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Progress in Tritium Retention and Release Modeling for Ceramic Breeders

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Abstract

Tritium behavior in ceramic breeder blankets is a key design issue for this class of blanket because of its impact on safety and fuel self-sufficiency. Over the past 10-15 years, substantial theoretical and experimental efforts have been dedicated world-wide to develop a better understanding of tritium transport in ceramic breeders. Models that are available today seem to cover reasonably well all the key physical transport and trapping mechanisms. They have allowed for reasonable interpretation and reproduction of experimental data and have helped in pointing out deficiencies in material property data base, in providing guidance for future experiments, and in analyzing blanket tritium behavior.

This paper highlights the progress in tritium modeling over the last decade. Key tritium transport mechanisms are briefly described along with the more recent and sophisticated models developed to help understand them. Recent experimental data are highlighted and model calibration and validation discussed. Finally, example applications to blanket cases are shown as illustration of progress in the prediction of ceramic breeder blanket tritium inventory.

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Introduction

An important aspect of the design and analysis of ceramic breeder blankets is the ability to predict the phenomenological behavior of tritium in the ceramic breeder under operating reactor conditions. By understanding the behavior 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 fuel self-sufficiency considerations. Development of accurate methodologies for modeling the behaviour of tritium in ceramic breeder materials represents a challenging task and requires close interaction with a parallel program of experimental activities. The schematic diagram of Figure 1 illustrates the high complexity of the problem under consideration and elucidates the evolutionary process of tritium modeling development and its close links with complementary laboratory and in-reactor fission experiments.

As results from progressively more integrated experiments become available, more accurate and sophisticated models are required to help in the analysis and interpretation of these results. Such analysis enables the estimate of missing property data and provides guidance for planning new experiments based on gaps in the understanding and/or data base. More accurate models also help to better extrapolate to blanket conditions and produce a more informed estimate of tritium inventory. The final step in modeling development is the development of reliable and calibrated design codes for overall blanket analysis.

This paper focuses on progress over the last 10-15 years in modeling of tritium behavior in ceramic breeders. First, a summary of model development is presented with a focus on the integrated modeling of the key transport processes. Advances in modeling are strongly linked to a well-planned experimental program which enables calibration and validation of the models. Some recent experimental data with high impact on the progress in modeling are discussed next. Examples of model application for estimating fundamental property data, and for

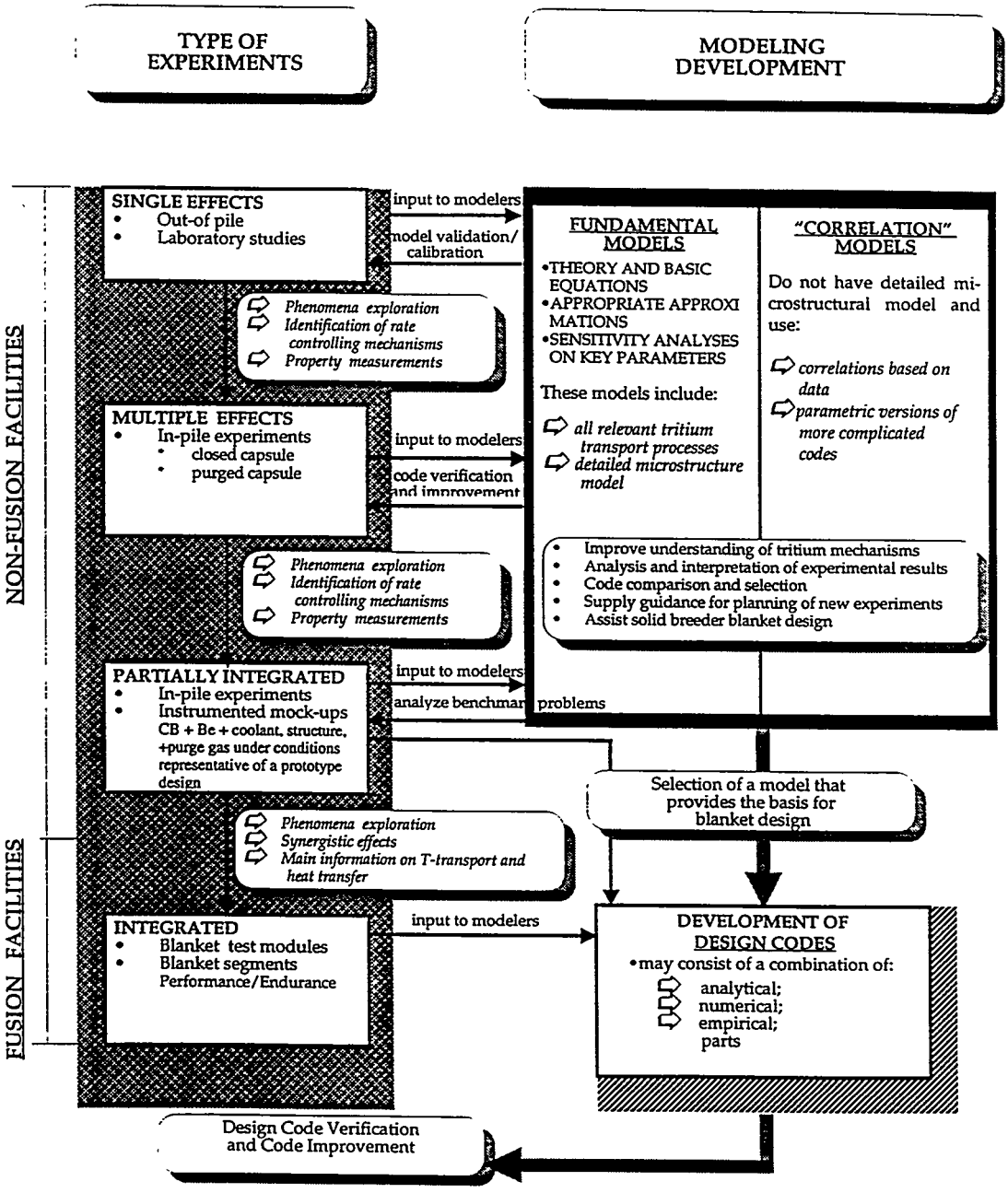

Figure 1 Experiment and modeling needs to develop a comprehensive understanding of tritium behavior in ceramic breeders.

calibration and validation purposes are then described. The parallel progress in modeling and experiments has enabled more accurate inventory predictions for blanket application, and the final part of the paper highlights the convergence towards better, more accurate blanket inventory predictions over the last 15 years.

Model Development

Model development involves two complementary approaches. First, the key tritium transport mechanisms need to be well understood and modeled. These include diffusion through the bulk, solubility, LiOT precipitation under certain conditions, surface processes and percolation through the interconnected porosity. Radiation effects are also important as they might create trapping in the bulk. This type of models helps in estimating property data from well-controlled experiments involving a limited number of transport mechanisms. The second modeling approach focuses on the development of more integrated models including several transport mechanisms based on the understanding of the different phenomena that the first approach helps to provide. These more comprehensive models would help in the analysis of the more integrated experiments and in extrapolating to prototypical conditions to provide blanket inventory estimates.

Overall, as evidenced for example by the achievements reported in five recent modeling workshops [1-5], co-ordinated through the IEA annex II implementing agreement on Radiation Damage in Fusion Materials, considerable progress has been made in recent years in the understanding of individual transport mechanisms and in the development of verified and validated methodologies to interpret available experimental data and to predict tritium inventory in fusion ceramic blankets. A comprehensive survey of modeling development has recently appeared in the literature [6], and the interested reader is there referred to find more detailed reviews of key transport/ release mechanisms, rate-limiting steps, material data-base status and

needs, model validation/ calibration issues, analysis on parameter ranges of applicability of available models, areas of latest advance and remaining open questions. Here, a summary of these advances in modeling over the last decade is given.

Tritium diffusion mechanism in lithium ceramics has been investigated by many researchers. Ohno et al.,[7-10] showed a close relationship between the lithium ion diffusion and the tritium diffusion by ion conductivity measurement. Noda et al.,[11-13] measured ion conductivity after ion irradiation and under ion bombardment .In post-irradiation experiments with Li_2O, a decrease in ion conductivity with ion fluence was found and attributed to the introduction of F^+ centers. Below 440 K, the observed increase of conductivity was attributed to the production of lithium ion vacancy. Ion irradiation was found to increase the ion conductivity. This showed the possibility of tritium-diffusion enhancement under irradiation. Kudo showed close correlation between tritium diffusivities of various lithium ceramics with lithium atomic density, the diffusivity increasing with lithium atomic density [14]. The activation energy for tritium diffusion was also found to be strongly dependent on Li to oxygen atomic-density ratio. These results suggest that there is an interaction between Li^+ and OT^- ions in the crystal.

The proposed diffusion mechanism from these studies was a lithium vacancy - tritium coupling process. Namely, for T jumping from an O^{2-} site to another O^{2-} site, Li^+ acts as a barrier. If a Li vacancy diffuses to this site the barrier for T^+ diffusion can be lowered, resulting in larger diffusivity. If a F^+ center traps a Li vacancy, then tritium diffusivity decreases. This mechanism was supported by Shulger et al., with quantum-chemical calculation [15]. They calculated the potential barrier (activation energy) for tritium migration in Li_2O crystal with lithium vacancies and F^+ centers. Activation energies below 0.9 eV were calculated for a T^+ ion in the lithium vacancy site to transfer to another lithium vacancy site. An activation energy of about 1 eV was calculated for T^0 in an interstitial site to jump to a

neighbouring interstitial site, and an activation energy of 1.1 eV for T^0 trapped in a Li vacancy to exchange with the nearest lattice lithium.

The understanding of neutron irradiation effect on tritium bulk diffusion in lithium ceramics has not known comparable progress. Experimental results do not show a decisive trend: post-irradiation tritium release showed decrease [16] and increase [17] of tritium diffusivity and no clear difference has been reported in tritium diffusivity between non-irradiation and irradiation experiments. However, recently the BEATRIX-II experiment indicated no serious decrease in tritium diffusivity in Li_2O up to lithium burn-ups of 5%[18].

Neutron irradiation effects can influence not only tritium diffusivity but other processes such as trapping by defect and change of the tritium chemical form (T^+, T^0, or T^-). Interaction with point defects such as F^+ and F^0 has been experimentally studied and modeled by Moriyama, et al., [19,20]. They suggested F^+ center traps tritium (T^+) and reduces it to T^0. By considering kinetic processes including diffusion of defects, they tried to explain the results of post-irradiation test of the tritium chemical form in Li_2O in-situ tritium release experiments and in-situ luminescence observation [21,22]. Interaction with other defects and synergism with tritium migration behavior require further experimental and modeling studies.

Tritium or hydrogen isotope solubility in lithium ceramics has been measured by several researchers [23,24]. However scattering of data exists in H_2 or T_2 absorption. Further experimental studies are required under controlled oxygen potential. No studies have been reported on physical modelling of tritium solubility although solubility data have been systematised by a phenomenological method.

In Li_2O, precipitation of LiOT or LiOH occurs when the equilibrium vapor pressure (or concentration in the bulk) is higher than the precipitation limit which is determined by the temperature. This is especially important when the blanket is operated at low temperature or in

pulsed operation mode. For physical modelling of this process, the mechanism of precipitation needs to be recognised. Tam tried to model this phenomena by using a technique similar to Zener's theory of precipitation [25]. The physical basis of the theoretical frame work proposes that, as a result of neutron irradiation, tritium/lithium vacancy complexes are formed. Precipitation occurs when these complexes migrate together to form clusters above certain critical sizes. This process is most likely to occur at grain boundaries, dislocation cores, and other regions of inhomogeneities.

Surface reactions have been considered to be an important process, because, if not enhanced by addition of H_2 or H_2O to the sweep gas, they can become the rate-controlling process to determine the tritium inventory in the solid breeders. Because of this, surface reactions in conjunction with bulk diffusion have often been the basis for the integrated modeling approach which built on the early single-mechanism models (e.g. diffusion only). Although useful, these early models were built on simple analytical solutions of governing equations under the assumption of simplified boundary conditions and, as such, could be applicable only over a limited range of experimental conditions.

Two mechanism-models, including a combination of diffusion step in the grains and desorption at the grain surfaces with a single desorption activation energy, were subsequently developed [26,27]. They represented an improvement but they too were unable to account for some of the experimental data [28]. As limitations in the predictive capabilities of these models were discovered, additional mechanisms thought to have a relevant effect were included and more sophisticated models developed. In particular, the complex morphology of the breeder, the presence of temperature gradients, the presence of different species in the purge stream (e.g., protium or moisture), the surface processes and their dependence on the amount of adsorbed species emerged to be key factors controlling the tritium build-up in and the release from the material and as such had to be included in the new models that were being developed.

A substantial conceptual improvement in the modelling has been achieved recently with the development of fairly comprehensive codes, based on finite-difference time-stepping algorithms which include details of the microstructure of the breeder, in conjunction with algorithms for some of the following mechanisms: diffusion, desorption, adsorption, solubility, chemical trapping (e.g., precipitation), and radiation-induced trapping (e.g., defects and/or He bubbles). They allow for one- and some of them a quasi-two dimensional description of the physical problem as they include space temperature and tritium production profiles together with variation of the concentration along the purge. Most of these state-of-the-art models have been described at length in the literature (i.e., MISTRAL [29,30], TIARA [31,32], DISPL2 [33], DAD [34]). In general, these models seem to cover an adequate selection of physical mechanisms and numerical methods and have allowed for reasonable interpretation and reproduction of experimental data, and they have helped in pointing out deficiencies in material property data base and in providing guidance for future experiments.

Compared to early models, present day models are fairly sophisticated; they allow for both steady-state and transient analyses, but the computational requirements have become more demanding and complex particular for parametric analysis which are often required for blanket analysis. In addition, they have been partially verified/ calibrated for results from in-fission reactor tritium release tests (typically small volume samples with quasi-uniform temperature distributions) on the basis of the data-base available, from laboratory studies and from in-pile experiments for some of the materials of interest.

All the blanket tritium inventory results presented in the past were obtained by performing the calculation with the local models at a limited number of representative poloidal and radial locations in the blanket. The results obtained for each of these selected control volume locations were then combined, externally to the code, to give whole-blanket tritium inventories and release rate. Therefore, it is clearly appealing to develop a tritium blanket design code

which 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. A review of different models available for analysis of tritium transport in ceramic breeders including a critical assessment of their applicability for blanket analysis and of their limits has been presented at ICFRM-6 [35]. 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 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 this type of model 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 blanket system code. Codes which use correlations based on experimental set of data or on parametric versions of more complicated models rather than the fundamental models themselves, could be attractive for applications that require reduced detail and accuracy for fast running times.

As a final remark, it must be mentioned that despite the modeling progress which has taken place, and the growing amount of experimental data available today, knowledge is still far from being complete and there are still several areas where additional work remains to be done to ensure development of more rigorous predictive capabilities. Major difficulties encountered in interpreting, comparing, and extrapolating the data for the different ceramic breeders are due to the lack of fundamental property data, incomplete characterisation of the sample micro-structure, impurity levels, and surrounding chemistry. The areas where additional modeling focus is required include: dissolution, LiOT precipitation in the bulk, and radiation-induced trapping and high burn-up effects which are not fully characterised yet and for which few data are currently available.

Experimental Data

The progress in modeling tritium retention and release in lithium–based ceramics has paralleled the expansion of the mechanistic laboratory data base on irradiated and unirradiated materials and the integral in–reactor purge–flow–capsule data base. The laboratory studies have been used to determine model parameters for the basic mechanisms of tritium diffusion through the lattice or bulk, surface adsorption and desorption, solubility and precipitation. The in–reactor purge–flow experimental data have been used to validate the prediction of computer models, as well as to refine both the models and the model parameters. Most of the data for Li_2O, Li_2ZrO_3, Li_4SiO_4, and $LiAlO_2$ available up until 1992 are summarized in Ref. [36]. This document will soon be updated to include more recent data on these ceramics, as well as to expand the data base to include another ceramic which shows considerable promise: Li_2TiO_3.

The in–reactor purge–flow data base includes on–line monitoring of transient tritium release during temperature, purge–composition, purge–flow rate, and tritium–generation–rate transients. Such data have proven invaluable in model development and validation. In addition, for many of these experiments, the tritium inventory was measured directly under controlled laboratory conditions following the last experimental run under relatively steady–state conditions. It is beyond the scope of the current effort to summarize all of these data. Table 1 gives a brief summary of the purged, in–reactor tests and the experimental conditions for four of the breeder ceramics. In general, the reference conditions for these tests include a He purge flow rate of 100 ml/min and a protium content of 0.1 vol. %. Purge flow rates from 50 to 200 ml/min have also been studied. The sample microstructures cover a wide range of densities and grain sizes. For designs involving sintered–products in the form of solid or annular cylinders or plates, densities of 80–85% are usually specified. Higher densities are sometimes specified (e.g., Li_4SiO_4) for pebble bed applications. The optimum grain size

Table 1. Summary of purged in–reactor experiments to determine tritium retention/release for several ceramic breeders.

depends on the material; for example, $LiAlO_2$ exhibits much better tritium release characteristics with sub–micron grain size. The wide range of temperatures investigated are ideal for model validation purposes, and they adequately cover the design ranges of interest. The tritium generation rates for most experiments are low relative to design applications. However, the peak generation rates approach those anticipated in design applications, with the Li_2O experimental peak corresponding to a peak design neutron wall loading of ~3 MW/m^2. The peak experimental burnups are low relative to anticipated design peaks at 3 $MW\text{-}a/m^2$. This may be an important factor for the ternary ceramics which undergo a change in stoichiometry, and perhaps performance, with burnup. Experiments to higher burnup in a fast–neutron spectrum are currently being planned for the ternary ceramics.

The end–of–life tritium inventory data are very good for Li_2O and relatively sparse for the other breeder ceramics. As will be shown in the Model Calibration and Validation section, these data are important in validating the steady–state inventory predictions. More data on the ternary ceramics (e.g., Li_2ZrO_3 from BEATRIX–II and CRITIC–II) are anticipated from experiments which are either completed or are nearing completion.

Model Application

Models vary in complexity from a one–mechanism model or correlation (e.g., tritium residency time correlation) to a multi–mechanism model with pre–exponentials and activation energies fit to experimental data to a multi–mechanism model with pre–exponentials and activation energies determined from fundamental principles. These models are calibrated and validated by comparing model predictions to experimental data. Essential to this process is the characterization of fundamental property data, such as bulk diffusion coefficient, which can be used as input in the model analysis. For illustration purposes, examples of two types of model application are described here. The first one is the application of the MISTRAL-SC code to a

set of tritium release results for $LiAlO_2$ single crystal to help determine the bulk diffusion coefficient. The second one is the application of the TIARA model to the end-of-life inventory data obtained from in-reactor purge experiments with Li_2O for calibration and validation purposes.

Example of Fundamental Property Data Estimate

MISTRAL-SC is a code developed specifically for the analysis of tritium release from lithium ceramic single crystals. It incorporates bulk diffusion and the four major surface processes and allows for variation of surface activation energies with coverage and for the presence of H_2 in the purge. The model is based on the methodology of Federici, et al., [29, 30] and is described in details in Ref. [37].

It was applied to a recent set of tritium release results from $LiAlO_2$ single crystal from Botter, et al., [38] including both out-of-pile isothermal and constant heating rate anneals. Simple diffusion, desorption and diffusion/desorption models were found to poorly reproduce the results suggesting the need for a more comprehensive model to better understand the results.

The tritium diffusion coefficient for the $LiAlO_2$ single crystal, D_{sc}, was estimated by reproducing the tritium release results for the constant temperature anneal cases based on a consistent set of input parameters.

$$D_{sc}(m/s) = 1.29x10^{-5}\exp(\frac{-141.5kJ/mol}{RT}) \qquad (2)$$

where R is the universal gas constant and T the temperature.

The results were verified by reproducing the constant heating rate experimental results with the same set of input parameters. Finally, sensitivity analyses based on the individual variation of several input parameters indicated the robustness of the results [37]. Example results for both the constant temperature and constant heating rate anneals are shown in Figures 2 and 3 to illustrate the comparison of the model calculations with the experimental results.

Multi-Mechanism Model Calibration and Validation

The experimental data in Table 1 are often summarized in terms of a tritium residency time ($\tau = \frac{I}{G}$, where I is the inventory and G the generation rate). Correlations can be developed for τ as a function of temperature within a narrow range of fabrication and operational parameters. Figure 4 shows the inventory data for Li_2O normalized to the generation rate for purged, in–reactor experiments for which the tritium inventory was measured directly after the test. The solid line in the figure can be represented by the correlation:

$$\tau(s) = 1.04 \exp\left(\frac{56 kJ / mol}{RT}\right)^2 \qquad (1)$$

This correlation is useful for quick, easy, bounding design calculations for a standard He purge with 0.1% H_2 and for densities in the range of 80–86% TD. However, for higher densities and/or lower H_2 concentrations in the purge, the correlation does not apply. A more fundamental approach is required.

The TIARA code[31,32] has been developed to analyze the steady–state inventory in Li_2O and the composition of the purge stream exiting the breeder region. Inventory predictions are based on models for bulk diffusion, surface desorption, and solubility. TIARA also has the capability of predicting LiOH/T precipitation. The diffusion model has a pre–exponential and activation energy which are determined from post–irradiation annealing of single crystals.

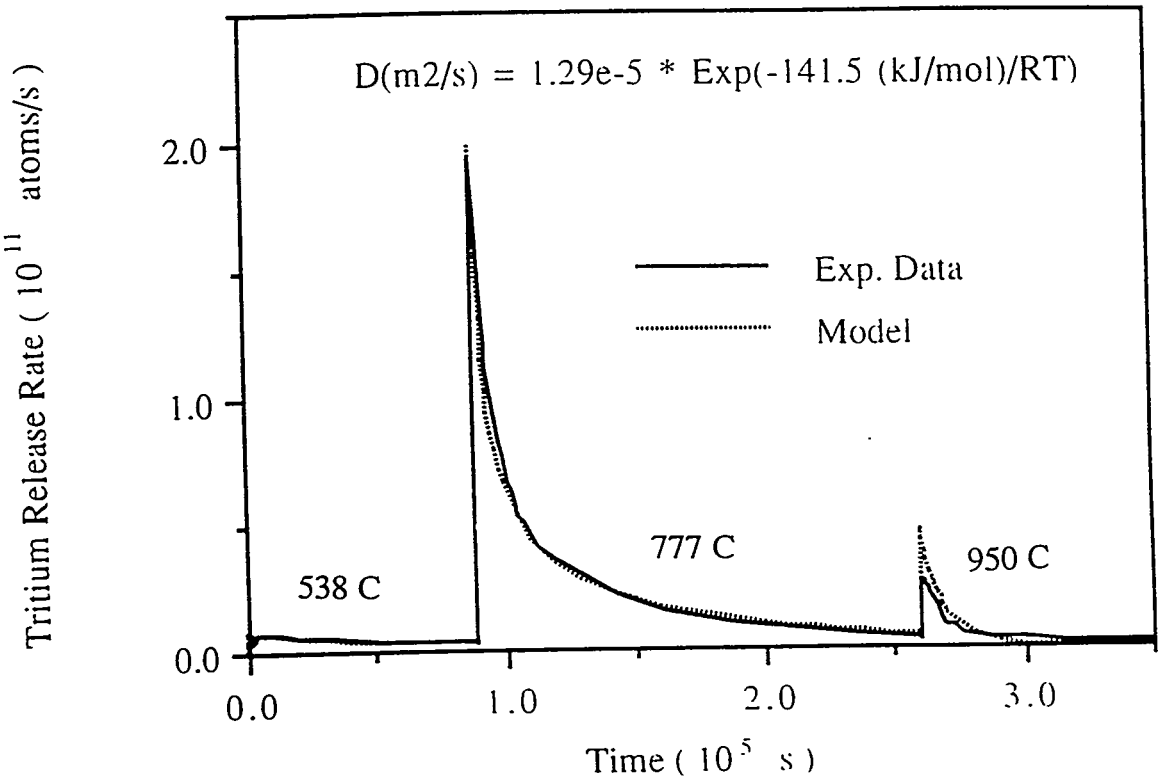

Figure 2 Comparison of MISTRAL-SC prediction with experimental data for tritium release from 2-mm $LiAlO_2$ single crystal under 3 successive constant temperature anneals (538 °K, 777°C and 950 °C) [35].

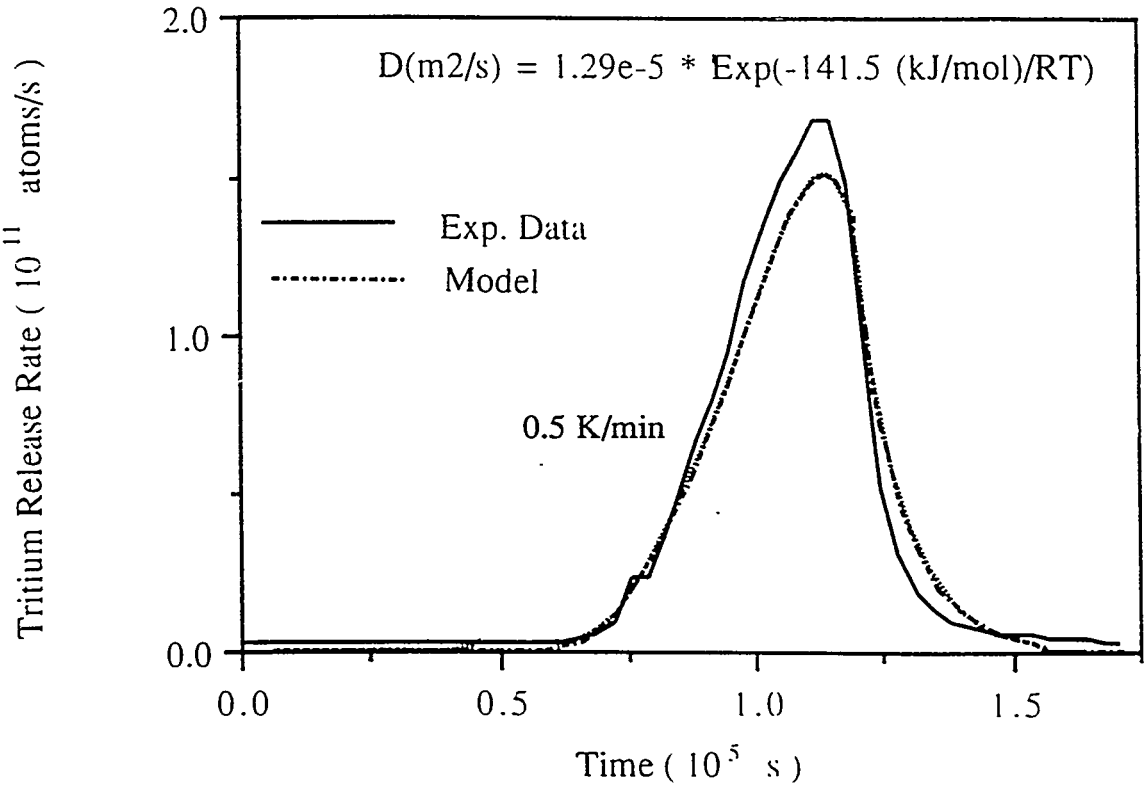

Figure 3 Comparison of MISTRAL-SC prediction with experimental data for tritium release from 2-mm $LiAlO_2$ single crystal under a constant heating rate corresponding to 0.5 K/min[35].

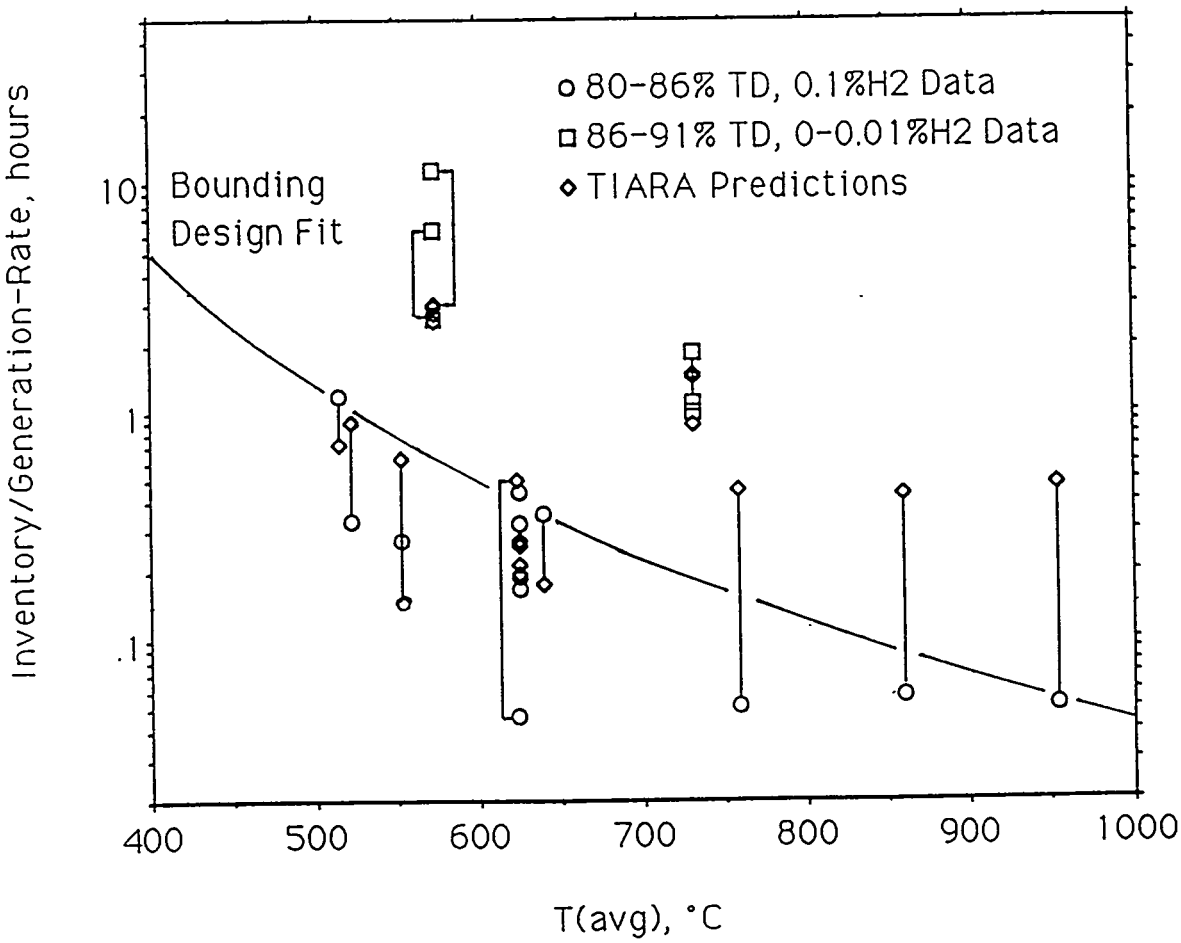

Figure 4 Comparison of TIARA and design correlation to experimental data for tritium residency times.

The surface desorption model has an activation energy determined from post–irradiation annealing of porous, polycrystalline samples and a pre–exponential dependent on protium pressure and pore–solid surface area. The solubility model is dependent on temperature and gas phase partial pressures. Thus, all of the model parameters in TIARA are determined from the results of laboratory experiments on unirradiated and irradiated Li_2O. Auxiliary models are also included to calculate the gas–phase partial pressures of tritiated species in the interconnected porosity and in the purge.

Of the 20 data points shown in Fig.4, two were used to fine–tune the model parameters for surface desorption at T > 470°C and for moisture solubility at T < 1000°C. The remaining 18 were used for validation in which no model parameters were changed, as described in detail in Ref. [32]. The validation results are also shown in Fig. 4. For all of the validation cases, only surface desorption and solubility were predicted to contribute significantly to the total tritium inventory. Although no model parameters were adjusted for this validation exercise, the mean of the TIARA predictions matches the mean of the inventory data (0.48 wppm). Good agreement was achieved between predictions and data for the BEATRIX–II thin–ring, BEATRIX–II thick–pellet near the outer radius, CRITIC–1, EXOTIC–2, and VOM–15H results, which include H_2 purge levels from ~0 to 0.1% and densities from 80 – 92%. However, one of the SIBELIUS data points and four of the BEATRIX–II thick–pellet data points were over–predicted. These data are all in the low range of 0.06 - 0.10 wppm whereas the predictions are in the range of 0.6 wppm. Notice that the TIARA results have the advantage over the residency correlation of being a better fit to the data over a wider range of densities and H_2 levels. In effect, TIARA reduces down to a two–mechanism model with the physically–realistic variables of pore–solid surface area and gas–phase pressures included in the model. However, there are some limitations even in this approach. The four BEATRIX–II thick–pellet data points which are over–predicted all occur in the dense, large–grain–size portions of the pellet. The over–prediction for these points, as well as the over–prediction of solubility and surface desorption inventories for single crystals studied in the laboratory,

suggest that the TIARA model parameters may not extrapolate well to microstructures approaching high density and large–grained crystals. Perhaps a more fundamental approach (e.g., MISTRAL) is needed to model the whole range of microstructures included in the experimental program. However, with regard to microstructures anticipated for design use, as well as relevant ranges of operating conditions, TIARA does a very good job of predicting tritium inventory under steady–state conditions.

Blanket Application

Ceramic breeders are considered as major blanket candidates. A key consideration in the design of these blankets is the tritium inventory. It affects safety as well as economics through its effect on the reactor tritium self-sufficiency. Over the last fifteen years, the ceramic breeder property data base has been considerably extended through a program of laboratory and irradiation experiments. This, coupled to the progress in model development, has enabled more accurate characterization of tritium transport mechanisms and has significantly reduced the uncertainties in tritium inventory prediction for fusion blanket application. As an example, one can consider the case of tritium diffusion in $LiAlO_2$.

The last 10-15 years have seen the completion of several in-situ purged capsule experiments, such as TRIO[39], LISA[40] and VOM[41], where the tritium release behavior of small-grained $LiAlO_2$ specimens was observed under temperature and purge flow composition transients. The diffusion coefficient of tritium in $LiAlO_2$ was estimated from these results under the assumption of diffusion as the rate-controlling mechanism. Order-of-magnitude type variations was observed among these diffusion coefficients, as summarized in Ref. [42] which, based on these results, recommended the following expression for the diffusion coefficient, D ± a factor of 5.

$$D(m/s) = 1.1x10^{-10} \exp(\frac{-135kJ/mol}{RT}) \qquad (3)$$

The diffusion coefficient estimated recently from single crystal results (Eq. (2)) is much higher than these previously estimated values for small-grained $LiAlO_2$. This indicates that in the latter case, other tritium transport mechanisms played a role in the tritium release process. Performing a purely diffusive analysis based on grain size would then yield "effective" diffusion coefficients which would tend to be lower than the pure bulk diffusion coefficient.

Basing design calculations on the diffusion coefficient of Eq. (3) can lead to wrong conclusions. For example, the diffusive time constant, ($t_{dif} = \frac{r_g^2}{15D}$), for a typical $LiAlO_2$ grain of radius, r_g = 0.1 μm, based on Eq. (3) and including the uncertainty band is shown in Figure 5 as a function of temperature. At temperatures below about 450 °C, the time constant is of the order of a day or more which is not acceptable based on blanket tritium inventory considerations.

On the same figure is also shown the diffusive time constant for a 0.2 μm grain $LiAlO_2$ based on the single crystal diffusion coefficient of Eq. (2). The conclusion changes significantly now since diffusion by itself appears not to be of particular concern for temperatures as low as 300 °C based on inventory considerations. The focus should then be on other mechanisms such as surface reactions which would have a strong influence on the total inventory and on measures which could alleviate the situation accordingly. For example, addition of small amount of protium (0.1%-1%) to the purge is known to reduce the tritium surface inventory through isotope swamping.

The effect of the overall progress in data base expansion, modeling and prediction capability can be illustrated by considering tritium inventory predictions for ceramic breeder blankets from past studies which in the absence of adequate analytical tools and fundamental

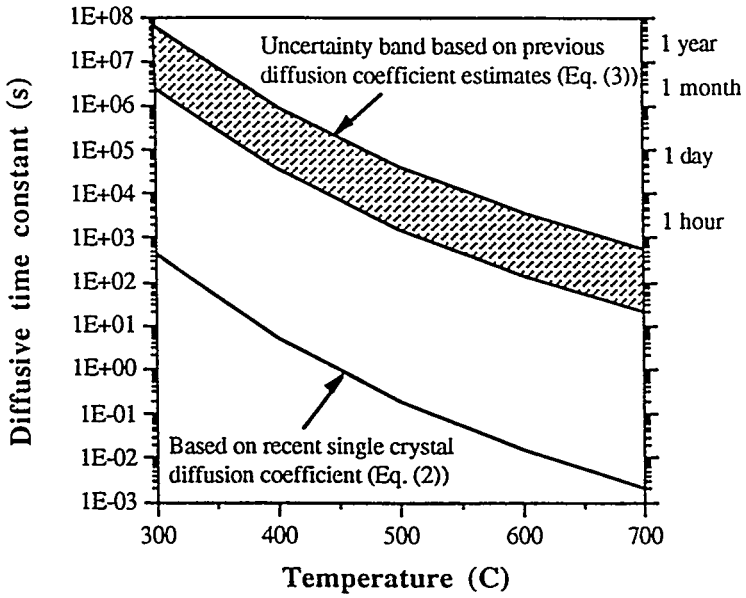

Figure 5. Diffusive time constant for 0.2 μm $LiAlO_2$ grain based on previous diffusion coefficient estimates (Eq. (3)) and single crystal diffusion coefficient estimate (Eq. (2)) as a function of temperature.

property data characterization tended to be overly conservative. As an illustration, Figure 6 shows a chronology of tritium inventory estimates for ceramic breeder blankets based on a number of different reactor studies performed over the last 10-15 years (including one inertial confinement reactor blanket [48]). Admittedly, specific inventory predictions are dependent on a number of parameters which may differ from study to study. These include reactor type and power, ceramic breeder material and configuration, and operating conditions. However, these parameters are closely interrelated and it is difficult, if not impossible, to scale or normalize the inventory estimates based on the independent effect of the above parameters in order to provide a consistent comparison. For example, a different fusion power level would result in different operating parameters and possibly in a different blanket configuration choice, the combination of which would dictate the overall inventory for this particular reactor system. Thus, Fig. 6 is intended more as a qualitative illustration than as a consistent comparison. Clearly, though,

both the magnitude of and uncertainty in ceramic breeder blanket tritium inventory have decreased substantially over the years with the expansion of the fundamental data base and the refinement of modeling tools.

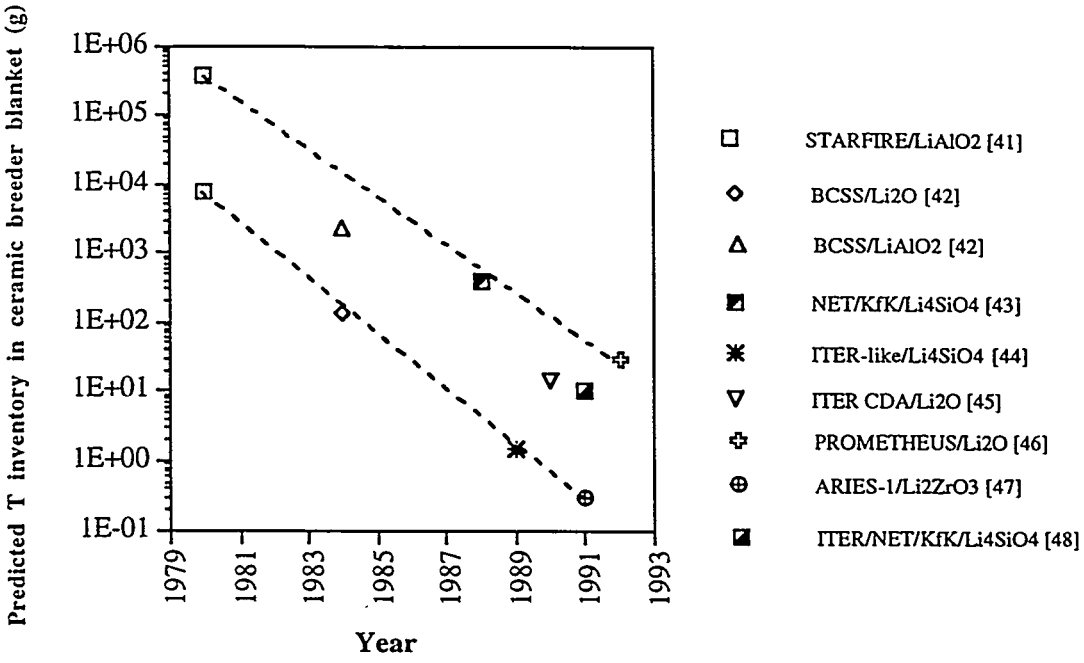

Figure 6. Blanket tritium inventory estimates in ceramic breeder reactor studies over the years.

Blanket tritium inventory estimates for the early STARFIRE study[43], where minimization of the tritium inventory was a key factor in the choice of $LiAlO_2$ as ceramic breeder, ranged up to 8 kg in the absence of radiation effects and up to 380 kg if radiation effects were included. In contrast, estimates of tritium inventory in the Li_2O ceramic breeder blanket-of the recent ITER CDA study [47], based on more detailed and better calibrated modeling tools, ranged up to about 40 g. In the most recent ceramic blanket reactor studies, the inventory estimates range from about 1 g in the Li_2ZrO_3 blanket of ARIES-I [49] to about 10 g in ther Li_4SiO_4 blanket of Ref. [50]. It is possible that at much higher fluence and burnup,

radiation effects could create some form of trapping not observed experimentally yet, which would result in higher inventory. However, it is clear that, in perspective, quite remarkable progress has been achieved in developing better and more accurate tritium release and inventory models for ceramic breeders. In combination with the larger experimental data base available for calibration, these models have enabled more accurate tritium inventory predictions of the order of 100-1000 lower than were conservatively predicted 10-15 years ago, thereby making ceramic breeder blankets much more attractive on this basis.

Conclusions

Remarkable progress has been achieved through the last decade in the understanding of tritium transport mechanisms in ceramic breeders through a complementary program of experiments and model development. The subject is so complex that much still needs to be done to arrive at a full understanding. However, advances have enabled development of models that have helped in the evaluation of fundamental property data and, through calibration and validation with experimental data, in more accurate estimates when extrapolating to blanket conditions.

Looking in perspective at the chronology of tritium inventory estimates in ceramic breeder blanket provides a simple illustration of the progress made in model development and data base expansion. Initial estimates in the absence of adequate modeling tools and data base were overly conservative and ranged as high as 380 kg. In contrast, recent estimates based on the more sophisticated and accurate tritium release and inventory models over the last few years range from about 1 to 100 g, thereby making ceramic breeder blankets much more attractive on this basis.

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Table 1. Summary of purged in–reactor experiments to determine tritium retention/release for several ceramic breeders.

Parameter	Li_2O	Li_2ZrO_3	Li_4SiO_4	$LiAlO_2$
Experiment	BEATRIX–II	BEATRIX–II	——	——
	COMPLIMENT	COMPLIMENT	COMPLIMENT	COMPLIMENT
	CRITIC–I	CRITIC–II	——	——
	EXOTIC–2	EXOTIC–3,4,6	EXOTIC–5,6	EXOTIC–1,5,6
	——	——	——	LiLa–1,3
	——	——	LISA–1,2	LISA–1
	MOZART	MOZART	——	MOZART
	SIBELIUS	SIBELIUS	SIBELIUS	SIBELIUS
	——	——	——	TEQUILA–1
	——	——	——	TRIO
	TTTex	——	——	——
	VOM–15H,22H	——	VOM–23	VOM–22H,23
	——	TRIDEX	TRIDEX	——
Density (%TD)	79–92	69–86	85–98	57–93
Grain size (μm)	5–60	0.4–40	10–120	0.2–20
Temperature (°C)	310–1000	200–1100	270–800	350–900
H_2 in purge (%)	0.0–1.0	0.0–1.0	0.0–1.0	0.0–1.0
Peak Tritium Gen. Rate (wppm/day)	35	10	13	6
Peak Li burnup (at.%)	5	5	3	3
Tritium Inventory (wppm)	0.05–1.5	0.01–2.0	3.0–52	0.1–20
Number of Inventory Points	21	3	3	5