



**HAL**  
open science

## **A study of diffusion coefficients: database and new device**

Alexis Ribet, Pascal Jardin, Mathieu Lalande

### ► **To cite this version:**

Alexis Ribet, Pascal Jardin, Mathieu Lalande. A study of diffusion coefficients: database and new device. Defect and Diffusion Forum, 2025, 439, pp.139-145. <10.4028/p-u7wKGE>. <hal-04965225>

**HAL Id: hal-04965225**

**<https://hal.science/hal-04965225v1>**

Submitted on 20 May 2025

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



HAL Authorization

# A study of diffusion coefficients: database and new device

A. Ribet<sup>1, a\*</sup>, P. Jardin<sup>1, b</sup>, M. Lalande<sup>1, c</sup>

<sup>1</sup>Grand Accélérateur National d'Ions Lourds, 14076 Caen, France

<sup>a</sup>alexis.ribet@ganil.fr, <sup>b</sup>pascal.jardin@ganil.fr, <sup>c</sup>mathieu.lalande@ganil.fr

**Keywords:** TULIP project, ISOL, diffusion coefficients, database, TARGISOL, MELODICA system

**Abstract.** The aim of the TULIP (Target ion soURce for short-Lived Isotope Production) device is dedicated to the production of short-lived neutron deficient ions using ISOL (Isotope Separator OnLine) method. The efficiency of such device depends on several parameters: diffusion, effusion and ionization processes, which must be optimized to maximize the efficiency. As short-lived, the elements must release from the stopping material in a brief time, explaining our focus on the diffusion parameters and our effort to build a database gathering diffusion parameters concerning a large variety of target-element combinations. For this purpose, important bibliographical work is underway to collect as much data as possible. Another offshoot of the TULIP ISOL project is to build a system (MELODICA device, MEsure en Ligne de cOefficients de Diffusion et de temps de Collage Atomique) to systematically measure the diffusion coefficients of atoms out of materials at elevated temperatures. Moreover, to get rid of constraints related to the use of radioactive elements, the MELODICA device uses stable atoms and embeds a FEBIAD (Forced Electron Beam Induced Arc Discharge) source to ionize a large panel of elements. With this device, it will be possible to systematically extract several diffusion coefficients during the same experimental period, and to compare the relative release of different atoms out of materials currently used in the ISOL systems.

## Introduction

Diffusion phenomena occupy a prominent place in material research since decades ago as this process can influence materials properties (electronic, mechanical, optical, ...). The knowledge and understanding of this process is hence a way to enhance properties, to obtain solutions of different metallurgical problems [1].

In ISOL (Isotope Separator OnLine) systems, radioactive atoms must leave the target material before decaying. Their release is often stimulated by heating the material up to temperatures of the order of 2000°C. Despite an impressive amount of experimental data on diffusion coefficients present in literature, they are not always sufficient to predict release time of atoms out of ISOL targets, especially at elevated temperatures. Our aim is thus to collect data particularly useful to ISOL system

designers, make them available and eventually extract global rules about the diffusion behavior, which could fill some gaps in experimental data.

Diffusion can be described as a process of migration and mixing particles via their irregular movements enhanced by kinetic energy input, while considering chemical equilibrium and native defect composition of material (interstitials, vacancies). Furthermore, diffusion phenomena depend on the size and the nature of diffusing atom, the structure, the density and the presence of porosity and impurity in the host material.

The analytical description of the transport of atoms in solids is identical to the one used for the transport of heat in solids. The atom flux in a particular direction is directly proportional to the atomic density gradient in that direction, as the thermal flux in a particular direction is directly proportional to the temperature gradient. For a one-dimensional system, the diffusion current per area unit of species,  $J$  (in particles.  $m^{-2}. s^{-1}$ ), is related to the concentration gradient ( $\frac{\delta c}{\delta x}$ ) of the species by Fick's first law:

$$J = -D \frac{\delta c}{\delta x}, \quad (1)$$

which defines the diffusion coefficient  $D$  of the species ( $m^2. s^{-1}$ ). Minus sign indicates that the flux of particles goes toward low concentration regions.

In addition, the temperature dependence of the diffusion coefficient  $D$  is described in the form:

$$D = D_0 \exp\left(\frac{-Q}{kT}\right), \quad (2)$$

where  $D_0$  is the pre-factor ( $m^2. s^{-1}$ ) and  $Q$  the activation energy for the atomic jump mechanism (in  $J. mol^{-1}$ ), which is often assumed to be independent of the temperature. The coefficient  $D$  is usually represented in the Arrhenius form:

$$\ln D = f\left(\frac{1}{T}\right), \quad (3)$$

from which the pre-factor and the activation energy coefficients can be determined by a linear fit.

Diffusion coefficients are often measured with the aid of radiotracers. A wide variety of radioisotopes with high purity and high specific activity exist as tracers in diffusion studies. Radiotracers are deposited on the material surface. The material is heated to enhance the diffusion process, and the surface is then analyzed.

Different destructive experimental techniques are used to study diffusion behavior, depending on required thickness and accuracy. Material surface is usually analyzed using IBS (Ion-Beam

Sputtering) or SIMS (Secondary Ion Mass Spectrometry). Several thin layers sectioned beforehand are studied. A depth profile can then be determined by counting the ejected radiotracers for each layer. Other techniques are also available to measure residual activity or radioactive decay directly. Nonetheless, these destructive methods are more time-consuming and the values obtained are less accurate [2].

Some experimentalists measure diffusion coefficients with non-destructive methods, *i.e.*, without analyses requiring layer sectioning. Several experimental techniques are available such as PIXE (Proton Induced X-ray Emission), RBS (Rutherford BackScattering), ERDA (Elastic Recoil Detection Analysis), NRA (Nuclear Reaction Analysis) and XRD (X-Ray Diffraction), which use ion beams and X-rays. Other techniques also exist for specific materials: EMPA (Electron MicroProbe Analysis), SRP (Spreading Resistance Profiling) or ionic conductivity.

All these analysis techniques are called direct methods since the diffusion coefficient is “directly” extracted from the experiment. Additionally, several so-called indirect methods are used, including nuclear analyses such as NMR (Nuclear Magnetic Resonance), QMS (Quasi-elastic Mossbauer Spectroscopy) and QNS (Quasi-elastic Neutron Scattering), which use different formalisms to extract the diffusion coefficients. In Fig. 1, the range where diffusion coefficients have been obtained by different experimental methods (direct and indirect) are presented [3]. Moreover, it shows the difference between techniques using or not radiotracers: a wide range of  $D$  parameters (order of magnitude) can be covered with the aid of radiotracers whereas a thin range of  $D$  is usually covered by each technique using no radiotracers. Nonetheless, all these techniques permit diffusion measurements over a broad range of magnitudes and provide complementary and comparable values.

Diffusion data are mostly based on experiments using tracers. When experimental data are not available, diffusion coefficients are evaluated using theoretical models. Aside from comparison with experimental measures, this process allows us to obtain an order of magnitude of coefficients about uncommon target-element combinations. Despite this, the experimental and calculated data available in literature do not cover all the materials which can be used in ISOL systems.

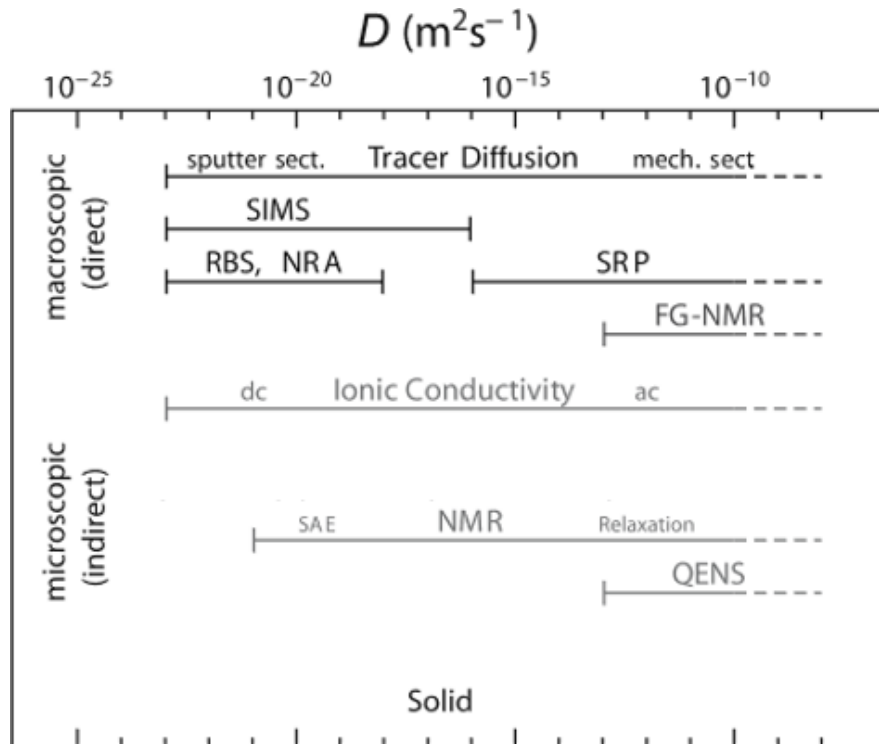


Figure 1. Accuracy of diffusion coefficients values according to the experimental methods (simplified figure from [3]).

It is noteworthy that measures of diffusion coefficients with radiotracers are always performed in two steps: implantation and then analyses. Each material sample requires several preparation steps. In addition, materials are evaluated during separate experiments, with different analysis methods, in different laboratories, by different people. In these conditions, the experimental process must be clearly defined and rigorously followed to make the results comparable. To make them easily comparable and later help the material users in their choices, it would be preferable to extract results from experiments conducted in conditions as close as possible, *i.e.*, with the same device and during the same experimental run. These ideal conditions are difficult to fulfil. Extracting diffusion coefficients from running gathering several materials assessed with the same device would be a first interesting step.

### Diffusion coefficients database

The TULIP (Target ion soURce for short-Lived Isotope Production) project [4,5] aims at producing short-lived neutron-deficient radioactive ions using the ISOL method. Due to the competition between the time of the atom-to-ion transformation process and the lifetime of the radioactive nuclei, it is necessary to minimize the time of each sub-process, *i.e.*, diffusion of atoms from the target material, effusion in the vacuum chamber and ionization. To guide future experimental developments

in terms of materials present in the production system and so in terms of time of the processes, the availability of a diffusion feature database is necessary.

Such a database was already conducted twenty years ago from extensive research in literature and from new measurements performed at CERN (Conseil Européen pour la Recherche Nucléaire) on ISOLDE (Isotope Separator OnLine DEvice) facility and at GANIL (Grand Accélérateur National d'Ions Lourds). The TargISOL database, called DifEfIsol, consisted of a set of tables gathering the process and delay parameters affecting the device design of rare isotope beam production systems [6,7]. Unfortunately, this database is no longer available at the web link. To rebuild it, articles of authors who have reviewed such experimental data on diffusion coefficients are still available [2,8-10]. Nevertheless, despite the important quantity of existing data, the available diffusion coefficients in these tables concern specific material groups. To overcome the lack of data grouping, we are in the process of designing our own database.

Usually, self-diffusion, impurity diffusion (also named heterodiffusion) and interdiffusion are the kind of data reported in literature. Self-diffusion refers to ion diffusion in a material with identical composition (*e.g.*, diffusion of  $^{59}\text{Fe}$  ion in pure iron material). When composition differs, diffusing ion is considered as an impurity (*e.g.*, diffusion of  $^{64}\text{Cu}$  ion in pure aluminum material). Interdiffusion is a diffusional process of atoms across two materials that are in contact. We have restricted this work to diffusion data interesting the ISOL community, and so excluded studies related to interdiffusion. This kind of diffusion may involve element or microstructure change during experiments, which can have an impact on the diffusion coefficients.

Regarding the parameters of the diffusing atoms, our database indicates the melting and boiling temperature, the first ionization energy and the atomic radius. For the host material, data collected relate to the structure, the lattice parameters, the density, presence or absence of porosity and impurity, and the grain size. Tables include diffusion coefficients and associated uncertainties for all temperature range available in literature, linked to the parameters in Eq. (2): the pre-factor  $D_0$ , the activation energy  $Q$ , the diffusion coefficient  $D$ , and the temperature  $T$ . For each temperature range, data represent average coefficients. Errors calculated on coefficients correspond to experimental scattering between data. For a diffusing atom, parameters can be available for several isotopes. Additionally, some authors have performed measures for various material conditions (solid and liquid), for specific orientations in single crystals (in parallel and perpendicular directions) and for polycrystals. These values are clearly stated in the current database for enhanced precision of diffusion coefficients. All the data contained in the database can be traced back to the reference in the original work. Furthermore, a graphical interface is being studied to facilitate database use.

Now, diffusion coefficients have been extracted from almost 1500 references. Data were gathered for sixty natural host materials of periodic table (out of 91 existing); correspond to 775 target-element combinations, for temperatures ranging from 200 K to 3000 K (depending on materials). This assessment shows the status of the project, which is still in progress.

The compilation of the large amount of diffusion data is one of the key to improve the future production of more and more exotic ions. The data from literature must be consolidated and completed by data sets from studies not published so far. Therefore, it will be useful to complete the database with data from research organizations interested in the project.

### **New experimental device: MELODICA system**

A second part of our project aims at building a device called MELODICA (MEsure en Ligne de cOefficients de Diffusion et de temps de Collage Atomique) to systematically measure the diffusion coefficients of atoms out of materials at different and elevated temperatures, and the atomic sticking times on a wall. Indeed, studies reported in the bibliography always present diffusion coefficients measured in two steps: implantation of radiotracers, followed later by analysis. To limit the risk of experimental errors induced by non-systematic measurements, our aim is to develop a device for characterizing the release and the sticking times of atoms on varied materials in a single step.

Few years ago, P. Jardin *et al.* have developed a device to perform online diffusion measurements [11]. The system used a primary beam of radioactive ions generated by fragmentation, which were implanted in a target contained in a cavity connected to ECR (Electron Cyclotron Resonance) ion source. Released atoms were ionized, accelerated, mass over charge separated and identified with gamma-spectrometer. The target cavity was heated at different temperatures to study the evolution of the release speed. Diffusion coefficients were deduced by a single on-line measurement in two distinct modes: continuous or pulsed. The device was also equipped with an automated target changing system, to assess several varied materials during the same run and enabling experiments to be conducted under similar conditions. Once the experiment was finished, the device was hardly reusable due to its activation. The results and principles are presented in C. Eléon's PhD thesis [12].

The reason the MELODICA system under study will use stable ions contrary to the TargISOL system, is to get rid of constraints related to the use of a radioactive ion beam. Implanting atoms from a low intensity stable ion beam and using a low intensity detector (1 pps sensitive) in place of a gamma-identification station should be possible to obtain information comparable to the one obtained with radioactive tracers. It will also be possible if the mass separation is sufficient to remove the contaminants. Principle consists in implanting pulses of ions into a thin foil target, heated to enhance

the atom diffusion speed while being in conditions comparable to the one of the ISOL systems (typical target temperature between 1000°C and 2200°C). Atoms effuse in a cavity until they reach an ionization source. Ions produced are then extracted and filtered to remove contaminants, before being collected on a low intensity detector (Fig. 2).

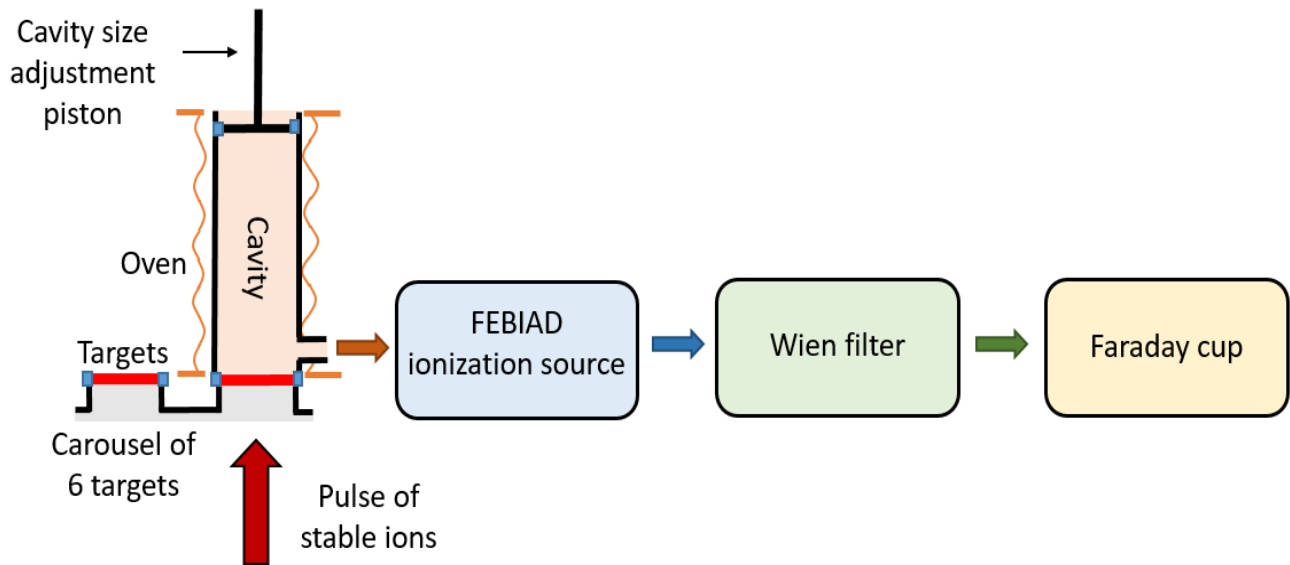


Figure 2. Scheme of the MELODICA principle.

Additionally, we want to measure the sticking time for different atoms, for various kinds of target and cavity materials. This parameter depends on several specific characteristics of the effusing atom and of the materials: physico-chemical features of effusing atom, structure and chemical composition of material, impurity, porosity, topography, ... To measure diffusion, effusion and ionization times for several target-element combinations, target can be exchanged easily using a carousel of several targets (up to 6 foils), allowing measurements with identical experimental conditions (vacuum, heating, ...). The cavity has an adjustable volume to differentiate diffusion and effusion times. Measuring the variation of the global response time versus the effusion volume should allow us to extract the diffusion time. Thanks to the carousel, targets can be changed easily between measurements without opening the vacuum chamber, which saves time and maintains the target environment unchanged.

As it can ionize a large panel of elements, a FEBIAD source (Forced Electron Beam Ion Arc Discharge) [13,14] is considered to transform the atoms coming from the cavity into ions. The ion source works at a specified temperature to limit the atom sticking time and to promote electro-emissivity, as the ionization principle is based on the production of energetic electrons colliding with

the atoms. The ions are then extracted using a potential difference applied between the FEBIAD system and an extraction electrode.

The maximum temperature aimed for the cavity is 2000°C. At this temperature, major part of the elements will be evaporated and ionized by the FEBIAD source, except refractory elements. Thus, contaminants can be collected during experiments. To separate the ion of interest from background noise for measurements, a Wien filter and beam optics are added at source output. The Wien filter consists of a combination of an electric field crossed with a fixed permanent magnetic field. By adjusting the electric field, it is possible to optimize the transmission of the filter for a particular ion mass to charge ratio. Beam optics consist of two electrostatic lenses localized at the injection and at the extraction of the Wien filter. Lenses allow the focus at the detection plane. As for the detection system, a Faraday cup for currents higher than 10 pA or microchannel plates for currents lower than  $1\text{E}+6$  pps can be used to measure extremely low currents.

For analysis, a theoretical model is set up to describe various stages of the physical processes involved in the MELODICA device: diffusion, effusion and ionization. These processes partly compete in time but are considered successive. The shape of the measured response time results from the convolution of the release time of atoms out of the foil, of the effusion time and of the ionization time. It thus directly depends on the diffusion coefficient.

For now, optical studies of the electrostatic lenses, the Wien filter, and optics up to the detection system are already performed. Thermal studies of the cavity are still under simulation and need to be completed to finalize the mechanical design. Regarding the process to properly extract the diffusion coefficients from the experimental curves of the ion release, it will benefit from previous experiments performed with ISOL systems and will be further developed and optimized by practicing the device.

Thanks to the MELODICA device, it will be possible to systematically extract several diffusion coefficients during the same experiment with identical experimental conditions. Even if the analysis and the separation of the several coefficients is complicated, we will at least be able to compare the relative release of atoms out of materials currently used in the ISOL systems.

## **Conclusion**

We are establishing a large and consistent database of diffusion coefficients. The construction of this diffusion database will help us to develop ISOL systems targets in a short term but also will help the ISOL community for the development of new atom-to-ion transformation systems.

Additional experimental diffusion data are needed to fortify the database. Hence, we are developing a novel device to systematically measure the diffusion parameters. Furthermore, carrying out

measurements of diffusion coefficients in identical experimental conditions, and in conditions similar to the ones present in the ISOL systems, will certainly ease their comparison and their choice to optimize the performance of the radioactive ion production systems, even if the determination of the absolute values of the diffusion coefficients is not insured.

## Acknowledgements

The French National Agency of Research (ANR) supports the TULIP project (Projet-ANR-18-CE31-0023).

## References

- [1] S.S. Naghavi, V.I. Hegde and C. Wolverton, Diffusion coefficients of transition metals in FCC cobalt, *Acta Materialia*, 132 (2017) 467-478.
- [2] G. Neumann and C. Tuijn, *Self-diffusion and impurity diffusion in pure metals: handbook of experimental data*, Elsevier (2008).
- [3] H. Mehrer, *Diffusion in solids: Fundamentals, methods, materials, diffusion-controlled processes*, Vol. 155, Springer Science & Business Media, (2007).
- [4] P. Jardin, M. MacCormick, V. Bosquet, P. Chauveau, S. Damoy et *al.*, Sub-millisecond atom-to-ion transformation in the TULIP ISOL system, *Nucl. Instrum. Methods Phys. Res. A*, 1055 (2023) 168332.
- [5] <https://anr.fr/Projet-ANR-18-CE31-0023>
- [6] O. Tengblad, M. Turrion and L.M. Fraile, TARGISOL: An ISOL-database on the web, *The European Physical Journal A-Hadrons and Nuclei*, 25 (2005) 763-764.
- [7] M. Turrión, M. J. G. Borge, and O. Tengblad, Database for ISOL-production optimization with web access, *Nucl. Phys. A*, 746 (2004) 441-444.
- [8] N.L. Peterson, Self-diffusion in pure metals, *Journal of Nuclear Materials*, 69 (1978) 3-37.
- [9] R. Kirchner, On the release and ionization efficiency of catcher-ion-source systems in isotope separation on-line, *Nucl. Instrum. Methods Phys. Res. B*, 70(1-4) (1992) 186-199.
- [10] H. B. H. P. Bakker, *Diffusion in solid metals and alloys*, Springer (1990).
- [11] P. Jardin, M. G. Saint-Laurent, F. Durantel, C. Eléon, C. Huet-Equilbec, R. Alvès Condé, J. Cornell, G. Gaubert, J. Y. Pacquet, F. Pellemoine, and M. Ozille. Set-up for systematic measurements

of diffusion of atoms from ISOL targets, Nucl. Instrum. Methods Phys. Res. B, 266 (2008) 4322-4325.

[12] C. Eléon, Recherche et développement concernant la production d'ions radioactifs dans le cadre de SPIRAL, Doctoral dissertation, Université de Caen (2007).

[13] R. Kirchner, and E. Roeckl, Investigation of gaseous discharge ion sources for isotope separation on-line, Nucl. Instrum. Methods, 133(2) (1976) 187-204.

[14] L. Penescu, R. Catherall, J. Lettry, and T. Stora. Development of high efficiency versatile arc discharge ion source at CERN ISOLDE. Review of Scientific Instruments, 81(2) (2010).