



A desorption model for the code SOLIDUSS and its experimental benchmarking

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ABSTRACT

The code SOLIDUSS is a Monte Carlo based solid-state diffusion software for radiation protection. It was developed to accurately estimate the amount of radionuclides that could escape activated material affected by an accidental fire. A desorption model based on the computation of the desorption probability of those radionuclides reaching the surface of an object was introduced to upgrade the software, proven to be a significant improvement with respect to earlier stages of the code.

A set of experiments was performed at CERN to estimate the out-diffusion of radionuclides from activated materials typically used in accelerator environments when exposed to high temperatures. In particular, a 49.3 μm thick Cu foil containing ⁶⁰Co and a 94 μm thick Al foil with ²²Na were exposed to approximately 1000 °C and 600 °C respectively for different time periods. Out-diffusion fractions of 1.5 ± 5.5% for ⁶⁰Co after 5 h and 22.5 ± 3.1% for ²²Na after 4 h were obtained.

A set of SOLIDUSS simulations was carried out replicating the experimental setup and using literature diffusion and desorption activation parameters. The results obtained are in good agreement with the experimental data within error bars. A high sensitivity of the simulation results to changes in the input parameters was observed.

1. Introduction

The code SOLIDUSS (Ogallar Ruiz et al., 2022) was developed at CERN (European Organization for Nuclear Research) (Anon, 2021a) in the context of the FIRIA project (Anon, 2021b; Gai, 2021) to be used to accurately estimate the contribution of the out-diffusion of radionuclides to radiological source terms in case of accidental fires affecting activated areas of particle accelerators facilities. It is a Monte Carlo (MC) based software able to track diffusing radionuclides through arbitrarily complex geometries according to user-defined 3-D temperature maps. It runs coupled with the general purpose MC code FLUKA (Battistoni et al., 2015; Ahdida et al., 2022; Anon, 2021c), making use of its geometry kernel.

In the present paper we discuss the model implemented in SOLIDUSS to assess the desorption of those radionuclides reaching the surface of the object that contains them. Then, we describe the experiments carried out to obtain data on the out-diffusion of radionuclides from activated materials when exposed to high temperatures. To conclude, the benchmarking of the SOLIDUSS code against these data is analysed and its results are discussed.

2. Desorption in SOLIDUSS

We are interested in estimating the release probability for a radionuclide that has reached the surface of the solid that contains it through thermally promoted diffusion. If we ignore potential chemical reactions with surrounding atmosphere elements, this probability equals the desorption probability. In such a situation, diffusion and desorption would be mutually exclusive processes. Nonetheless, if the radionuclides diffuses back into the solid matrix, its trajectory may encounter again the surface of the object and have a second chance of desorption.

2.1. Desorption rate

Let us imagine a surface with a given number of adsorbed atoms, θ_0 . If we assume that the only process these atoms can undergo is first-order desorption, the number of atoms on the surface will vary with time t as follows (Frenkel, 1924):

$$\frac{d\theta}{dt} = -\theta v_0 \exp\left(-\frac{E_{des}}{k_B T}\right), \quad (1)$$

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where E_{des} is the desorption activation energy,¹ ν_0 the vibration frequency of the atom, T is the absolute temperature and k_B the Boltzmann's constant. Solving this differential equation one obtains that:

$$\theta(t) = \theta_0 \exp\left(-\frac{t}{\tau}\right), \quad (2)$$

where τ is the so-called sojourn time, which characterizes the desorption process and is given by:

$$\tau = \frac{1}{\nu_0} \exp\left(\frac{E_{des}}{k_B T}\right). \quad (3)$$

The desorption rate r_{des} can be obtained from the previous expression as follows:

$$r_{des} = \frac{1}{\tau} = \nu_0 \exp\left(-\frac{E_{des}}{k_B T}\right). \quad (4)$$

The vibrations of adsorbed atoms are induced by the vibrations of the adsorbent crystalline lattice. Hence, ν_0 is assumed to be a property of the adsorbent only and supposed to be independent of the temperature (Zvára, 2008). The diffusion rate is typically given by a very similar expression:

$$r_{dif} = \nu_0 \exp\left(-\frac{Q}{k_B T}\right), \quad (5)$$

where Q is the activation energy of diffusion. We have assumed that the effective vibration frequency of the atom is equivalent to that of the desorption process. We can see that whenever a radionuclide reaches a surface there will be a competition between those two processes, and the desorption probability would be given by:

$$P_{des} = \frac{r_{des}}{r_{des} + r_{dif}}. \quad (6)$$

From a Monte Carlo perspective, we could sample the process taking place by sampling a random number ξ from a uniform distribution $U(0, 1)$. If $\xi < P_{des}$, desorption will follow. Otherwise, we shall assume that the radionuclide jumped back inside the solid.

2.2. Desorption model for SOLIDUSS

The code SOLIDUSS simulates the thermally promoted diffusion of radionuclides in activated solids sampling their path at a given scale above the atomic level. It uses adaptive time steps, which condense multiple microscopic steps into a single one. The MC model for desorption described above is therefore not suitable to be implemented in SOLIDUSS as is because one would need to follow each atomic step to decide between desorption and diffusion every time the radionuclide reaches a surface. Nonetheless, it can be adapted to be implemented in SOLIDUSS on top of its diffusion treatment. The key would be to find out the number of times N_{enc} that a given radionuclide would reach a surface during a simulation time step Δt , provided that it does not desorb in any of the encounters. With this data, we can obtain the probability for a radionuclide to desorb in one of those encounters, P_{desT} , as follows:

$$P_{desT} = 1 - (1 - P_{des})^{N_{enc}}. \quad (7)$$

Knowing this and proceeding analogously as before, we could sample a random number ξ from a uniform distribution $U(0, 1)$ and compare it with P_{desT} . If $\xi < P_{desT}$, the radionuclide desorbed after one of the surface encounters that took place during the time step. If not, it is still inside the solid and its new position should be properly sampled.

Once a radionuclide is found to reach a surface while diffusing, the desorption model decides whether or not it desorbs in the current simulation step. In order to properly sample the stochastic quantity

¹ Also known as desorption enthalpy or desorption energy. Note that we are assuming ambient pressure.

N_{enc} , it is essential to know the number of jumps that the radionuclide performs within the solid lattice during a simulation time step, as well as the moment of the first surface encounter (or hit). To obtain the latter, we first estimate the distance from the initial position of the radionuclide to the surface by exploiting SOLIDUSS' internal link to the FLUKA geometry navigator. Then, we sample the time t_0 at which the first hit to the surface takes place, since it is well-known that the time at which a 1-D Brownian particle² with no drift first reaches a wall-like boundary follows a Lévy Distribution (Bachelier, 1900) which depends on the initial distance from the radionuclide to the surface and its diffusion coefficient (see for instance Redner, 2001 for an extended discussion). After sampling the first hitting time, we know the time left in the current simulation step at the radionuclide's disposal to diffuse and successively encounter the surface. The number of atomic jumps N performed in a time t is given by:

$$N = \frac{6Dt}{A^2}, \quad (8)$$

where D is the diffusion coefficient and A is the jump spatial size, which can often be approximated by the atomic bond length. This parameter, together with the desorption activation energy should be provided by the user in the so-called diffusion input file³ of SOLIDUSS if desorption is to be simulated. Once a random walker reaches a boundary for the first time, the average number of times that it will encounter the boundary again can be well approximated by the following expression:

$$\langle N_{enc} \rangle \simeq a \cdot \sqrt{N}, \quad (9)$$

where a is a parameter that depends on the lattice structure and its orientation with respect to the boundary. This has been inducted from the results of numerous 3-D random walk simulations, but it can be easily and analytically obtained for particular cases, such as a 1-D random walk. The orientation of the lattice with respect to the surface of the object will be unknown in the scenarios in which SOLIDUSS would be employed to assess the potential out-diffusion of radionuclides. Furthermore, it is probable that the crystal lattice structure would also be unknown. We must therefore chose a value for the parameter a to be used as default. We deemed it appropriate that, given the radiation protection motivation of this work, we should stay on the safe side and overestimate the number of surface encounters (and as a consequence, the desorption probability) rather than underestimate it. This quantity is maximized when the jumps of the random walker are considered to be isotropic, obtaining:

$$\langle N_{enc} \rangle \simeq \sqrt{\frac{8N}{3\pi}}. \quad (10)$$

The Probability Density Function (PDF) of the number of surface encounters for a given number of atomic jumps is well approximated by a half-normal distribution. The mean of the half-normal distribution is a function of its standard deviation ($\mu = \sigma\sqrt{2/\pi}$) and therefore one single parameter characterizes the PDF. Since we already have a method to calculate $\langle N_{enc} \rangle$, which is the mean of the PDF, we can sample N_{enc} for every time step in which a radionuclide is found to have reached a surface. Finally, we can make use of Eqs. (6) and (7) to obtain the desorption probability and sample if the radionuclide would desorb or not during the time step under consideration. If the outcome of the sampling determines that the radionuclide did not desorb, the code would continue tracking its trajectory through the solid, which could bring it to the surface again.

² Given the microscopic distances travelled by radionuclides in the time steps under consideration (order of seconds) and the macroscopic size of the objects for which SOLIDUSS is conceived, we can safely assume 2-D boundaries. Therefore, although we deal with 3-D trajectories, the displacements bringing the radionuclide closer or farther from the boundary take place in 1-D.

³ Text file specifying the information necessary to perform the out-diffusion simulations using SOLIDUSS.

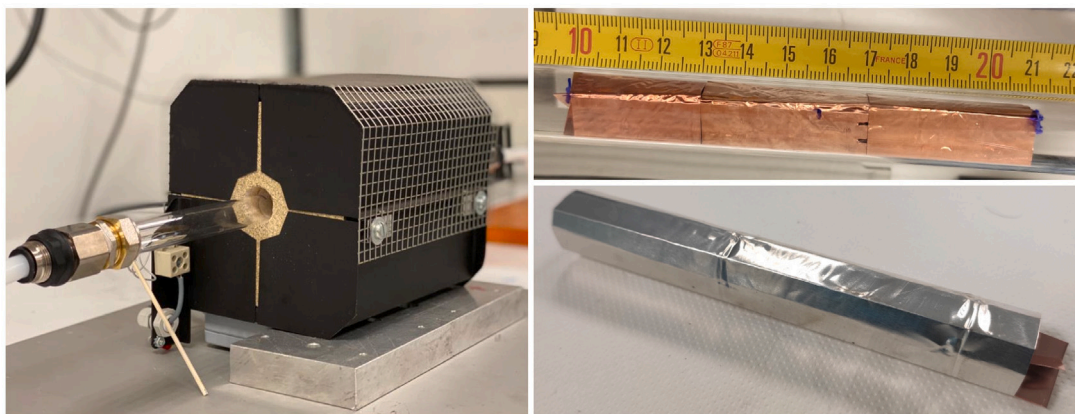


Fig. 1. Furnace used in the experiments (left), Cu sample (right top) and Al sample (right bottom), where the inner cross-shaped Cu bar is slightly visible.

3. Out-diffusion experiments

An experimental campaign was carried out intended to provide data on the out-diffusion of radionuclides from activated materials typically used in accelerator environments when exposed to high temperatures. This data was later used to benchmark SOLIDUSS.

3.1. Methodology and samples

Two radioactive metallic foils were selected to be used as the samples of the experiments. The first one was made out of Cu, with 0.65 ± 0.05 Bq of ^{60}Co , and dimensions 11.0 ± 0.2 cm \times 4.8 ± 0.1 cm \times 49.3 ± 1.3 μm thick. The second one was made out of Al, with 18 ± 1 Bq of ^{22}Na , and dimensions 11.0 ± 0.1 cm \times 4.8 ± 0.1 cm \times 94 ± 2 μm thick. Both were chosen according to two criteria: their low and yet measurable activity to guarantee the safe outcome of the experiments, and their thinness, which increases the chances of radionuclides reaching their surface. They were not specifically irradiated and thus activated for these experiments, but they had been previously located in the CERN accelerator complex. Given the extended radiation fields they were exposed to in comparison to their sizes as well as the high energies of the impacting particles, it is justified to assume a uniform activation of the samples.

The procedure followed for each experiment can be summarized in the following steps:

1. Gamma-ray spectrometry measurement to accurately determine the radionuclides present in the sample and their activities. These measurements were performed by the staff of the HSE-RPCS section at CERN in their gamma-ray spectrometry laboratory using a Ge detector.
2. Heat the sample up to high temperatures during a certain period of time. This is performed to maximize the out-diffusion fraction (ODF), which is the fraction of radionuclides that manages to escape from the material. This has been done in the Solid-State Physics (SSP) labs of ISOLDE (Anon, 2021d,e) using a furnace (see Fig. 1) able to reach temperatures well beyond 1000 °C (HTM Reetz GmbH type LOBA 1100-25-150), tuned by means of a voltage regulator. The temperature was maximized while keeping it sufficiently low to avoid melting. The temperature profile inside the furnace used in the experiments was characterized.
3. Gamma-ray spectrometry of the samples. This second measurement is performed under the same experimental conditions as the first one. Subtraction of both results allows to estimate the ODF.

The central element of the performed experiments is the heating up of radioactive samples, which is carried out in a controlled oxygen-free atmosphere of pure N_2 in order to avoid their combustion. To do

this, the samples are placed inside a gas circuit that consists of the following elements: the N_2 gas source, a pressure regulator, a 20 mm outside diameter quartz tube in which the samples are placed, an inline HEPA filter to capture radioactive aerosols that may be released from the samples during the experiments, and a bubbler to monitor the gas flux. Formation of volatile radioactive compounds is not expected given the radionuclides species present in our samples. In addition to this, their very low activity ruled out the need for using activated charcoal filtration. Nonetheless, the gas is released inside a fume hood when exiting the circuit, preventing its liberation in the laboratory. The radioactive foils are folded following a squared cylinder (see Fig. 1) in order to be placed inside the quartz tube, which minimizes the contact between the samples and the tube. A cross-shaped bar of Cu was introduced inside the foils cylinder, in order to increase their structural stability while reducing the re-deposition of radionuclides out-diffusing from the sample.

3.2. Results and discussion

The Cu sample was exposed to temperatures around 1000 °C, and the Al sample to 600 °C. The results of the characterization of the temperature maps are shown in Fig. 2. We assume them to be purely longitudinal given the short transverse aperture of the furnace. The sources of uncertainty taken into account are: the accuracy of the thermometer and the thermocouple probe used to measure the temperature, the existence of temperature fluctuations, and the uncertainties associated to the determination of the position of the thermocouple tip. A correction was performed when computing the temperature profiles, taken into account that the thermocouple is placed outside the quartz tube, and therefore outside the flowing N_2 in which the samples are immersed. To define the correction to be implemented, several auxiliary tests were carried out.

The samples were placed in the region of the furnace where the most uniform temperatures are found. Specifically, the Cu sample was placed between position = 4 cm and position = 15 cm, and the Al one between the position = 5 cm and position = 16 cm. Several heating iterations were performed, two for the Cu sample (60 min and 240 min), and three for the Al one (120 min, 250 min and 370 min). After each of the iterations, a gamma-spectrometry measurement was performed. Combining the results and computing the ODF we obtain Table 1.

In addition to this analysis, we have carried out combined spectrometry analysis of the HEPA filter used in the experiments and the cross-shaped Cu bars introduced inside the samples. Both elements were replaced by new ones after each iteration. There was no activity detected for those used with the Cu sample but, in contrast, the results for those of the Al one are shown in Table 2.

In Table 1 we can see that the results for the Cu sample show an ODF of $1.5 \pm 5.5\%$ after 5 h of heating. As a consequence, we cannot

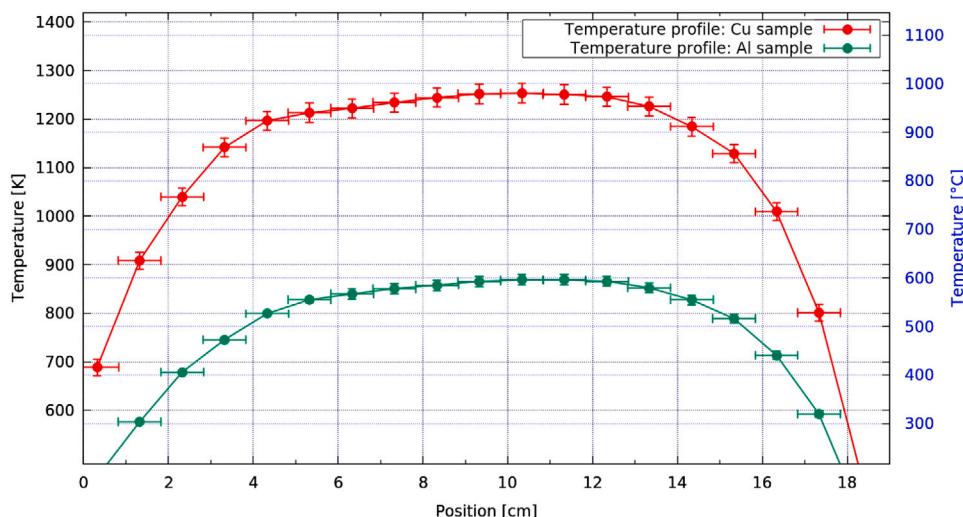


Fig. 2. Temperature profiles to which the Cu and Al samples were exposed inside the furnace.

Table 1

Experimental ODFs for Cu and Al samples. Note that since the ODF is given as a percentage, its uncertainty, although absolute, is also a percentage.

Sample	Nuclide	Heating time [min]	ODF [%]
Cu	^{60}Co	60	4.9 ± 5.4
		300	1.5 ± 5.5
Al	^{22}Na	120	14.6 ± 3.3
		250	22.5 ± 3.1
		370	40.4 ± 2.5

Table 2

Results of the combined gamma-ray spectrometry measurements of the cross-shaped Cu bars and the HEPA filter used in each of the iterations. MDA stands for Minimum Detectable Activity.

Nuclide	Iteration	Activity [Bq]	MDA [Bq]
^{22}Na	1st	0.95 ± 0.14	0.155
	2nd	0.88 ± 0.06	0.128
	3rd	0.64 ± 0.14	0.159

conclude that any ^{60}Co isotope escaped from the sample; but we can say that the amount that escaped was less than 7.0% with 1σ certainty. In the Al case, it is clear that some of the radioactive content of the sample out-diffused, somewhere in the range of [11.3,17.9]% in 2 h and up to [37.9,42.9]% in approximately 6 h. These results are also supported by those in Table 2, which are a second confirmation of the fact that a significant portion of the ^{22}Na escaped from the sample. Of course, the radionuclides deposited on the Cu bars and those captured by the HEPA filter are only a portion of the released ones since many of them are expected to be deposited in the gas circuit between the sample and the filter. Note also that heating times reported correspond to the time that the sample spent inside the furnace at the maximum temperature. Yet, several minutes are needed for the sample and the tube to be brought up to this temperature once introduced in the furnace. Similarly, few minutes are needed to cool them down after their extraction. The decay of the radionuclides has been taken into account to correct the sample activities, since the spectrometry analysis were performed in different days. This way they all refer to the same moment in time and are therefore directly comparable.

During visual examination after the experiments, we noticed the appearance of a small hole (approx. 2 mm^2) in the Al sample. Since the temperature was continuously monitored during the process and it stayed at all times well below the Al melting point, we believe it may have been caused by some (momentary or sustained) hot spot inside the furnace.

4. Experimental benchmarking of SOLIDUSS

In this section we present the simulations performed using the SOLIDUSS code with the aim of reproducing the experiments described above.

4.1. Simulation setup

The simulation procedure is as described in Ogallar Ruiz et al. (2022). A FLUKA geometry has to be created and the generation of the radionuclides that are created in the material of interest has to be simulated. Then, SOLIDUSS will take over and track each one of them, accounting for its potential out-diffusion. For these specific simulations, the radionuclides have been uniformly sampled along the material since we assume a uniform activation of the experimental samples. We note in passing that the spatial distribution of the radioisotopes can also be determined with FLUKA based on a beam impacting on a material and subsequent generation of radionuclides. Two radioactive foils were modelled as those used in the experiments according to the dimensions specified in the previous section. The temperature maps provided to SOLIDUSS are those measured in the experimental campaign but, since SOLIDUSS is currently unable to deal with uncertainties in the temperature maps, we need to make different simulations using the extreme values of the temperature profile in order to find out its impact on the out-diffusion of radionuclides. To provide the necessary parameters needed to simulate diffusion and desorption, a diffusion input file is provided together with the FLUKA input. For this simulation, the values used are shown in Table 3, which have been obtained from the sources specified in the table caption.⁴

⁴ Some of the parameters' uncertainties are not specified because they are not reported in the original references. Nonetheless, DNN values are well known for the materials of interest, with an accuracy of the order of 1 pm. Variations of such order would have no visible impact in our results and therefore such uncertainties are negligible. Uncertainties for the A_1 parameter for Na inside Al are also not provided in the original reference. Note that A_1 and Q_1 are extracted through the fit of a set of diffusion coefficients D_i experimentally obtained for different temperatures T_i , and therefore the values of both parameters and uncertainties are strongly correlated. Since there is no uncertainty reported we must consider no uncertainty for this parameter since any other assumption would be highly arbitrary. Furthermore, the authors suspect that the uncertainty of the diffusion coefficients obtained in Sudár et al. (1977) is well represented by the reported uncertainty of the activation energy Q_1 . Regarding the desorption activation energies E_{des} , there is no

Table 3

Parameters provided to SOLIDUSS to simulate the out-diffusion of radionuclides of interest (Nuc.) from their host materials (Mat.). A_i and Q_i are the frequency factors and the diffusion activation energies respectively, which have been obtained from Brandes and Brook (1992), Sudár et al. (1977), Neumann and Tölle (1988). E_{des} is the desorption energy, taken from Roßbach and Eichler (1984) and DNN is the distance between nearest atomic neighbours, extracted from Kittel (2005).

Mat.	Nuc.	A_1 [$\text{cm}^2 \text{s}^{-1}$]	Q_1 [kJ mol^{-1}]	A_2 [$\text{cm}^2 \text{s}^{-1}$]	Q_2 [kJ mol^{-1}]	E_{des} [kJ mol^{-1}]	DNN [pm]
Cu	^{60}Co	$0.74^{+0.22}_{-0.17}$	217.2 ± 1.9	$736^{+2.9 \cdot 10^6}_{-735.8}$	312.8 ± 96.5	416.1	256
Al	^{22}Na	$6.7 \cdot 10^{-4}$	97.1 ± 13	–	–	134.5	286

Table 4

ODF results obtained by means of SOLIDUSS simulations imitating the experiments performed.

Material	Nuclide	Heating time [min]	ODF [%]
Cu	^{60}Co	60	$1.5^{+0.4}_{-0.9} \cdot 10^{-2}$
		300	$7.7^{+1.7}_{-3.4} \cdot 10^{-2}$
Al	^{22}Na	120	$34.7^{+55.5}_{-24.2}$
		250	$49.2^{+48.2}_{-34.1}$
		370	$59.7^{+39.6}_{-41.3}$

4.2. Results and discussion

We have used three different sets of data in order to obtain a mean for the ODF as well as lower and upper limits to it taking into account the uncertainties of the different parameters employed (see Table 3, Fig. 2 and samples sizes reported above). The simulation results are shown in Table 4.

At a first glance, a result like the one for ^{22}Na in Al after 250 min may seem to be covering almost the whole range of values and therefore may be misinterpreted as a poor result when thinking linearly on the [0–100]% range. To better understand why this is a valuable result, let us consider a simple example. If an activated object contains 10 GBq of a given radionuclide, the out-diffusion of few GBq (case of Na in Al) in case of fire exposure may be radically different in terms of radiation protection than the out-diffusion of few MBq (case of Co in Cu), which is still far from zero and could be important, even if the relative amount escaping may be considered as low. Therefore, a logarithmic approach to the ODF results is advisable.

The comparison between experimental and simulation results is illustrated in Fig. 3, where we can see that both are in good agreement within error bars.⁵ Unfortunately, the uncertainties could not be further reduced. In the Co case, this is because of the experimental procedure, which does not allow for establishing a lower limit above zero when the ODF is found to be below few percent due to the uncertainties of the gamma spectrometry analysis. In the case of Na, the largest error bars are those of the simulation results, since the uncertainties in temperature and the diffusion activation energy have a remarkable impact on the results due to the exponential behaviour of the diffusion coefficient and the desorption rate as functions of these parameters. The contribution of the simulation statistical uncertainty is minor in comparison.

The results clearly support the importance of the implemented desorption model. According to further simulation results, approximately

experimental data published for these quantities to the best of the authors' knowledge. The data used here was reported to be calculated by means of a theoretical model (details can be found in Roßbach and Eichler, 1984) and is not accompanied by any uncertainty estimation. Therefore, we do not consider any uncertainty for these parameters in our calculations in order to avoid the introduction of rather arbitrary estimations. Nevertheless, the reader should be aware of this fact.

⁵ Resuming the discussion about the lack of uncertainties for some parameters used in the calculation, the reader may have notice that the agreement of experimental and simulated results within error bars would remain true even if further uncertainties would be introduced, since those could only extend the error bars of our simulated results.

96% of the ^{60}Co reached the sample's surface during the 5 h of heating (considering mean values for all input parameters), while only a very small fraction of them managed to desorb. Therefore, the results would have been definitely off without the desorption model. This is of especial significance when compared with the ^{22}Na in Al scenario, in which the desorption model becomes almost transparent (due to the high desorption chances of Na).

The appearance of the aforementioned small hole in the Al sample suggests the existence of a certain heterogeneity of the temperature on the transversal axes of the furnace. Nonetheless, the longitudinal characterization of the temperature in real conditions of use and its continue monitoring in a point of reference during the experiments exclude a generalized spatial or temporal heterogeneity. The appearance of hot or cold spots should then have been very localized in space and/or time. The post-experiment Al sample presented evidence for a single hot-spot, which supports this idea. We believe the uncertainty introduced in the final result by such heterogeneity would be rather negligible when compared to the rest of uncertainties encompassed in this analysis. Note that even the complete out-diffusion of the radionuclides contained in the piece of material originally placed in the hole position would increase the ODF by less than 0.05%.

Let us take a closer look to the results obtained for ^{22}Na in Al. Three heating iterations of approximately 2 h each were carried out during the experimental campaign. The ODF in successive iterations is expected to behave like that of the simulation results: in every iteration, the ODF is smaller than in the previous one, asymptotically decreasing as the sample loses radionuclides. The reason behind is that those radionuclides closer to the surface of the sample would out-diffuse in earlier iterations and, as a consequence, the concentration of radionuclides in the outer regions of the sample would decrease. The ODF would then diminish due to the lower probability of deeper radionuclides reaching the surface. This is consistent with the behaviour of the ODF mean that we observe in the 2nd experimental iteration with respect to the 1st, but is clearly not the case for the 3rd one. There is a variation in the tendency, which could be related to the potential damage caused to the structure of the metal as a consequence of repeated and rapid heating and cooling of the sample. This may have resulted in the creation of significant lattice defects, which would enhance the diffusion of radionuclides inside, modifying the behaviour of the ODF since, as mentioned, it is limited by diffusion in the case of Na in Al.

Diffusion and desorption are phenomena with an exponential dependence on several parameters, such as diffusion and desorption activation energies and temperature. Such type of dependence make results highly sensitive to variations of those parameters. For this reason, any future user of the code is advised to consider their uncertainties whenever possible and carry out sensitivity analysis if necessary to estimate their potential impact. However, it is worth mentioning that there are situations in which changes in those parameters would have little impact. Take, for instance, the case of Na in Al discussed in the present document: out-diffusion is limited by diffusion since all radionuclides reaching the surface of the sample manage to desorb, and therefore small variations of the desorption activation energy would have no impact on our result. On the contrary, if we consider the out-diffusion of Co from Cu: small changes in the diffusion activation energy would have a much smaller impact than similar changes in the desorption activation energy, since the process limiting out-diffusion in this case is desorption. Changes in temperature, on the other hand,

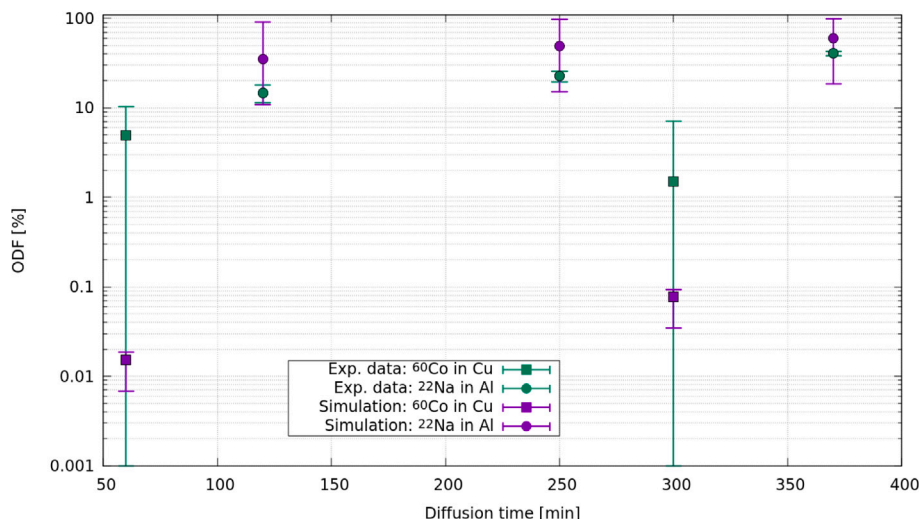


Fig. 3. Comparison between experimental and simulations results. The plotted data can be found in Tables 4 and 1.

have always a considerable impact on the results. The potential user should also keep in mind that the correct modelling of the geometry of the activated material is key, as well as a realistic representation of the radionuclides' initial distribution inside it, in particular for bulky objects.

5. Conclusion

A Monte Carlo based desorption model has been implemented in SOLIDUSS to assess whether or not those radionuclides reaching the surface of a solid would desorb. The main parameter governing the desorption probability is the desorption activation energy.

We have obtained experimental data on the out-diffusion of ⁶⁰Co from Cu and ²²Na from Al when exposed to approximately 1000 °C and 600 °C respectively while immersed in a N₂ atmosphere. The fraction of radionuclides out-diffused in the latter case ($22.5 \pm 3.1\%$ after approx. 4 h) was substantially larger than the former ($1.5 \pm 5.5\%$ after 5 h).

The code SOLIDUSS was benchmarked against the obtained experimental data and a good agreement was observed between simulation and experimental results.

In Ogallar Ruiz et al. (2022) and the present article, the different approximations used by SOLIDUSS to perform calculations have been exposed. In addition, we have seen in the previous section that results are extremely sensitive to variations of the parameters provided by the user (e.g. activation energies, frequency factors, temperature, etc.), which are often estimations with important uncertainties. Given the tests performed so far and in absence of further supporting experimental data, the authors consider it as prudent to take SOLIDUSS' results as order-of-magnitude estimations, while encouraging the user to perform accompanying sensitivity analyses.

CRediT authorship contribution statement

F. Ogallar Ruiz: Methodology, Software, Formal analysis, Investigation, Writing – original draft, Visualization. **H. Vincke:** Conceptualization, Writing – review & editing, Supervision. **I. Porras:** Writing – review & editing. **C. Theis:** Conceptualization, Software, Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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References

- Ahdida, C., Bozzato, D., Calzolari, D., Cerutti, F., Charitonidis, N., Cimmino, A., Coronetti, A., D'Alessandro, G.L., Donadon Servallo, A., Esposito, L.S., Froeschl, R., Alía, R. García, Gerbershagen, A., Gilardoni, S., Horváth, D., Hugo, G., Infantino, A., Kouskoura, V., Lechner, A., Lefebvre, B., Lerner, G., Magistris, M., Manousos, A., Moryc, G., Ogallar Ruiz, F., Pozzi, F., Prelipcean, D., Roesler, S., Rossi, R., Sabaté Gilarte, M., Salvat Pujol, F., P. Schoofs, V. Stránský, Theis, C., Tsinganis, A., Versaci, R., Vlachoudis, V., Waets, A., Wiodorski, M., 2022. New capabilities of the FLUKA multi-purpose code. *Front. Phys.* 9, 788253.
- Anon, 2021a. CERN website, <https://home.cern/>, accessed 2021.
- Anon, 2021b. FIRIA project website, <https://hse.cern/content/firia>, accessed 2021.
- Anon, 2021c. FLUKA website, <https://fluka.cern>, accessed 2021.
- Anon, 2021d. ISOLDE website, <https://home.cern/science/experiments/isolde>, accessed 2021.
- Anon, 2021e. Solid-state physics at ISOLDE, <https://espace.cern.ch/ISOLDE-SSP/>, accessed 2021.
- Bachelier, L., 1900. Théorie de la spéculation. *Ann. Sci. éc. Norm. Supér.* 17, 21–86. <http://dx.doi.org/10.24033/asens.476>.
- Battistoni, G., Boehlen, T., Cerutti, F., Chin, P.W., Esposito, L.S., A. Fassò, A. Ferrari, Lechner, A., Empl, A., Mairani, A., Mereghetti, A., Ortega, P. Garcia, Ranft, J., Roesler, S., Sala, P.R., Vlachoudis, V., Smirnov, G., 2015. Overview of the FLUKA code. *Ann. Nucl. Energy* 82, 10–18.
- Brandes, E.A., Brook, G.B. (Eds.), 1992. *Smithells Metals Reference Book*, Seventh ed. Reed Educational and Professional Publishing Ltd.
- Frenkel, J., 1924. *Theorie der Adsorption und verwandter Erscheinungen*. *Z. Phys.* 26, 117–138.
- Gai, G., 2021. FIRIA Methodology. Tech. Rep., (EDMS 2426715), CERN, <https://edms.cern.ch/document/2426715>.
- Kittel, C., 2005. In: Wiley, J., Sons (Eds.), *Introduction to Solid State Physics*, Eighth ed.
- Neumann, G., Tölle, V., 1988. Monovacancy and divancy contributions to the impurity diffusion in face-centred cubic metals. *Philos. Mag. A* 57 (4), 621–630. <http://dx.doi.org/10.1080/01418618808214411>.
- Ogallar Ruiz, F., Theis, C., Porras, I., Vincke, H., 2022. SOLIDUSS: SOLID-state diffusion software for radiation protection. *Appl. Radiat. Isot.* 179, 109997. <http://dx.doi.org/10.1016/j.apradiso.2021.109997>.

- Redner, S., 2001. A Guide to First-Passage Processes. Cambridge University Press.
- Roßbach, H., Eichler, B., 1984. Adsorption von Metallen auf metallische Oberflächen und Möglichkeiten ihrer Nutzung in der Kernchemie - Ermittlung von Adsorption-senthalpien mit dem Rechenprogramm AMO. Tech. Rep., (ZfK-527), Zentralinstitut für Kernforschung Rossendorf bei Dresden.
- Sudár, S., Csikai, J., Buczkó, M., 1977. Diffusion of ^{24}Na in polycrystalline aluminum. Z. Metallkde. 68, 740–741.
- Zvára, I., 2008. the Inorganic Radiochemistry of Heavy Elements. Springer Netherlands, <http://dx.doi.org/10.1007/978-1-4020-6602-3>.