

# Analysis of tritium kinetics of SIBELIUS beryllium

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## Abstract

A tritium transport model called BETTY has been developed to describe and predict the kinetics of tritium transport in irradiated beryllium in fusion blanket applications. Some USA SIBELIUS experimental data were released and provided useful input data for tritium transport models and code development. An analysis of these USA SIBELIUS experimental data was performed in this paper. A comparison of the tritium kinetics results predicted by the model with those from the experiments shows reasonable agreements. In addition, a parametric sensitivity analysis for the variance of surface activation energies and diffusion coefficient is performed and the minimal effects are observed based on the modeling results. © 2000 Elsevier Science B.V. All rights reserved.

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## 1. Introduction

Beryllium is considered as both a neutron multiplier material and a plasma-facing component material for fusion reactors. Tritium release kinetics in beryllium is an important issue in fusion blankets. In order to handle this issue, a thorough understanding of the mechanisms of tritium kinetics in beryllium is required.

Several experiments [1,2] were carried out in the HFR (High Flux Reactor) reactor to investigate the tritium release kinetics in irradiated beryllium pebbles. The beryllium irradiated in these experiments consists of a mix of about 2 mm and 0.1–0.2 mm diameter beryllium pebbles. Since the

beryllium pebbles may be used as the neutron multiplier of the DEMO relevant blanket as well as the International Thermonuclear Experimental Reactor (ITER) breeding blanket, it is important to obtain the tritium release/retention characteristics in the beryllium pebble beds. However, to understand the basic mechanism of the tritium kinetics and material properties of beryllium, required are the beryllium specimens with low BeO content and high fabrication density, such as SIBELIUS samples [3–6].

The tritium release data from the beryllium irradiated in ATR (Advanced Test Reactor) at low temperature are available for the post-irradiation annealing results and SIBELIUS experiments. The EC/USA collaborative SIBELIUS experiment was performed at CEN Grenoble to obtain information on the compatibility between beryllium and steel, as well as beryllium and ceramics,

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in a low neutron fluence condition and to provide additional understanding of tritium behavior in irradiated beryllium [6]. Baldwin [7] reexamined previously presented measured and calculated tritium inventories of USA experimental data and provided useful input data for tritium transport models and code development. Those reexamined experimental data has the uncertainty of  $\pm 5\%$ .

Based on the long-term tritium release data from irradiated beryllium (SIBELIUS irradiation), the tritium inventory as a function of annealing time for capsules 2 and 5 are compared with the predictions of ANFIBE [8] that considered the tritium transport with helium bubble migration. The code ANFIBE predicted the residence time of neutron generated tritium in dense beryllium [9]. But for the investigation of short-term tritium release and material characteristics, it would be better to introduce the code BETTY [10].

The purpose of the work is to analyze these USA experimental data, and to validate a developed model, BETTY, for tritium release from Be. In addition, to help interpret and plan future experimental data for fusion blanket applications, a parametric sensitivity analysis for the variance of surface activation energies and diffusion coefficient is performed and the results of the analysis are discussed.

Table 1  
Characteristics of SIBELIUS beryllium sample

Sample name	Sample 2-1	Sample 5-1
Purge gas flow rate	$1.667 \times 10^{-6} \text{ m}^3/\text{s}$	
Purge gas component	He + 0.1% H <sub>2</sub>	
Irradiation temperature and time	550°C for 1690 h	
Neutron flux (#/cm <sup>2</sup> -s)	$1.1 \times 10^{14}$ (th) + $1 \times 10^{14}$ (fast)	
Neutron fluence	$6 \times 10^{20}$ (#/cm <sup>2</sup> )	
Weight (kg)	$1.829 \times 10^{-4}$	$1.865 \times 10^{-4}$
Sample disc geometry	(dia. × thick.) = 8 × 2 (mm)	
Average density	1805 (98% TD) (kg/m <sup>3</sup> )	
Tritium released (measured) (MBq)	112	130
Tritium released (calculated) (MBq)	198	188
<sup>4</sup> He contents (wppm)	58.56	63.19
BeO contents (wppm)	≤ 300	

## 2. Analysis of experimental data

The representative two SIBELIUS Be samples, 2-1, and 5-1, that are adjacent only to steel, were considered as baseline specimens for investigation of tritium release characteristics and material properties of Be. These samples are also chosen in tritium release analysis using ANFIBE [11]. The beryllium specimens, with high fabrication density (98% TD), were irradiated with low fluence ( $6 \times 10^{20}$  n/cm<sup>2</sup>) at relatively high temperature of 550°C for 1690 h [7]. Stepped isothermal anneal tritium release testing was performed, measuring tritium release rate over the temperature range from 550 to 850°C, in 100°C steps for about 24-h periods at each temperature. The main characteristics and features of these two samples are listed in Table 1. The total amount of tritium released are 112 MBq for sample 2-1 and 130 MBq for sample 5-1 [7]. However, based on the tritium release experimental data given by Baldwin, the total tritium release of 198 MBq for sample 2-1 and 188 MBq for sample 5-1 were obtained. In order to be consistent for these two different tritium amount, the tritium release experimental data are modified and the adjusted experimental data are shown in the figures below comparing with the modeling results.

In the SIBELIUS experiments, the effects of tritium trapping in He bubbles (He < 60 wppm) and tritium retardation in the BeO layer (BeO < 300 wppm) on tritium release would be relatively less important than bulk diffusion as well as surface processes at the solid/gas surface. So, in this analysis only pure Be bulk diffusion and surface processes are considered. The formulation of governing equations in the simplified model is described in the next section. In this work, BETTY was applied to the tritium release experimental data of samples 2-1 and 5-1 and a comparison of the results from the SIBELIUS experimental data to those from the model is performed. The reference property data and input parameters used in this analysis are summarized in Table 2 [12–15].

Table 2  
Reference property data and parameters used in this analysis

Parameters	Values	Reference
Pre-exponential diffusion coefficient ( $\text{m}^2/\text{s}$ )	$4.56 \times 10^{-7}$	[12]
<i>Activation energies (kJ/mol)</i>		
Diffusion ( $E_{\text{dif}}$ )	63	[12]
Bulk adsorption ( $E_{\beta}$ )	55.34	[12]
Dissolution ( $E_{\text{diss}}$ )	158.25	–
Solution ( $E_s$ )	96.48	[13]
Adsorption ( $E_{\text{ads}}$ )	48.24	[14]
Desorption ( $E_{\text{des}}$ )	54.67	[15]

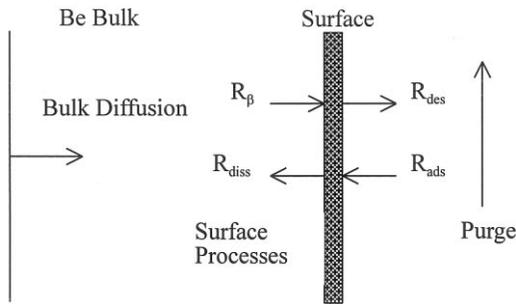


Fig. 1. Schematic of the model presented in this analysis for tritium kinetics in beryllium

### 3. Governing equations in the simplified model, BETTY

A tritium transport model, BETTY, has been developed to describe and predict the kinetics of tritium transport in beryllium in fusion blanket application [10]. A schematic of the simplified model presented in this analysis is shown in Fig. 1. The tritium released from the Be bulk may be considered to take place in several steps: (1) a tritium atom is produced in the bulk; (2) the tritium diffuses to the solid–gas interface; (3) it recombines with another atom and escapes as a molecule to the gas; and (4) it is removed by purge gas stream.

The 1-D model can be representative of the high density Be sample for normal release by using the appropriate definition of the length of bulk region. The bulk is modeled as slab geometry. In order to investigate the effect of surface

processes on the tritium release from the irradiated beryllium, a surface kinetics model for tritium release from Be was suggested. In this model, tritium diffusion in the bulk and all possible surface processes at the surfaces are considered. Considering four surface fluxes at the Be surface, the governing equation used in this model are summarized as follows.

#### 3.1. Be bulk diffusion equation

$$\frac{\partial C(x, t)}{\partial t} = D_{\text{Be}}(T) \frac{\partial^2 C(x, t)}{\partial x^2} + G_{\text{Be}} \quad (1)$$

where  $G_{\text{Be}}$  (atoms/ $\text{m}^3\text{-s}$ ) is the tritium generation rate assumed constant in the Be bulk. The diffusion coefficient of tritium in Be can be expressed as an Arrhenius function:

$$D_{\text{Be}}(T) = D_0 \exp(-E_{\text{dif}}/RT) \quad (\text{m}^2/\text{s}) \quad (2)$$

The boundary conditions are: (a) symmetry condition at the center of the sample:

$$\frac{\partial C(x, t)}{\partial x} = 0 \quad \text{at } x = 0 \quad (3)$$

(b) flux balance at the interface between Be bulk and purge flow:

$$-D_{\text{Be}} \frac{\partial C(x, t)}{\partial x} = R_{\beta} - R_{\text{diss}} \quad \text{at } x = L \quad (4)$$

where  $R_{\beta}$  and  $R_{\text{diss}}$  are the tritium flux from the bulk to surface and dissolution flux respectively, and the detail mathematical expressions are illustrated in Ref. [16]. The initial condition is

$$C(x, 0) = C(x) \quad \text{at } t = 0 \quad (5)$$

#### 3.2. Rate equation at the surface

$$N_s \frac{d\theta}{dt} = R_{\text{ads}} - R_{\text{des}} + R_{\beta} - R_{\text{diss}} \quad (6)$$

where  $\theta$  is the tritium surface coverage and  $N_s$  (sites/ $\text{m}^2$ ) is the number of surface sites per unit area. Also,  $R_{\text{ads}}$  (atoms/ $\text{m}^2\text{-s}$ ) is the absorption flux from the purge to the surface assuming dissociative adsorption and  $R_{\text{des}}$  (atoms/ $\text{m}^2\text{-s}$ ) is the desorption flux from the surface to the purge. The detail mathematical expressions of these fluxes are shown in Ref. [16]. The initial condition is

$$\theta = \theta_0 \quad \text{at } t = 0 \quad (7)$$

The governing equations consist of a parabolic partial differential equation and a stiff ordinary differential equation. The overall numerical scheme used for the solution is based on the well-proved scheme [16].

#### 4. Results and discussion

Based on the limited available data for activation energies and diffusion coefficient as shown in Table 2, the modeling results are compared to the experimental data for USA SIBELIUS sample 2-1 by release rate and integrated release amount and the results are shown in Fig. 2 and Fig. 3, respectively.

When the surface is not considered, only desorption and adsorption between bulk and purge are included. When the surface is considered, four surface fluxes are included in the endothermic Be surface coverage [6]. The modeling results without surface coverage show that only 79% of the total tritium are released at the end of the annealing temperature. Therefore, the overall shape of the

results is much below the experimental data. The modeling results considering surface coverage show that the amount of tritium released at 550 and 650°C are closely matched to the experimental data. The peaks of the modeling results at the next two temperatures reach those of the experimental data. The shape of the modeling results at 650°C shows a monotonous decrease which is reverse phenomena compared to the experimental data, where an increase of release rate after a small peak. The shape at 750°C decreases more slowly than the experimental data. However, the shape at 850°C follows the experimental data well at the first part and then decreases fast. Therefore, the modeling results reasonably reproduce the experimental data as the peak and the cumulative tritium release at each temperature as concerned.

Similarly, the modeling results are compared to the experimental data for sample 5-1 by release rate and integrated release amount and the results are shown in Fig. 4 and Fig. 5, respectively. When the surface coverage is not concerned, the modeling results show that only 80% of the total tritium are released. The shape of the modeling results at 850°C only follows the experimental data at the

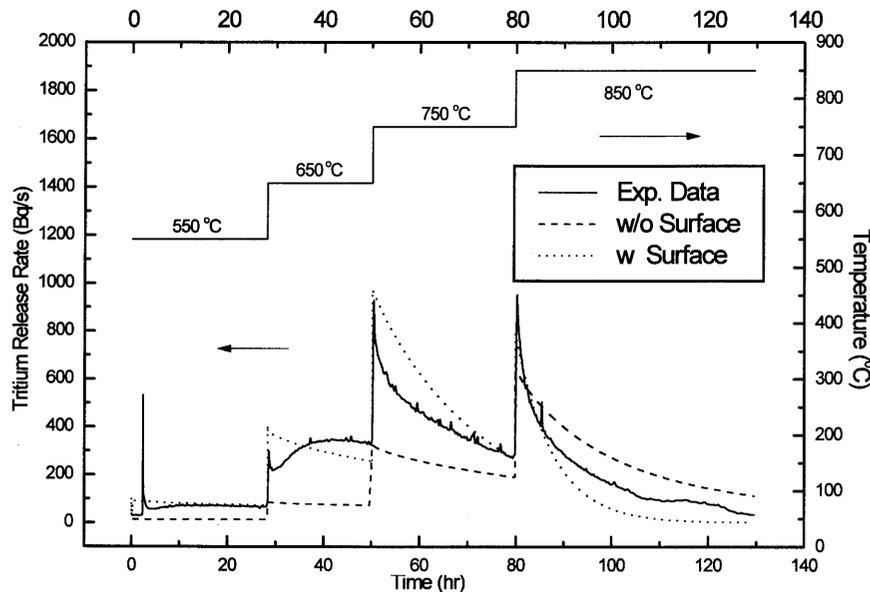


Fig. 2. Comparison of BETTY results with the experimental results for tritium release from SIBELIUS sample 2-1 under four successive temperature anneals.

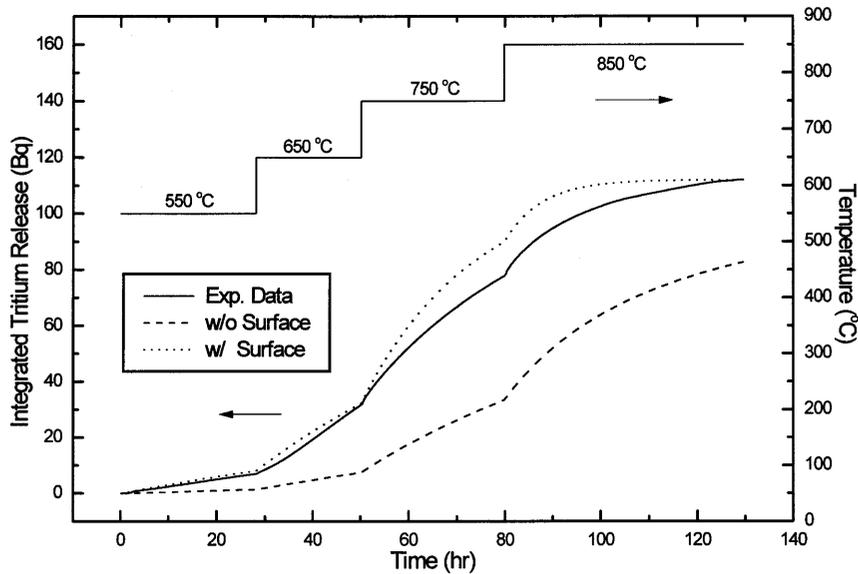


Fig. 3. Comparison of BETTY results with the experimental results for integrated tritium release from SIBELIUS sample 2-1 under four successive temperature anneals.

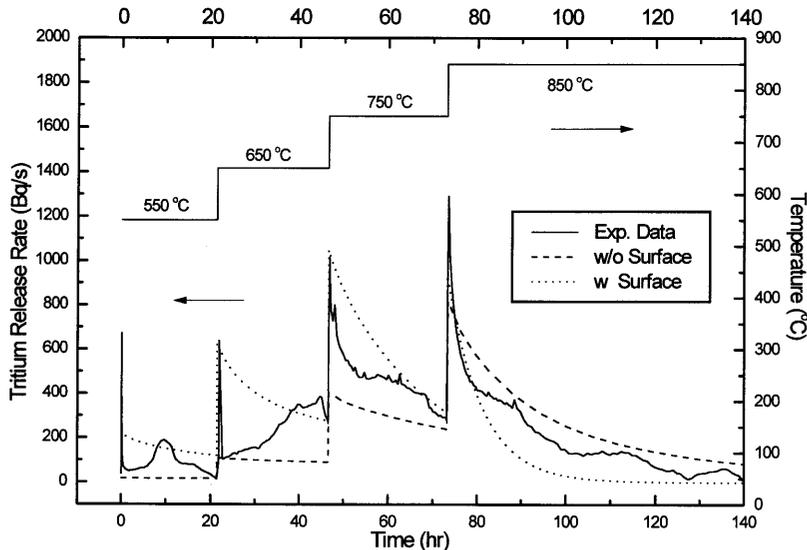


Fig. 4. Comparison of BETTY results with the experimental results for tritium release from SIBELIUS sample 5-1 under four successive temperature anneals.

rear part. The modeling results considering surface coverage show that the peak at 650 and 750°C reaches that of the experimental data but the shape decreases slowly relatively to the experimental data. The modeling results for both sam-

ples 2-1 and 5-1 show similar phenomena with regard to the peak and the overall shape. The experimental data locates between two modeling results. Overall tritium releases fast when the surface coverage is considered, but it releases slowly

when the surface is not considered. A retardation mechanism at relatively low temperature with surface coverage might be needed in the future work.

Since the property data assumed in this analysis are not dominant values, a sensitivity analysis for the variation of surface activation energies are performed. As shown in Fig. 6, the modeling

results for  $E_{\text{ads}} = 60$  kJ/mol are exactly the same as the modeling results for the reference adsorption activation energy, except the results at 850°C. The release rate at 850°C and at the end of 750°C decrease somewhat more sharply. The modeling results, however, have not changed in each case, which shows that the sensitivity of the activation

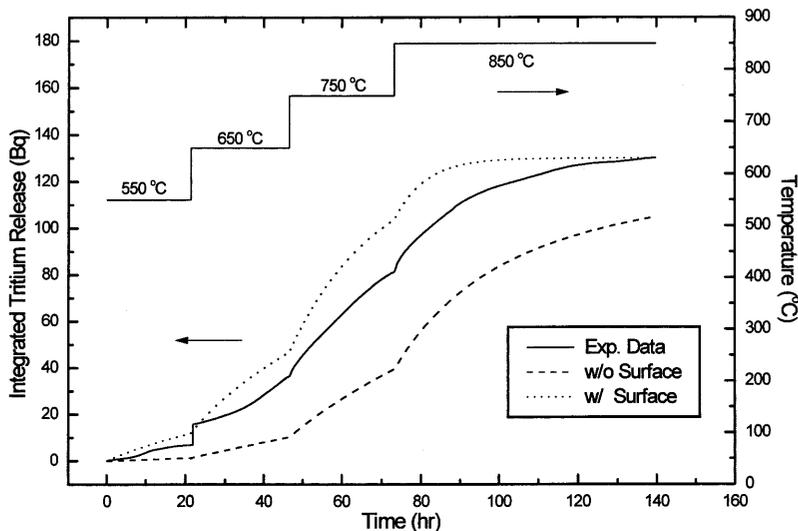


Fig. 5. Comparison of BETTY results with the experimental results for integrated tritium release from SIBELIUS sample 2-1 under four successive temperature anneals.

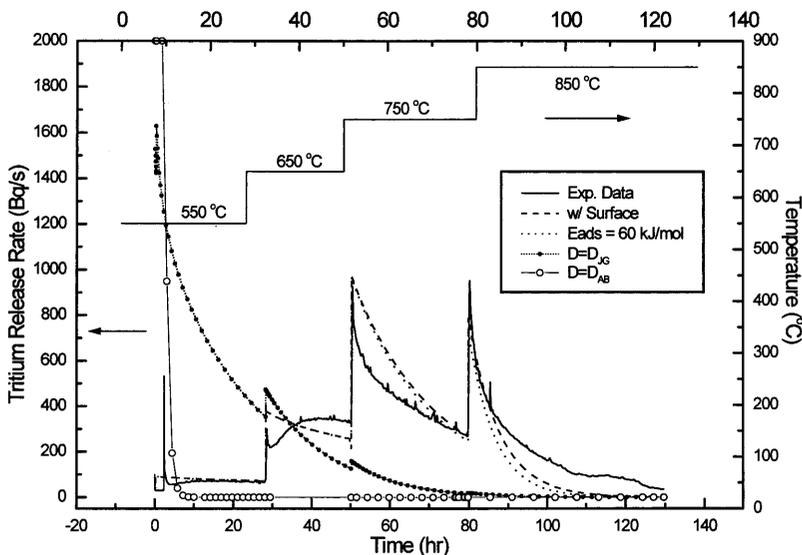


Fig. 6. Effect of adsorption activation energy and diffusion coefficients on the modeling results.

energy is minimal. The modeling results for the diffusion coefficients given by [17,18] are also shown in Fig. 6. Almost all the tritium is released at the low temperatures due to their high value of diffusion coefficient. Therefore, the reference diffusion coefficient used in this analysis is verified to be more reasonable than theirs.

## 5. Conclusions

Among the recently released USA SIBELIUS experimental data for Be samples, sample 2-1 and 5-1 were chosen due to their relatively well-behaved tritium release rate curves and these selected experimental data were analyzed in this work. Also, a simplified model of the developed integrated tritium transport model, BETTY, was described. Based on the limited property data, this simplified model was applied to the SIBELIUS experimental data and the modeling results were compared to the tritium release experimental data. The results show reasonable agreement. Also, the effect of surface activation energies on the modeling results was investigated and found to be minimal. The reference diffusion coefficient used in this analysis is reasonable. Based on this analysis performed in this work, more mechanisms might be needed for the perfect analysis of the experimental data in the future work.

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