Grain size influence on the release of radioactive isotopes out of target materials made of powder.

V. Kuchi[1]* and P. Jardin[1]

[1]Grand Accélérateur National d'Ions Lourds, Bvd H. Becquerel, BP 55027, F-14076 Caen cedex 5

*Kuchi.venkateswarlu@ganil.fr

Radioactive ion beam production by Isotope Separator On Line method (ISOL) has motivated the construction of several nuclear facilities over the world. The method consists in impinging solid target material with beams of stable nucleus. Radioactive nuclei produced during the collision are stopped in the target material and must diffuse out of it as fast as possible to transform them into ions before their radioactive decay. The release time must thus be as short as possible to avoid their losses. The release of the nuclei depends on several parameters, which are related to the chemistry of the atoms in the target matrix, to the geometry and micro-structure of the target, and to its temperature. In the case of targets made of grains, we assumed that an optimum grain size of the grains existed. To make possible its easy determination, we aimed to calculate it analytically. Thus we have built a description of the propagation of the atoms in the target material, while conserving the different physico-chemical parameters and avoiding the use of adjustable parameters. The description of the propagation process will be presented as well as the assumptions. Finally, the optimum grain size will be given for the radioactive ^{35}Ar atoms out of graphite.

INTRODUCTION

To explore the synthesis of the elements which form our world, the nuclear matter must be studied to understand how, from initial very hot plasma of relatively simple particles the nuclear processes have led to current complex and heavy elements. Using a large variety of nuclear reaction, the initial plasma has transformed into stable elements, which are the starting point of the "archeological nuclear studies". "Exotic" isotopes of elements, so called as they no longer exist on the earth must be produced to be observed before their transformation by radioactive decay up to the valley of stability. One experimental principle consists in bombarding targets of matter (made of stable nuclei) with ions (also made of stable nuclei). Above energy of some Mega Electron volt per nucleon to overcome the coulombienne barrier, the collisions are violent enough to modify the structure of each collision partners and lead to exotic nuclei. The exoticism of the nuclei is evaluated looking at their position in the chart of nuclides, which depends on their proton and neutron numbers, and where very exotic nuclei show an important number disequilibrium. Moreover, the more exotic, the more interesting they are as theoretical descriptions can be tested far from the region where they were born. The demands of physicists for these exotic nuclei is important and several facilities have been built over the last 40 years all over the world [1] to deliver beams of radioactive nuclei. Today's main facilities are ISOLDE at CERN [2] (Switzerland), ISAC at TRIUMF [3] (Canada), SPIRAL-1 at GANIL [4] (France), GSI [5] (Germany), RIKEN at RIBF [6] (Japan), NSCL at MSU [7] (USA) and JINR at FLNR [8] (Dubna).

Two main methods are commonly used [9]. For both, one figure of merit of the beams is the rate of radioactive ion beams (RIB), which must be high enough to fulfill the statistic requirements of the experiment analysis. The rate depends on two main parameters: the rate of nuclear reactions during the collision between the bombarding beam and the target, and the efficiency of the setup to deliver the

nuclei of interest to the physicist. In the frame of this work, only the Isotope Separator On Line (ISOL) Method is considered. It consists in a primary beam bombarding a solid target, thick enough to stop the reaction products. Once stopped, they are neutralized and diffuse out the matter of the target, effuse up to a "source" where they are ionized before being accelerated to form a radio-active ion beam.

- The rate of nuclei produced during the collision depends on the intensity and energy of the "primary" beam, and on the matter of the target.
- The efficiency depends on the technique used and on its response time (time between the creation of the nuclei and their exit from the production system), which is in competition with the radioactive decay time of the nuclei of interest.

The design of the production system is thus a compromise between different constraints the production system has to cope with I) - maximizing the reaction rate, which often induces an enlargement of the production system (higher primary beam intensity, larger target to sustain the primary beam power), II) - minimizing the response time (strongly related to the size of the system), what is of first importance in the case of short-lived isotope production.

The atom-to-ion transformation process can be described using three different sub-processes: release of the atoms out of the target matter, effusion up to the ion source and ionization. From the time point of view, other processes like nuclear reaction and acceleration of the ions are neglected. Among the processes considered, the release time of the atoms out of the target matter is often dominating. Its determination needs to take into account the physico-chemical properties of the couple formed by the radioactive atom and the target matter, the macro and micro structure of the target and the temperature, which must be maximal to speed up the release while low enough to avoid the target damage.

Targets made of compressed and sintered powders, as graphite, have been widely used in ISOL facilities. So far, GANIL/SPIRAL1 (Grand Accélérateur National d'Ions Lourds/Système de Production d'Ions Radioactifs Accélérés en Ligne phase 1) facility has been producing radioactive ions exclusively by fragmenting the nuclei of different primary beams on carbon nuclei of graphite targets, explaining our interest for such target material. Diffusion of atoms out of the grains of graphite and effusion within the spaces between the grains are two important processes, but their respective weights on the release are difficult to separate. That is why the release process out of the target is often described using a global "diffusion" coefficient, mixing both diffusion and effusion processes. Several attempts and receipts have been performed and used to optimize the release according to the microscopic structure, playing on the density and on the grain size. Obvious conclusions about the grain size, which must be as small as possible, and about the porosity, which must be as open as possible, have been extracted. But are these conclusions sufficient to optimize a target? If one considers a given material density, reducing the size of the grains will lead to increase the complexity of the path the atoms have to go through to leave the target material. If the density is not fixed, one can imagine a very low density target material, possibly increasing the effusion process. But lower density induces larger target, and lower release out of the target vacuum chamber. So for a given radioactive isotope, what is the optimum grain size?

This work is part of a global study aiming to improve the efficiency of radioactive ion beam production systems [10]. This part aims to give an analytical description of the release out of a target made of powder and to extract an optimum grain size, avoiding adjustable parameters and computing simulation to conserve usable equations and give trends. It does not pretend to provide absolute values of grain size, which therefore will have to be compared to experimental results. Assumptions are given to let the reader appreciate the reliability of the results.

Description of release process

Our approach is based on the following schematic Figure 1. We consider a target made of grains, having a spherical average shape. Two cases are considered Figure 1: on the left part, grains have a

radius two times larger than in the right part. The atoms diffuse out of the grains, effuse up to the surface of the target and once they reach the left or the right side, they are released out of the target.

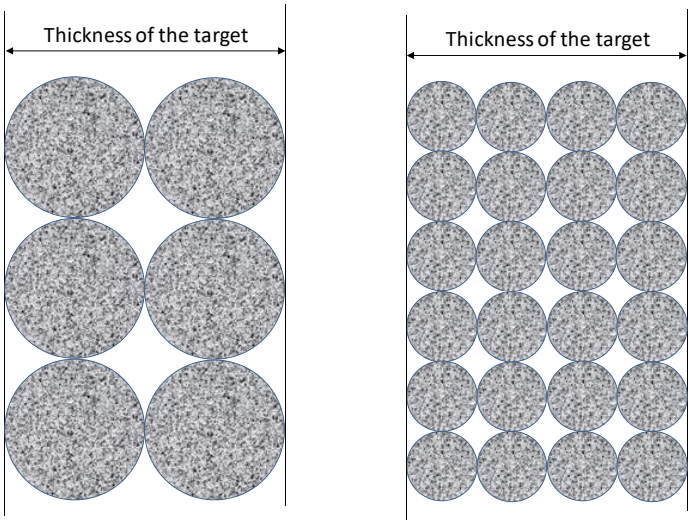

Figure 1 : Basic representation of the graphite structure.

In the left part, grains are larger. The diffusion time out of the grains will thus be longer than in smaller grains. In case of effusion, the process flow two steps. One is the Effusion out of an elementary volume formed by a group of 8 grains and the second is the effusion out of target through the sum of elementary volumes. If the grains are smaller, the effusion out of an elementary volume is smaller but the effusion out of the target is longer since the number of elementary volumes increases with decrease of grain size. So one could think that optimizing the release of the atoms out of the target could merely consists in reducing the size of the grains. But to reach the outer surface of the target, the atoms also have to go through a series of elementary volumes, whom number increases as the grain size decreases. Diffusion and global effusion processes have thus opposite dependences versus the grain size. **The question we would like to address is to determine the optimum size of the grains?**

Therefore we built an analytical description taking these two processes into account, while conserving the presence of the grain size in the equations. Our goal was to build a description which could directly extract the tendencies of the effect of the grain size on the release behavior, and a rough estimation of the optimum grain size.

We did the following assumption:

1. The temperature is uniform over the target volume. Rk: targets and their containers are designed for that but the power deposited by the primary beam can induce temperature differences up to some hundreds of Kelvin, to be compared to temperature of the order of 2300 K.
2. The production of radioactive atoms in the matter of the target is uniform. Rk: the ions of the primary beam can be slow down in the target material, modifying their energy and thus the nuclear reaction cross section along their path in the target material
3. The diffusion of the atoms within the matter of the grains is isotropic.
4. The effusion regime of the atoms in free volume in the matter is molecular (no collision between the atoms).
5. The extend of the target in the directions perpendicular to the thickness is infinite, so the flux of atoms along these directions is equal to zero
6. Once the atoms are out of a grain, they will never come back into the grains, as the propagation in the grain is more difficult than in the free volume between the grains. Rk:

this effect can be neglected only if the diffusion coefficient in the matter can be neglected in front of the effusion coefficient.

7. Once the atoms are out of the target, they will never come back into the target. Rk: this assumption is valid if the effusion rate out of the production system is high enough compared to the effusion rate in the matter.

As diffusion-effusion-exit processes are consecutive, the flux of atoms released by the grains corresponds to the feeding term of the effusion process, and the flux of atoms released by effusion corresponds to the flux released out of the target. Combining the expression of the release out of a sphere (diffusion) and the expression of the release out of a foil (effusion), both extracted from the article of M. FUJIOKA [11], we can build the analytical expression of the flux $F(t)$ coming out the target at time t:

$$\boldsymbol{F(t) = \ 48.\lambda_D.\lambda_E.N_0 \sum_{n=0}^{\infty} \sum_{m=1}^{\infty} \frac{1}{\lambda_{En}-\lambda_{Dm}} \left[e^{-\lambda_{Dm}\, t} - e^{-\lambda_{En}\, t}\right]} \qquad (1)$$

Where N_0 is the number of atoms produced at $t = 0$ in the target. λ_D is the diffusion rate equal to $D/4R^2$, where D is the diffusion coefficient and R the grain radius. λ_{Dm} is the diffusion rate from the shells m according to the mathematical formalism given in ref [11], and is equal to $m^2\pi^2\lambda_D$. λ_E is the effusion rate equal to $D_E/4d^2$, where D_E is the effusion coefficient and d is the thickness of the target. λ_{En} is the effusion rate of the atoms out of the shell n and is defined as $(2n+1)^2\pi^2\lambda_E$.

If the radioactive decay rate λ is taken into account during the diffusion and effusion processes, the total release rate of the radioactive elements out of the target can be expressed as:

$$\boldsymbol{F(t) = \ 48.\lambda_D.\lambda_E.N_0.e^{-\lambda t} \sum_{n=0}^{\infty} \sum_{m=1}^{\infty} \frac{1}{\lambda_{En}-\lambda_{Dm}} \left[e^{-\lambda_{Dm}\, t} - e^{-\lambda_{En}\, t}\right]} \qquad (2)$$

To optimize the grain size, one criterion can be the mean release time, which depends on the diffusion and effusion rates and thus on the radius of the grains:

$$< t >= \frac{\sum_{m=1}^{\infty} \sum_{n=0}^{\infty} \frac{\lambda_{En}+\lambda_{Dm}+2\lambda}{(\lambda_{Dm}+\lambda)^2(\lambda_{En}+\lambda)^2}}{\sum_{m=1}^{\infty} \sum_{n=0}^{\infty} \frac{1}{(\lambda_{Dm}+\lambda)(\lambda_{En}+\lambda)}} \qquad (3)$$

Determination of the effusion coefficient

As for a diffusion coefficient D, the effusion coefficient D_E can be determined considering it as an intensive value, which does not depend on the size of the target. Nevertheless, these coefficients are statistical coefficients, needing a sufficiently large media. So there are minimum volumes within which these coefficients have no meaning. In the framework of our approach, the minimum size considered to define the effusion coefficient D_E is the size of a cube made of eight grains. We consider that atoms propagate in the target by effusion, from one inter-grain volume to another one through the aperture defined by the spheres. For that elementary system, the effusion rate λ_E can be calculated by making the ratio of the inter-grain volume V by the conductance C between inter-grain volumes, what leads to:

$$\lambda_E = \frac{V}{C} = \frac{\gamma.\beta}{R} = \frac{D_E}{4R^2}$$

where γ is a coefficient fixed by the geometry of the elementary system of eight spheres, and β is the average speed of the atoms $\sqrt{\frac{8kT}{\pi m}}$ having a mass m and at temperature T. We can then deduce a rough

effusion coefficient D_E from the geometry of the microscopic structure and from the speed of the atoms, versus the grain size

$$D_E = 4\gamma\beta R$$

For ^{35}Ar atoms diffusion in spheres of 1 µm in diameter, and a density of 1.8 g/cm^3, D_E is equal to 0.693 cm^2/s.
For a target thickness d, the effusion rate is then equal to

$$\lambda_E = 4\gamma\beta R/d^2$$

Extraction of the diffusion coefficient

We know that the diffusion coefficient varies with materials and temperature. We have extracted the diffusion coefficients of different metallic materials from G.J. Beyer [12]. They all range from 10^{-13} to 10^{-5} cm^2.s^{-1} in a temperature range of 900 K to 2200 K. We assume that the diffusion coefficient of atoms in the grains of graphite is within this range.

In the frame of C. Eléon's work, effusion and diffusion processes in the matter were not separated. Only a global release coefficient was extracted from the experiment. In the present work, effusion and diffusion processes in the matter are separated. Whatever is the description of the release, using separated or non-separated process description, the release efficiencies must be the same:

$$\boldsymbol{\varepsilon_{global}}\big|_{\boldsymbol{C.Eléon}} = \boldsymbol{\varepsilon_{Diffusion}}\cdot\boldsymbol{\varepsilon_{Effusion}}\big|_{\boldsymbol{present\ work}} \qquad (4)$$

The effusion coefficient and thus the effusion efficiency can be deduced from the previous paragraph. The diffusion coefficient has been extracted from the above equation, by adjusting the diffusion efficiency up to get equality between the efficiencies. The corresponding diffusion coefficient has been found equal to $1.8.10^{-8}$ cm^2.s^{-1}.

Density effect

A graphite density of 1.8 g/cm^3 can not correspond to the arrangement of the spheres given Figure 1, where the spheres are in punctual contact with the other spheres, leading to a density close to 1.2 g/cm^3. The size of the grains being selected by a sieve during the synthesis process of the material, grains smaller than the sieve mesh are also collected and contribute to the final material density, what explains this high density of 1.8 g/cm^3. To take the presence of this extra-material into account in our description, we assumed it equivalent to an overlap between the spheres. This overlap has a direct consequence on the effusion and diffusion coefficients: the inter-grain volumes are smaller, the apertures between the inter-grain volumes are narrowed and the surface of the sphere trough which the atoms can leave the grains is reduced. These consequences of the overlap lead to a reduction of the effusion and diffusion coefficients by factors of 0.6 and 0.55 respectively. These factors have been taken into account in the present article.

Optimum grain size

The optimum grain size is defined as the size corresponding to the minimum value of the release mean time. The mean release time is necessarily related to the half-life of the isotope considered, but the minimum mean release time only depends on a balance between the diffusion and effusion processes.**Fehler! Verweisquelle konnte nicht gefunden werden.** Figure 2 shows the evolution of the release mean time versus the grain size of graphite in which ^{35}Ar isotopes diffuse and effuse. The conditions of the calculation correspond to C. Eléon [13] work, *i.e.* a graphite having a density of 1.8 g/cm^3, diameter grains of 1 µm, thickness of 3 mm and at a temperature of 2100 K. The mean release time drops approximately to ~ 25 ms for a grain radius of 0.3 µm.

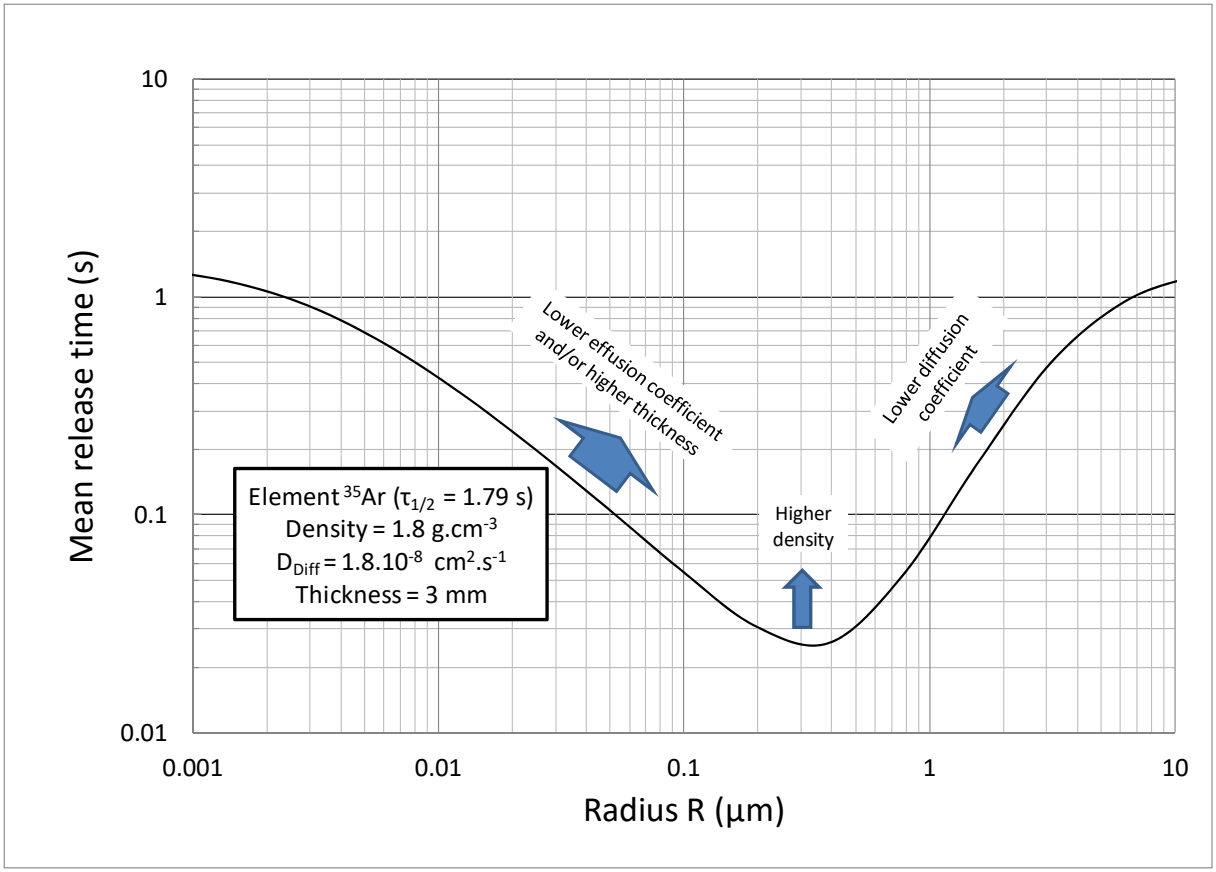

Figure 2 : influence of grain size on mean release time

For larger grains or smaller diffusion coefficient, the diffusion process increases the release time. For smaller grains or higher diffusion coefficient, the diffusion process becomes negligible compared to the effusion process which thus dominates. Even if this description does not pretend to provide an absolute value of the optimum grain size, it gives trends and shows there is no point in looking for grains smaller than an optimum, what is technically often difficult leads to the risk of increasing the release time and thus the production losses.

Release efficiency versus radius and density

As the processes of diffusion of the atoms out of the grains and effusion through the inter-grain volumes are considered consecutive, the release efficiency can be calculated by the product of the diffusion efficiency ε_{diff} by the effusion efficiency ε_{eff}:

$$\varepsilon = \varepsilon_{Diffusion} \cdot \varepsilon_{Effusion} = 3.\frac{\lambda_D}{\lambda}\left(\sqrt{\frac{\lambda}{\lambda_D}}.coth\sqrt{\frac{\lambda}{\lambda_D}} - 1\right).2\sqrt{\frac{\lambda_E}{\lambda}}.tanh\left[\frac{1}{2}\sqrt{\frac{\lambda}{\lambda_E}}\right]$$

For ^{35}Ar isotopes, the release efficiency as a function of the grain size and the density is given in Figure 3.

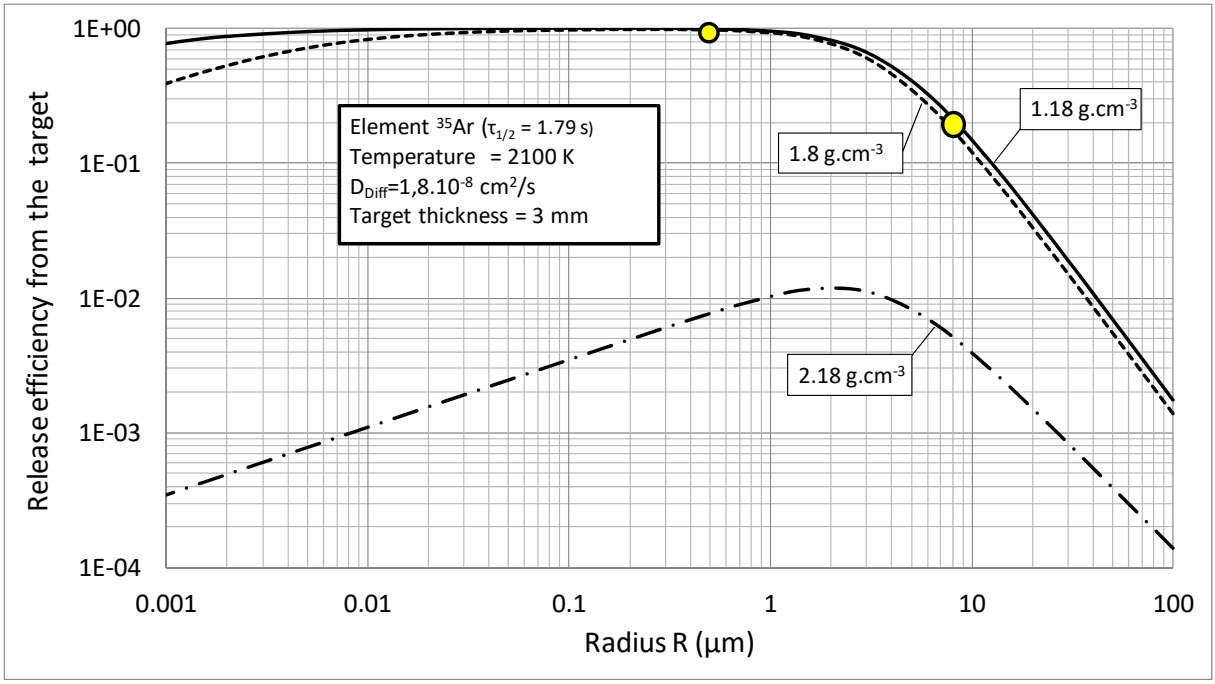

Figure 3 : Release efficiency versus grain size and three different densities. Yellow dots correspond to experimental results.

The 1.18 g/cm^3 density assumes no overlap between the grains. A density of 1.8 g/cm^3 corresponds to a material previously tested and assumes an overlap between the grains. A density of 2.18 g/cm^3 corresponds to a density close to the maximum.

For grain size ranging from 50 nm to 1 µm and a density equal to 1.18 g/cm^3, the release efficiency is close to 100%. In this range, the efficiency difference between 1.8 g/cm^3 and 1.18 g/cm^3 densities is not significant. So there would be no point in looking for too low density materials, which are often more difficult to synthesis, which lead to larger target volumes and from which the energy deposited by the primary beam is more difficult to remove.

As shown in the figure, decreasing the density from 2.18 g/cm^3 to 1.18 g/cm^3 leads to increase the release efficiency by two orders of magnitude for 1µm grains. The effect of the density is more important on the effusion than on the diffusion, as the difference of efficiency is more important for small grain region, where the effusion mainly governs the release time.

One conclusion which can be extracted from this observation is that if thick targets aim to produce short lived isotopes, it is of first importance to develop target structure made of very small parts of matter, separated by sufficiently large and open spaces, which principle corresponds to a large open porosity.

Comparison with experiment

A previous experiment has been performed to compare the release efficiency of ^{35}Ar atoms out of graphite having different grain size but same density (1.8 g.cm^{-3}). Ignoring *a priori* the diffusion coefficient of Ar atoms in the carbon grains, we have adjusted the diffusion coefficient to make the global efficiency of our description correspond to the one observed during the experiment (0.98 for 1µm grain and a target thickness of 3 mm). The diffusion coefficient deduced was equal to 1.8.10^{-8} cm^2/s. Then it has been used to calculate the mean release time and the release efficiencies presented in Figure 2 and Figure 3. Going from 0.5 µm to 8 µm radii, the experimental release efficiency dropped from 0.98 to 0.22. This efficiency is close to the one predicted with our description (0.18).

Outlook

This comparison is indeed not sufficient to validate our description. More results would be necessary for a wide range of grain size. And to use them, they must have been obtained in similar experimental conditions. A dedicated system could be helpful.

So far the study has been led in the relatively simple case of a noble gas isotope, avoiding the effects of chemical reactions. Continuing the study with reactive elements will be interesting and probably more difficult as each reactive element has a particular behavior in the matter and on its surface.

A complete description allowing a stringent design of the target seems to be an unreachable goal as additional effects should be taken into account, one of the most important being the damage of the material under irradiation, which changes the characteristics of the material along the production process. This effect is presently far from being mastered. Awareness of trends, orders of magnitudes, existence of mechanisms having sometimes opposite effects can help to optimize TISS, particularly if one wants to explore regions of the nuclide chart where half-lives are of the order of some milliseconds.

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