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MATHEMATICAL MODELS DESCRIBING GENERATION AND DIFFUSION OF TRITIUM AND HELIUM-4 IN SOLID BREEDERS FOR FUSION REACTORS

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MATHEMATICAL MODELS DESCRIBING GENERATION AND DIFFUSION OF TRITIUM AND HELIUM-4 IN SOLID BREEDERS FOR FUSION REACTORS

Abstract

Several mathematical models, based on spherical geometry, were developed to describe generation, accumulation, and release of ^4He and T_2 in solid breeders for fusion reactors. The applicability of these models was extended to include a collection of particles that have a log-normal distribution. An assessment of the models focused attention on the fact that there is a paucity of data on the diffusion coefficients and solubilities of hydrogen isotopes and helium-4 in solid breeders.

For generation rates greater than 10^{13} atoms/cm³·s, the limiting concentrations of helium-4 and tritium may exceed their solubilities in solid breeders, and bubbles could form. These bubbles could be accommodated by swelling without rupturing. In some cases the forces imparted by trapped gases may be sufficient to rupture the sphere. In other cases new exit paths for these gases, such as grain boundary separations, cracks, or fissures, could be formed.

Introduction

Some conceptual designs of fusion reactors have selected solid breeders such as lithium oxide (Li_2O) as blanket materials.^{1,2} Since there are no natural sources of tritium (T_2), all fusion reactors must breed their own tritium. In order to establish methods for recovering T_2 it is of considerable importance to evaluate transport properties of T_2 and ^4He in solid breeders and structural materials.

In lithium-containing compounds the only nuclear reactions that produce tritium with a high enough cross section to be useful are $^6\text{Li}(n,\alpha)\text{T}$ and $^7\text{Li}(n,n',\alpha)\text{T}$.^{1,2} These two neutron-induced reactions have rates that are about a factor of 10 greater than other possible gas producing nuclear reactions.

Seki¹ has suggested that trapped ^4He in Li_2O could cause swelling. Hickman³ has also stated that prolonged exposure of solid breeders to neutrons may induce swelling and internal void formation that could act as permanent sinks for much larger amounts of tritium than presently expected.

The objective of this study was to establish mathematical formulations that would provide a method for describing the time-dependent accumulation and release of ^4He and T_2 in solid breeders.

Theoretical Considerations

GENERATION AND ACCUMULATION OF HELIUM-4 AND TRITIUM IN A SPHERE

The differential equations that describe heat conduction in a solid sphere with a homogenous heat source were derived by Carslaw and Jaeger.⁴ By assuming that the accumulation and release of neutron-induced gases, such as He and T_2 , are analogous processes, it is a simple matter to obtain transport equations for these gases in the solid.

For a sphere with radius a , the moles of gas retained (Q) at time t , for $t > 0$, with zero initial concentration in the sphere and at the sphere surface is described by⁵:

$$Q = \frac{4\pi A_0 a^5}{45D} \left[1 - \frac{90}{\pi^4} \sum_{n=1}^{\infty} \frac{1}{n^4} \exp(-n^2 D \pi^2 t/a^2) \right], \quad (1)$$

where $D(\text{cm}^2/\text{s})$ is the diffusion coefficient of the gas in the solid, and $A_0(\text{moles}/\text{cm}^3 \cdot \text{s})$ is the generation rate of the gas. At long times ($t \rightarrow \infty$) Eq. (1) becomes $Q = 4\pi A_0 a^5/45D$, and the average concentration becomes $\bar{c}(\text{moles}/\text{cm}^3) = A_0 a^2/15D$.

Equation (1) shows that the limiting quantity that accumulates in the sphere is proportional to the gas generation rate A_0 and to a^5 . If diffusion is the only process by which the generated gases ^4He and T_2 can exit from the solid, then for generation rates greater than $10^3 \text{ atoms}/\text{cm}^3 \cdot \text{s}$ the solubilities of these gases in solids will probably be exceeded and bubbles could form.

In some cases the forces imparted by trapped gases may be sufficient to rupture the sphere. In other cases new exit paths for the gases, such as grain boundary separations, cracks, or fissures could be formed. If the forces imparted by the trapped gases are small, the trapped gases will be accommodated by swelling without rupturing.

For generation rates less than $10^{12} \text{ atoms}/\text{cm}^3 \cdot \text{s}$, the solubilities are not exceeded. For some applications this condition may be desirable for temporarily storing T_2 in the solid.

Equation (1) and others that follow can be applied not only to processes with a continuous generation rate, but also to a process with a pulsed high-frequency gas generation.

RELEASE OF GASES

During exposure to neutrons not only are gases generated in lithium containing solids, but they are also released to the surrounding gases. For ^4He or T_2 , the amount released as a function of time can be derived by balancing the amounts of gas involved. That is, the gas quantity released = gas quantity generated - gas quantity retained by the solid. Therefore, the quantity released Q_R (moles) can be expressed as

$$Q_R = \frac{4}{3} \pi a^3 A_0 t - \frac{4\pi A_0 a^5}{45D} \left[1 - \frac{90}{\pi^4} \sum_{n=1}^{\infty} \frac{1}{n^4} \exp(-n^2 D \pi^2 t / a^2) \right]. \quad (2)$$

At long time ($t \rightarrow \infty$), the quantity generated equals the quantity released; i.e.,

$$Q_R = A_0 t. \quad (3)$$

At early exposure times most of the gases generated are retained by the solid and the release rate increases according to the formulation

$$\frac{dQ_R}{dt} = \frac{4}{3} \pi a^3 A_0 \left[1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp(-n^2 D \pi^2 t / a^2) \right]. \quad (4)$$

A sphere size, temperature, and generation rate could be selected such that the limiting quantity, $4\pi A_0 a^5 / 45D$, retained by the solid could be achieved in a short time and any additional gas generated would go directly to the gas phase. Equation (3) expresses the moles released, Q_R , as a function of time at constant temperature under these conditions.

CONCENTRATION AT SPHERE CENTER

Internal forces tending to rupture the solid are proportional to the sum of ^4He and T_2 concentrations at the sphere center. These concentrations can be described by the equation⁶

$$C \left(\frac{\text{moles}}{\text{cm}^3} \right) = \frac{A_0 a^2}{6D} \left[1 + \frac{12}{\pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \exp(-Dn^2\pi^2 t/a^2) \right] \quad (5)$$

It follows that the limiting concentration for each gas as $t \rightarrow \infty$ becomes $A_0 a^2/6D$.

Because in most fusion reactor applications it is easier to process T_2 in the gas phase, it would be desirable to accumulate most of the ${}^4\text{He}$ and T_2 in the surrounding gas by allowing their limiting concentrations in the solid to be achieved, thereby forcing any additional gases generated to go directly to the gas phase. A knowledge of the maximum deformation that the solid will undergo before rupturing is required to establish limiting concentrations. The extent of deformation can only be experimentally determined.

APPLICABILITY OF DIFFUSION MODEL

The solubilities of T_2 or ${}^4\text{He}$ in lithium-containing solids are not known, but they are expected to be low. It is not clear whether the accumulation or release of these gases can be described by Eqs. (1) and (2) after the solubility is exceeded and bubbles are formed. If no new exit paths are created as the solid deforms, then Eqs. (1) and (2) will probably describe the accumulation and release of gases.

Accumulation of Tritium in Solid by Absorption from Surrounding Gas and by Internal Breeding

It is plausible to assume that negligible amounts of T_2 are absorbed from the surrounding gas during early neutron exposures until limiting concentrations are achieved. In a contained system, after the limiting concentration $A_0 a^2/6D$ is achieved through irradiation, the amount of T_2 surrounding the solid is expected to be enough to allow transfer of T_2 from the surrounding gas to the solid.

The interaction between the two sources of T_2 and Li_2O can be assessed by assuming initial and boundary conditions that apply immediately following an initial neutron exposure. That is, set the conditions such that, for a new neutron exposure the starting concentration of T_2 in the solid is the limiting concentration resulting from a previous neutron exposure. This constraint on the initial concentration can be described by the expression:

$$C(r) = A_0(a^2 - r^2)/6D. \quad (6)$$

In order to continue the derivation, we assumed that transfer of T_2 into and out of the solid can be described by the formulation⁴

$$\frac{dc}{dr} = h(c_b - c_s),$$

where h is a proportionality constant with units of cm^{-1} , c_s is the concentration of T_2 at the sphere surface ($r = a$), and c_b is the concentration of T_2 in the solid. The moles Q of T_2 , which come from T_2 in the surrounding gas and from neutron-induced nuclear reactions, can be described by:⁷

$$Q = \frac{4\pi A_0 a^5}{45D} + \left(SV + \frac{4\pi A_0 a^4}{9hD} \right) \times \left\{ 1 - 6h^2 \sum_{n=1}^{\infty} \frac{\exp(-\alpha_n^2 Dt)}{\alpha_n^2 [a^2 \alpha_n^2 + ah(ah-1)]} \right\}, \quad (7)$$

where α_n is the n th nonzero positive root of

$$A\alpha_n \cot a\alpha_n = 1 - ah, \quad (8)$$

S is the solubility of T_2 in a solid breeder, V is volume of the sphere, and the remaining symbols mean the same as defined above. At long times ($t \rightarrow \infty$), the limiting amount $Q_{\max}$ of T_2 retained by the sphere is

$$Q_{\max} = \frac{4\pi A_0 a^5}{45D} + Q_{\infty} + \frac{4\pi A_0 a^4}{9hD}. \quad (9)$$

Because the value of h can only be experimentally determined, it is not yet possible to estimate $Q_{\max}$ for either ^4He or T_2 .

Particle Size Distribution

IRREGULARLY SHAPED PARTICLES

The analysis described above only applies to one sphere or to many spheres of the same size. For practical applications such an ideal situation will not exist, but more likely will consist of a size distribution of irregularly shaped particles.

For many regularly shaped particles the surface-to-volume ratio varies as k/r , where k is a constant approximately equal to 3 and r is a dimension such as radius or half-thickness. For small particles deviations from spherical shape could be adjusted by applying a surface-to-volume shape correction so that an irregularly shaped particle could be represented as having a regular shape such as a sphere.

LOG-NORMAL DISTRIBUTION

The log-normal distribution serves as an excellent model for particle size distribution analysis.⁸ It provides a convenient method for describing a dispersed particulate system with only two parameters.

The Normal density function is defined by:

$$\frac{dn}{N} = \frac{1}{\sqrt{2\pi}} e^{-\frac{x^2}{2}} dx, \quad (10)$$

where $x = (\bar{r} - r)/\sigma$, r is particle radius, $\bar{r}$ is the arithmetic mean radius, σ is the standard deviation and N is the total number of particles. It is recognized⁸ that the majority of dispersed systems tend to follow a logarithmic variant of the normal density function. If Z is defined as $Z = (\overline{\ln r} - \ln r_i)/\sigma$ where $\overline{\ln r} = (\sum \ln r_i)/\sum 1$, by making the substitution $\ln \bar{r} = \overline{\ln r}$ ($\bar{r} \neq \bar{r}$), Z becomes $(\ln \bar{r}/r_i)/\sigma$ resulting in a working form of the log-normal distribution,

$$\frac{dn}{N} = \frac{1}{\sqrt{2\pi}} \exp(-Z^2/2) dZ \quad (\ln = \log_e). \quad (11)$$

The area of an individual sphere is $A = 4\pi(r_1)^2 = 4\pi(\bar{y})^2 \exp(-2\sigma Z)$ and the total area of a powder can be expressed as:

$$A = \frac{4\pi N(\bar{y})^2}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} \exp\left(-\frac{Z^2}{2} - 2\sigma Z\right) dZ, \quad (12)$$

which integrates to:

$$A = 4\pi N(\bar{y})^2 \exp(-2\sigma^2), \quad (13).$$

Similarly, the total volume is:

$$V = \frac{4}{3} \frac{\pi N}{\sqrt{2\pi}} (\bar{y})^3 \int_{-\infty}^{+\infty} \exp\left(-3\sigma Z - \frac{Z^2}{2}\right) dZ, \quad (14)$$

which results in:

$$V = \frac{4}{3} \pi N (\bar{y})^3 \exp\left(\frac{9}{2} \sigma^2\right). \quad (15)$$

The surface-to-volume ratio is uniquely defined with two parameters σ and $\bar{y}$ as follows: $A/V = 3/\bar{y} \exp(-5/2 \sigma^2)$.

We can extend the analysis from one sphere to include all spheres by superimposing the log-normal distribution to Eqs. (1), (5), or (6). For example, by combining Eqs. (1) and (9) the average concentration $\bar{C}$ in all spheres having a log-normal distribution can be described by the expression:

$$\bar{C} = \frac{A_0(\bar{y})^2 N e^{2\sigma^2}}{5 \cdot 2\pi D} \int_{-\infty}^{+\infty} e^{-\frac{1}{2}(x + 2\sigma)^2} \times \left[1 - \sum_{n=1}^{\infty} \frac{1}{n} e^{-n^2 D \pi^2 t / N(\bar{y})^2 e^{2\sigma^2} e^{-\frac{1}{2}(x + 2\sigma)^2}} \right] dx. \quad (16)$$

A complete particle size analysis would be necessary to determine the empirical parameters $\bar{y}$ and σ . The expected behavior of spherical particles should not be large for nonspherical particles of comparable dimensions. Large deviations from spherical geometry, which might occur from extreme irregularities, would be manifested as errors in the average dimension and would change the empirical determination of the diffusion constant D .

Limitations of Models

It is reasonable to question the validity of the mathematical models used for these assessments. It is unrealistic to believe that the initial and boundary conditions used for obtaining the various solutions rigidly apply to fusion reactors. These conditions and the resulting equations are only intended to represent approximate conditions in a fusion reactor, but are used because they simplify the problems sufficiently so that the appropriate differential equations can be analytically solved.

Since long-time extrapolations from the models are not strongly dependent on the geometry based as a frame of reference, the limiting values are useful for any regular geometry. On the other hand, the limiting values are strongly dependent on the initial conditions assumed. A better definition of the conditions in a fusion reactor are necessary to refine the mathematical descriptions and to allow a more realistic representation of the time dependence and limiting values.

Conclusions and Recommendations

An assessment of some problems associated with tritium and helium-4 generation and accumulation in solids as blanket materials can be made by using spherical geometry as a frame of reference for Li_2O particles. The evaluation can be conveniently extended to include a distribution of particles that can be adequately represented by a log-normal distribution.

Perhaps the most useful part of the study is that it focuses attention on some areas where no experimental information is available. We can only provide information of lasting value by experimentally determining the appropriate constants S and D . The following experiments are listed in order to delineate those areas where experimental work is sorely needed.

- Determine solubility and diffusion constants for T_2 and He and mixtures of both in solid breeders.
- Determine the accumulation rates of T_2 and 4He in solid breeders when both gases are generated in the solid as well as absorbed from the gas phase. This experiment would be done by exposing in the temperature range 1000-1300°K, a solid breeder to neutrons.
- Determine the amount of deformation that a solid breeder can undergo before rupturing by exposing it to neutrons and allowing gas bubbles of T_2 and 4He to form.
- Use techniques such as transmission electron microscopy and nuclear magnetic resonance to study bubble formation.
- Study the release mechanisms and how they are affected by deformation of the solid.
- Determine an optimum particle size that will provide effective gas release and minimize the T_2 inventory.

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