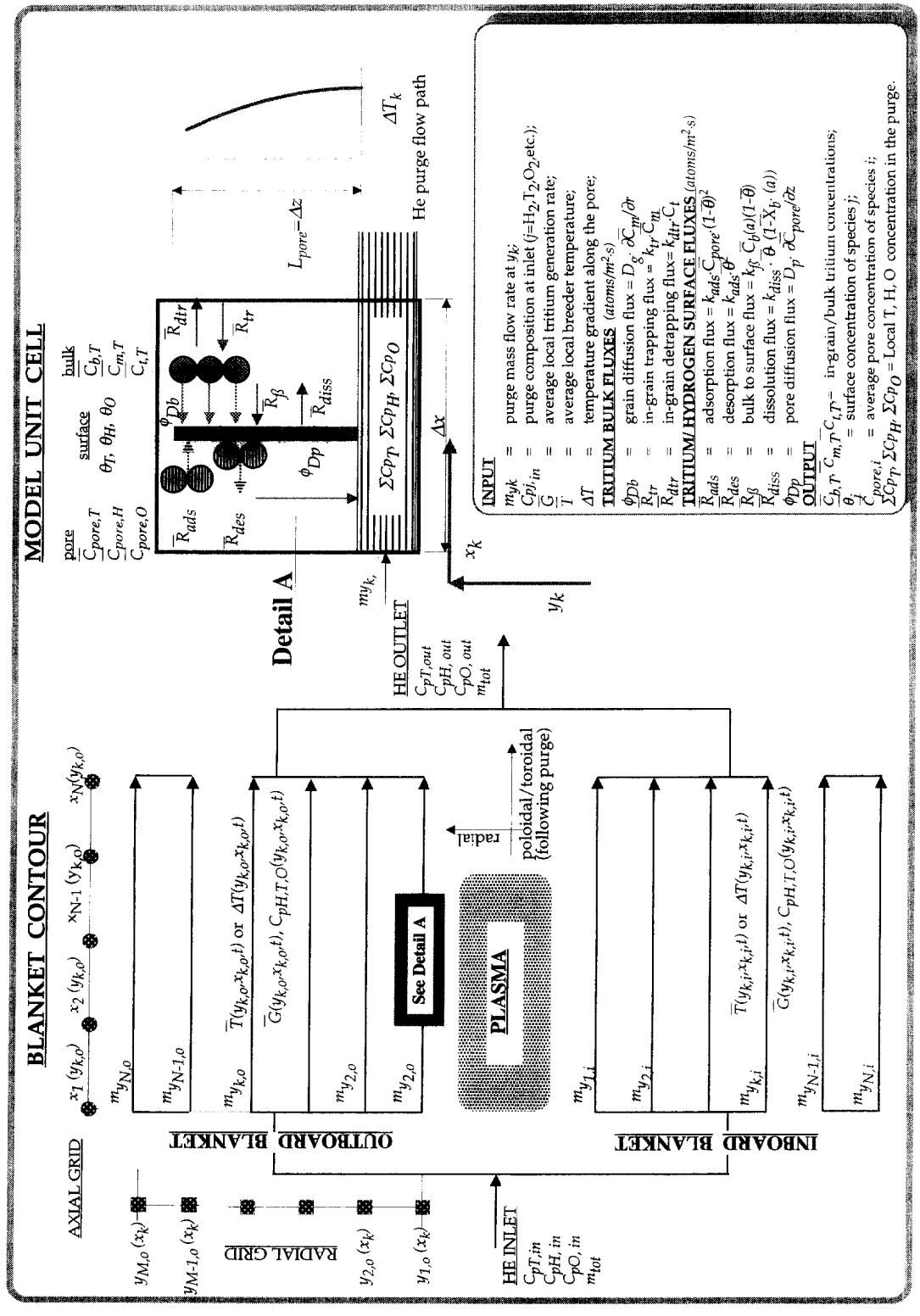

Fig. 1. Conceptual schematic of a tritium blanket design code (TBDC).

diffusion equation along the pore channel length, Δz, assumed perpendicular to the increment Δx along the purge flow path, which is required whenever pore diffusion is rate-controlling or when a steep temperature gradient exists along the pore network.

In order to avoid lengthy iterative schemes to advance the solution of the system of coupled simultaneous finite-difference time-stepping and rate equations at each blanket grid point, the integration procedure has to be consistently decoupled for each model zone by defining adequate interfacial assumptions for the different regions of the model. In particular, care must be taken to identify those release transport mechanisms, if any, which, according to the material and operating conditions being analysed, could be very fast (i.e., not rate-controlling) and, therefore, would not significantly affect the overall tritium release and inventory. These mechanisms could then be neglected to simplify the analysis.

Therefore, before effectively starting the calculation, a check would be done to estimate the inventory, at least for a number of key nodes of the blanket contour assuming possible worse-case input combinations (from the inventory buildup standpoint) which could occur at certain times (e.g., during burn or dwell) during the transient being analysed.

By comparing the inventory components it is possible to determine the transport mechanisms which could be neglected without affecting significantly the accuracy of the overall analysis.

To illustrate the modelling approach, a few examples of equations to be solved in the TBDC are described below. The formalism which will be adopted in the equations is summarised in Fig. 1 and refers to the case of averaged input/output values (e.g., temperature, tritium generation, concentration) along the increment Δz. The set of equations which follows needs to be solved at each nodal point x_k, y_k along the blanket contour and the proposed solution procedure consists of starting from the inlet purge composition and flow rate and starts at this node to solve for the grain/bulk first, followed by the surface regions, the network of interconnected pores and the purge flow path.

The concentration of tritium in the grains, $\bar{C}_{b,T}$ (both trapped $\bar{C}_{tr,T}$ and mobile $\bar{C}_{m,T}$ fractions) is governed by an equation of the type

$$\frac{\partial \bar{C}_{m,T}(r,t)}{\partial t} = D_b \nabla^2 \bar{C}_{m,T}(r,t) + \bar{G}(r,t) - \frac{d\bar{C}_{tr,T}(t)}{dt}. \quad (1)$$

The three terms on the r.h.s. of Eq. (1) account for diffusion of the mobile fraction of tritium, for generation by breeding reactions, and for some sort of trapping (e.g., chemical trapping), respectively. In Eq. (1), the total concentration of tritium can be expressed as the sum of the mobile and trapped tritium concentration in the bulk. The last term in Eq. (1) can be expressed by the following rate equation:

$$\frac{d\bar{C}_{t,T}(t)}{dt} = \bar{R}^N_{tr,j}(t) - \bar{R}^N_{dtr,j}(t). \quad (2)$$

Coupling of the bulk with the surface of the pores is given by

$$A^N_1 D_b \left[\frac{\partial C_{b,j}(r,t)}{\partial r}\right]_{r=a} = \bar{R}^N_{\beta,T}(t) - \bar{R}^N_{diss,j}(t), \quad (3)$$

where A^N_1 is a normalisation area.

For analysis of conditions where in-grain diffusion is not rate-controlling, Eq. (1) can be simplified and the following rate equation could be solved instead:

$$\frac{d\langle \bar{C}_{b,T}(t)\rangle}{dt} = \bar{G}^N_T(t) - \frac{d\bar{C}_{t,T}(t)}{dt} - R^N_{\beta,T}(t) + R^N_{diss,T}(t). \quad (4)$$

In Eq. (4) the last two terms represent the tritium flux from the bulk to the surface and the dissolution flux from the surface to the bulk. The symbol N implies that a normalisation factor has to be included for reason of consistency. For conditions where diffusion in the grains is rate controlling (i.e., both desorption from the surfaces and diffusion through pores are very fast), Eq. (1) can be solved decoupling bulk and pore surfaces, by setting for example the concentration at the grain surface equal to zero.

The buildup of coverage of species j at the pore surfaces, $\bar{\theta}_j$, is governed by an equation of the type

$$n_s \frac{d\bar{\theta}_j(t)}{dt} = \chi \bar{R}^N_{\beta,T}(t) - \bar{R}^N_{diss,j}(t) + \bar{R}^N_{ads,j}(t) - \bar{R}^N_{des,j}(t), \quad (5)$$

where n_s represents the density of surface sites and the terms appearing on the r.h.s. of Eq. (5) are indicated in Fig. 1 (note that $\chi = 1$ per $j = T$ and $\chi = 0$ per $j \neq T$). The total surface coverage depends upon the number of condensable species considered (e.g., H, T, O) and is given as the summation of coverages of all condensable species present.

If pore diffusion has a major impact on the tritium behaviour, the regular diffusion equation along the pore will need to be solved with a source term given by

$$S^N_j(t) = \bar{R}^N_{des,j}(t) - \bar{R}^N_{ads,j}(t). \quad (6)$$

When desorption/adsorption at the surfaces are very fast (i.e., not-rate controlling), and bulk diffusion is rate controlling, the source term can be expressed as the l.h.s. of Eq. (3).

However, if pore diffusion has a minor impact, a simplified version can be used. The concentration of species j in the pore, $\overline{C}_{\mathrm{pore},j}$, in this case can be given by the following equation:

$$\frac{\mathrm{d}\overline{C}_{\mathrm{pore},j}(t)}{\mathrm{d}t} = S_j^{\mathrm{N}}(t) - A_2^{\mathrm{N}}\left(\overline{C}_{\mathrm{pore},j}(t) - C_{\mathrm{p},j}(x,t)\right). \tag{7}$$

In Eq. (7) the first term on the r.h.s. represents the normalised source of species j in the pore given by Eq. (6) while the second accounts for the rate of release to the purge.

Finally, the conservation of species j along the purge, $\overline{C}_{\mathrm{p}j}$ assuming that convection predominates over diffusion, can be expressed by an equation of type

$$\frac{\partial C_{\mathrm{p},j}(x,t)}{\partial t} = A_3^{\mathrm{N}}\left(\overline{C}_{\mathrm{pore},j}(t) - C_{\mathrm{p},j}(x,t)\right) - A_4^{\mathrm{N}}\frac{\partial C_{\mathrm{p},j}(x,t)}{\partial x}. \tag{8}$$

In Eq. (8), the first term on the r.h.s. represents the rate of accumulation of species j in the He purge flow path calculated for the control-volume at the point x_k along the purge flow path. The second term represents the contribution of convection. Note that if pore diffusion can be completely neglected, the first term on the r.h.s. of Eq. (8) can be replaced by the source term of Eq. (6).

By solving the foregoing set of equations, the code calculates and keeps track of the tritium concentration distribution in all the regions together with the release resulting from any given transient history.

6. Conclusions and summary

Tritium behaviour in ceramic breeder materials of breeding blankets is a complex process which cannot be predicted accurately using a simple model. Some of the better known models have been surveyed in this paper emphasising the key transport mechanisms included, the application capabilities and the computational requirements. As blanket tritium analyses require the integration of the tritium inventory and release over the complete tritium generation rate and temperature contours within the blanket space (e.g., hundreds of node for each time step), the computational requirements of such an integrated analysis are complex and pose limit to the selection of the basic model to be used. Simple models are, in this respect, useful and prone to perform parametric design analyses, however, they lack the completeness of more comprehensive models and as such can easily overlook effects associated with excluded mechanisms which can be important in time and space as conditions change. Comprehensive models provide the right tool for a thorough analysis of tritium transport in the blanket, but could be cumbersome and complex if used for blanket parametric design calculations. Although, emphasis should be placed on fast running times, a TBDC has to model most of the mechanisms in sufficient detail to enable reliable predictions with variations in important parameters and/or operation conditions. Mechanistic models, which allow for detailed breeder microstructural descriptions, are in this case desirable, although, it is likely that an integrated TBDC will be composed of a mix of mechanistic and correlation models. Comprehensive versions of the mechanistic models would be most useful for final confirmatory analyses, which would build on the foundation set by extensive data analysis using simpler models. Although at an early development stage, desired characteristics of the model which would be best suited for blanket analysis are described in this paper together with some introductory considerations on the basic formulation of the phenomenological models included and some examples of the governing equations.

References

[1] M. Dalle Donne, C. Ferrero, C. Ronchi and F. Scaffidi-Argentina, in these Proceedings (ICFRM-6), J. Nucl. Mater. 212–215 (1994).
[2] S. Cho, A.R. Raffray and M.A. Abdou, in these Proceedings (ICFRM-6), J. Nucl. Mater. 212–215 (1994).
[3] I.J. Hastings, ed., Proc. 1st Specialists' Workshop on Modelling Tritium Behaviour in Fusion Blanket Ceramics, Chalk River, Canada, April 23–24, 1987.
[4] C.E. Johnson, ed., Proc. 2nd Specialists' Workshop on Modelling Tritium Behaviour in Fusion Blanket Ceramics, Indianapolis, Indiana, USA, April 27–28, 1989.
[5] H. Werle, ed., Proc. 3rd Specialists' Workshop on Modelling Tritium Behaviour in Fusion Blanket Ceramics, Karlsruhe, Germany, June 10–11, 1991.
[6] M. Yamawaki, ed., Proc. 1st Int. Workshop on Ceramic Breeder Blanket Interactions, Tokyo, Japan, October 26–29, 1992.
[7] G. Federici, C.H. Wu, A.R. Raffray and M.C. Billone, J. Nucl. Mater. 187 (1992) 1.
[8] G. Federici, C.H. Wu, A.R. Raffray and M.C. Billone, Proc. 17th Symp. on Fusion Technology (SOFT), vol. 2 (1993) pp. 1350–1354.
[9] J.P. Kopasz, S.W. Tam and C.E. Johnson, J. Nucl. Mater. 155–157 (1988) 500.
[10] J.P. Kopasz, S.W. Tam and C.E. Johnson, Modelling of Tritium Behaviour in Ceramic Breeder Materials, Argonne National Laboratory, Report ANL/FPP/TM-231 (1988).
[11] M.C. Billone, C.C. Lin, H. Attaya and Y. Gohar, Fusion Technol. 19 (1991) 976.
[12] G. Federici, A.R. Raffray and M.A. Abdou, J. Nucl. Mater. 173 (1990) 185.

[13] G. Federici, A.R. Raffray and M.A. Abdou, J. Nucl. Mater. 173 (1990) 214.
[14] A. Badawi and A.R. Raffray, Analysis of Surface Fluxes of Hydrogen Species in Lithium Ceramics, University of California, Los Angeles, Report UCLA-FNT-50 (1991).
[15] A. Badawi, A.R. Raffray and M.A. Abdou, Modelling and analysis of time-dependent tritium transport in lithium-containing ceramics, J. Nucl. Mater., submitted.
[16] The ITER Team, ITER Blanket, Shield and Material Data Base, ITER Documentation Series No. 29 (International Atomic Energy Agency, Vienna, 1991).
[17] A.R. Raffray, S. Cho and M.A. Abdou, Modelling of tritium transport in ceramic breeder single crystal, J. Nucl. Mater., submitted.
[18] G. Federici, A.R. Raffray, and M.A. Abdou, Proc. IEEE 13th Symp. on Fusion Engineering, Knoxville, TN, 1989, vol. 2 (IEEE Catalog. No. 89CH2820-9) p. 886.
[19] A. Badawi, A.R. Raffray, A. Ying and M.A. Abdou, Fusion Technol. 19 (1991) 1532.
[20] R. Toschi and the NET Team, Special Issue of Fusion Eng. Des. 21 (1993) 1–358.