




Investigating solid-state reactions in UAl_2 -Al dispersions for Mo-99 target fabrication

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ABSTRACT

Molybdenum-99 (Mo-99), the parent isotope of the widely used medical radioisotope technetium-99m (Tc-99m), is primarily produced via neutron irradiation of uranium-containing targets in research reactors. UAl_2 -Al dispersions, with UAl_2 particles embedded in an aluminum matrix, are commonly employed for target fabrication. This study employs a new method for investigating the solid-state reactions between UAl_2 and the Al matrix during target fabrication using stepwise isothermal dilatometry (SID). The study focuses on the formation of UAl_3 and UAl_4 , which are less dense than UAl_2 and result in material expansion detected by the linear displacement, which relates to reaction conversion. By analyzing the linear expansion data from 560 °C to 630 °C, SID determines apparent activation energies and identifies the D2 diffusion rate model as the best fit, indicating a fast reaction rate along the UAl_2 /Al interfaces and highlighting the crucial role of interface diffusion in the process. This information is crucial for optimizing target fabrication and controlling the final composition of Mo-99 targets.

1. Introduction

The application of radioisotopes in nuclear medicine is an important social utilization of nuclear energy. The use of technetium-99m (Tc-99m) (Papagiannopoulou, 2017), a radionuclide originated from the nuclear decay of molybdenum-99 (Mo-99), has extensive use for acquisition of single-photon emission computed tomography (SPECT) images. The distribution of Tc-99m is facilitated through Mo-99/Tc-99m generators, where Tc-99m is produced as a result of Mo-99 decay. With a global annual demand exceeding 30 million procedures, Tc-99m constitutes approximately 85% of all nuclear medicine diagnostics (OECD-NEA, 2019).

On an industrial scale, the primary method for Mo-99 production involves the fission of U-235 in research reactors, through neutron irradiation of uranium-containing targets (Verbeek, 2008). The use of dispersion-based plate-type targets (Kohut et al.; Ali et al., 2013; Mushtaq, 2011; Ryu et al., 2013; Ryu et al., 2015; Durazzo et al., 2021), employing UAl_2 dispersed in an aluminum matrix (Kohut et al.; Ali et al., 2013; Durazzo et al., 2021), is the predominant technology for the

manufacture of irradiation targets. Employing the well-established "picture-frame technique" (Kaufman, 1962), a set composed of a UAl_2 briquette (fuel meat), an aluminum frame plate, and two aluminum cladding plates undergo assembly and rolling operations, resulting in small aluminum plates containing a dispersion fuel meat where UAl_2 particles are embedded in a continuous aluminum matrix. Fig. 1 illustrates the procedure for preparing the sets for rolling and the manufactured target according to the picture-frame technique.

The uranium-aluminum binary system exhibits a phase diagram outlining intermetallic compounds, namely UAl_2 , UAl_3 , and UAl_4 , collectively referred to as UAl_x . The mixture of aluminides within the final fabricated target (or the x value in the UAl_x formula) depends on the manufacturing process. In this type of target, UAl_2 has been used as the starting material (Kohut et al.; Ali et al., 2013; Durazzo et al., 2021). The use of UAl_2 instead of other aluminides offers advantages in terms of its synthesis, since UAl_2 has a congruent melting point and as a result can be synthesized in a single step, without the need for post-synthesis annealing. The intermetallics UAl_3 and UAl_4 have incongruous melting points and, therefore, are formed through peritectic reactions, requiring

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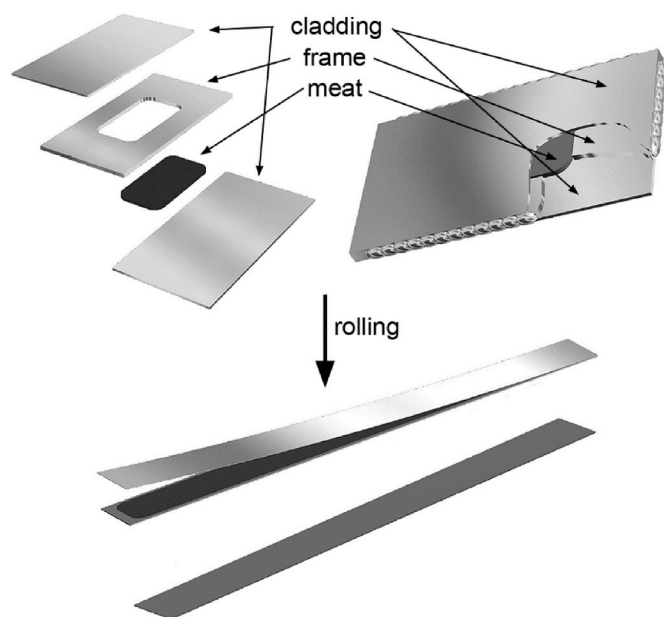


Fig. 1. Picture frame technique used to manufacture targets.

long thermal treatments to complete the synthesis (Bramfitt and Leighly, 1968; Okamoto, 2012). Additionally, UAl_2 has a higher U content (81.52 wt% of U) and density (8.14 g/cm^3) than the other uranium aluminides, maximizing the U-235 content in the target. UAl_2 reacts with aluminum during the target manufacturing, especially during hot rolling and heat treatments, resulting in a mixture of UAl_3 and UAl_4 after fabrication.

The phase composition in $\text{UAl}_x\text{-Al}$ target after fabrication is important because both UAl_3 and UAl_4 are more easily dissolved in alkaline solutions than the UAl_2 , which defines, ultimately, the radiochemical processing throughput after the irradiation (Cols et al.). Thus, the presence of UAl_2 in the $\text{UAl}_x\text{-Al}$ dispersions target must be controlled and preferably avoided.

The phase composition in the meat of $\text{UAl}_x\text{-Al}$ dispersion target can be controlled during manufacturing by means of the rolling temperature, or the duration of heat treatment between hot rolling passes, or final annealing before the cold rolling, or a combination of these. To determine the optimal thermal treatment, the study must focus on the kinetics of reactions involving the aluminides UAl_2 , UAl_3 , and UAl_4 . The aim of the present study is to employ Stepwise Isothermal Dilatometry techniques (SID) to estimate reaction kinetic parameters, such as activation energy and diffusion-related coefficients.

Sørensen (Sørensen, 1981; Husum and Sørensen, 1987) described a method for kinetic investigation known as Stepwise Isothermal Analysis (SIA). In this technique, the user stipulates the range of reaction rate by establishing maximum and minimum values for it (Sørensen, 1992). Once a heating rate is established, when the reaction rate reaches a programmed maximum value, the system initiates an isotherm. As the reaction progresses, the reaction rate decreases to a predetermined minimum value, when the control program commands a new heating until the predetermined maximum reaction rate is reached again. This procedure is repeated until the end of the reaction.

The SIA method allows for more accurate kinetic modeling of solid-state reactions than techniques that do not use isotherms. This method also has the advantages of enabling the separation of consecutive and overlapping reactions and obtaining kinetic information (mechanisms and activation energy) for each reaction without overlap. Reactions are measured in isotherms, providing more reliable data for kinetic studies, allowing for the determination of more precise reaction temperatures, and enabling a comprehensive kinetic study in a single analysis. The generated isotherms provide data for Arrhenius plots, enabling a

sequential quasi-isotherm analysis in a single run. On the other hand, this method requires more time (Gotor et al., 1998).

Another kinetic technique is known as Forced Stepwise Isothermal Analysis (FSIA), which is a variant of the SIA method adopted for the study of fast reactions, where the temperature and the amount of time in each isothermal are pre-defined by the operator and not by the decrease in the reduction rate (Sørensen, 1992).

In the present study, the Forced Stepwise Isothermal Dilatometry (FSIA) technique was applied, using the ΔL signal, corresponding to the expansion resulting from the solid-state reaction between Al and UAl_2 , UAl_3 , and UAl_4 compounds with progressively lower densities: 8.1 g/cm^3 , 6.8 g/cm^3 , and 6.1 g/cm^3 (Kim, 2012), respectively. The methods employed in this study, based on volume expansion measurements via dilatometry, are novel and applied here for the first time.

2. Experimental

The intermetallic compound was synthesized through the combination of metallic uranium and metallic aluminum in stoichiometric proportions, yielding UAl_2 (with a uranium content of 81.5 wt%). Metallic uranium was synthesized via magnesiothermic reduction of UF_4 following the methodologies outlined by Durazzo et al. (2017). The initial components were introduced into a zirconium crucible and subjected to melting using a 15 kW induction furnace. Preceding the melting process, the furnace underwent argon purging subsequent to achieving a vacuum level of 2.6×10^{-3} mbar. Subsequently, the UAl_2 ingot was pulverized within a mortar under an argon atmosphere.

X-ray diffraction data were acquired from UAl_2 powder employing a Bruker diffractometer, operating with $\text{Cu-K}\alpha$ radiation at 40 kV and 30 mA, utilizing a scanning step size of 0.02° and an acquisition time of 8 s per step. The quantification of crystalline phases was executed through the application of the Rietveld method (Rietveld, 1969; Hill and Howard, 1987), utilizing TOPAS V 4.2 for data refinement, as per the methodology delineated by Conturbia et al. (2018). The resultant phase composition derived from the UAl_2 powder is presented in Table 1.

Powder metallurgy techniques were employed to fabricate specimens designed to simulate the fuel meat of the target. The specimen, comprised of powdered UAl_2 and pure aluminum powder, was formulated to result in volumetric fractions of 45% and 50%, respectively. The resulting mixture was pressed at 500 MPa to yield specimens for dilatometric analyses.

The resultant specimens exhibited a microstructure characterized by dispersed UAl_2 particles embedded within an aluminum matrix. The UAl_2 powder had a size composition of 80 wt% between 88 and $44 \mu\text{m}$, and 20 wt% of particles smaller than $44 \mu\text{m}$. This granulometric composition is traditionally used in the manufacture of targets. Additionally, specimens incorporating UAl_2 powder with particle sizes below $44 \mu\text{m}$ (fine powder) were tested to assess the influence of UAl_2 powder particle size on its reaction with aluminum within the dispersion matrix.

The investigation of solid-state reactions was conducted using dilatometry with a Setaram Setsys 1750 °C Thermal Analyzer, employing the Forced Stepwise Isothermal Dilatometry (FSIA) technique in the newly proposed expansion mode. A forced stepwise analysis was programmed on the dilatometer, with a heating rate of $10 \text{ }^\circ\text{C}/\text{min}$ and 15-min isothermal steps ranging from $560 \text{ }^\circ\text{C}$ to $630 \text{ }^\circ\text{C}$, with intervals of $10 \text{ }^\circ\text{C}$. To study rapid reactions occurring in the initial stages with fine

Table 1
Phase composition on the UAl_2 powder.

Phase	Quantity (%)
UAl_2	91.7 ± 0.3
UAl_3	5.5 ± 0.2
Oxides (UO_2, UO)	2.8 ± 0.2
Empirical Formula	$\text{UAl}_{1.97}$

UAl₂ powder (<44 μm), the isothermal times were reduced to 10 min. The atmosphere maintained a pure argon environment at a dynamic flow rate of 30 mL/min to prevent oxidation. The involved reactions lead to expansion of the pellet sample. In this sense, the measured dilatation Lt was computed to obtain the reaction conversion α according to eq. (1), assuming the expansion is isotropic:

$$\alpha = \frac{Lt^3 - Lo^3}{Lf^3 - Lo^3} \quad (1)$$

where Lf and Lo are the final and initial sample length.

The differential kinetic method states the derivative of conversion is proportional to the model-representative function at each isotherm:

$$\frac{d\alpha}{dt} = k(T) \cdot f(\alpha) \quad (2)$$

k being the reaction rate obeying Arrhenius equation.

A correct selection of $f(\alpha)$ is the key point for kinetic analysis. One first approach is to choose diffusion reaction rate equations D1, D2 and D3 according the ICTAC Kinetics Committee recommendations (Vyazovkin et al., 2011), since UAl₂ → UAl₃ → UAl₄ solid state reactions depend on Al diffusion into the aluminides lattice.

$$\text{D1 diffusion: } 1/(2\alpha) \quad (3)$$

$$\text{D2 diffusion: } [1/\ln(1 - \alpha)] \quad (4)$$

$$\text{D3 diffusion: } [3(1 - \alpha)^{2/3}]/[2(1 - (1 - \alpha)^{1/3})] \quad (5)$$

The natural logarithm of conversion derivative $\ln(d\alpha/dt)$ (eq. (2)) allows the graphic determination of $k(T)$ which, by its turn, leads to the apparent energy E determination by plotting $\ln K$ versus $1/T$. The equation becomes:

$$\ln\left(\frac{d\alpha}{dt}\right) = \ln K + \ln f(\alpha) \quad (6)$$

followed by:

$$\ln(K) = -\frac{E}{R}\left(\frac{1}{T}\right) + \ln A \quad (7)$$

where the activation energy E can be determined by the slope.

Activation energies can also be estimated using the Jump Method (Sorensen, 1992; Atkins et al., 2018), a technique commonly employed in chemical kinetics to measure very rapid reaction rates. In this approach, a reacting system initially at equilibrium is subjected to a rapid perturbation, and its relaxation back to equilibrium is subsequently observed. For temperature jump methods, the perturbation involves a sudden increase in temperature, altering the equilibrium constant, followed by the system's relaxation to equilibrium at the new temperature. The method considers the conversion derivatives at two consecutive isotherms at the same dataset, being valid if the function $f(\alpha)$ does not change through two subsequent isotherms. The related equation is:

$$\ln\left(\frac{\frac{d\alpha}{dt}_1}{\frac{d\alpha}{dt}_2}\right) = \frac{E}{R}\left(\frac{1}{T_2} - \frac{1}{T_1}\right) \quad (8)$$

where the subscripts 1 and 2 refer to the values at the end of an isotherm and the beginning of the subsequent one, respectively, being R the gas constant.

This method does not demand applying any rate model function whereas is very useful to detect mechanism transitions.

3. Results and discussion

The kinetic analysis was performed by plotting the reaction conversion according to eq. (1), with its corresponding derivative illustrated in Fig. 2. For the traditionally sized UAl₂ powder (Fig. 2A), a distinct change in the conversion rate is observed, marked by a minimum point at 2.6 h, indicating that the formation of UAl₃ is nearing completion. This result suggests that a heat treatment at 580 °C for 2.6 h is effective in fully consuming UAl₂ during the target fabrication process. The reaction to form UAl₄ continues at a lower conversion rate, reaching a maximum at 3.4 h, and is completed after approximately 5 h.

In the case of fine UAl₂ (<44 μm) (Fig. 2B), a much faster reaction is observed, with the nearly simultaneous formation of UAl₃ and UAl₄. The formation reaction of UAl₃ is completed after 2.4 h.

Arrhenius plots depicting the reaction rates determined at each isothermal condition for the traditionally sized powder are presented in Fig. 3, in accordance with the diffusion rate equation (3) through (5). The application of the one-dimensional diffusion equation (D1) yields unrealistic results (very high activation energy) within the higher temperature range and exhibits significant deviation when applied to the low-temperature fitting. Conversely, the diffusion equations D2 and D3 demonstrate superior accuracy, providing results that are more reliable.

To further investigate, the Jump Method (Sorensen, 1992; Atkins et al., 2018) can be utilized to select the most appropriate diffusion model. Table 2 demonstrates that the activation energy values determined via the jump method closely align with the results obtained from

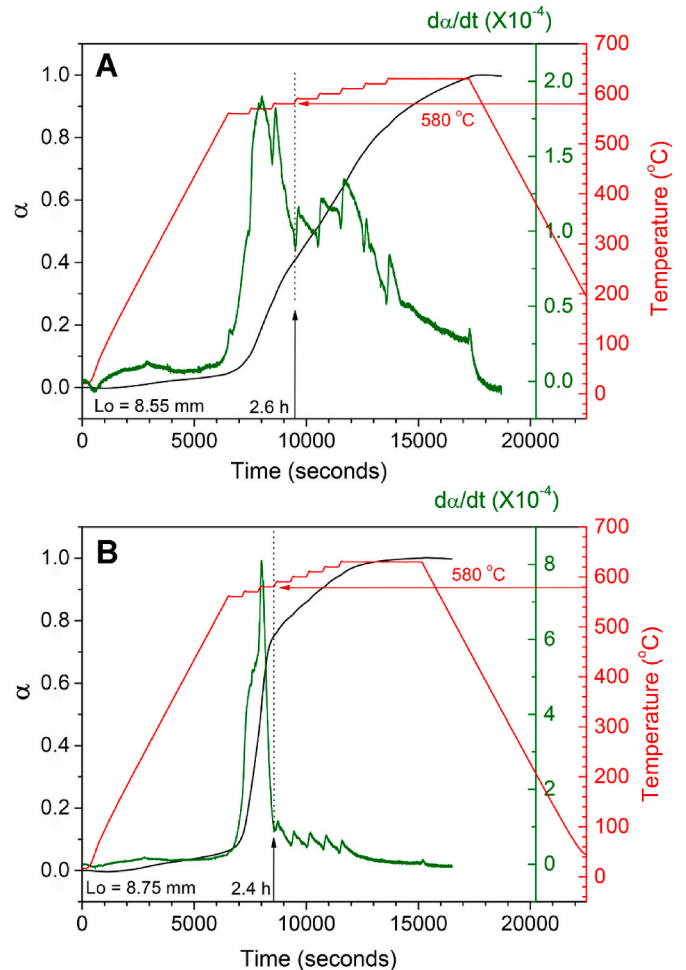


Fig. 2. Reaction conversion and derivative obtained by FSIA analysis A – traditionally sized powder (80 wt% 88-44 μm/20 wt% < 44 μm), 15-min isotherms. B – fine powder (100 wt% < 44 μm), 10-min isotherms.

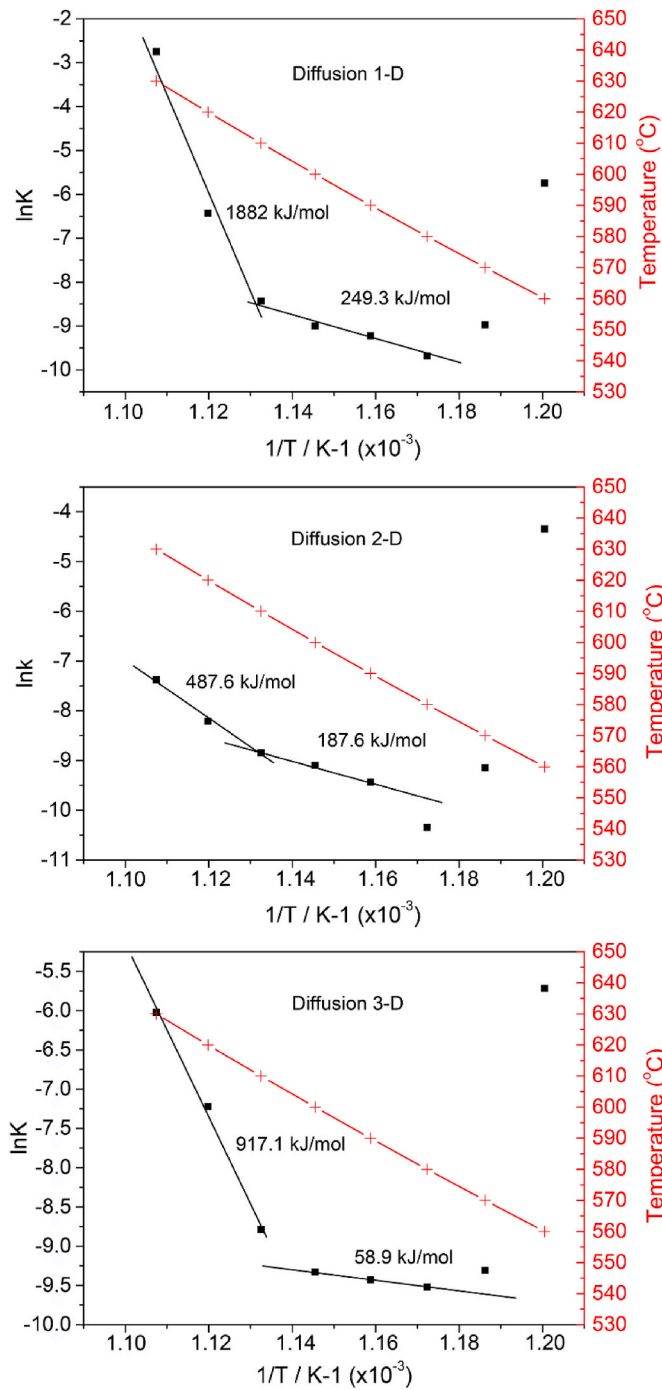


Fig. 3. Arrhenius plots for three diffusion rate controlling model determined by FSIA analysis (traditionally sized powder).

Table 2
Activation energies determined by the Jump Method for traditionally sized powder.

T1 (°C)	T2 (°C)	(dα/dt)1	(dα/dt)2	AE (kJ/mol)
560	570	9.92727E-5	1.73227E-4	325.0
570	580	1.61508E-4	1.71588E-4	36.2
580	590	1.00548E-4	1.10211E-4	56.2
590	600	9.72603E-5	1.19309E-4	128.0
600	610	1.10592E-4	1.32043E-4	113.6
610	620	8.90634E-5	1.07594E-4	123.9
620	630	6.10102E-5	7.72745E-5	158.4

the D3 diffusion model based on Arrhenius plots during the initial stages of the reaction. However, these values shift to higher levels at 590 °C, exhibiting greater concordance with the D2 model. This suggests a possible mixed reaction control involving both the D2 and D3 diffusion models.

The two techniques indicate a transition in the reaction mechanism around 600 °C, accompanied by a corresponding increase in activation energy. This phenomenon may be associated with the formation of a greater quantity of UAl₄, which possesses a defective crystal structure that complicates diffusion pathways, thereby potentially slowing down atomic diffusion, particularly for uranium atoms (Kniznik et al., 2011; Borgstedt et al., 1989).

The results for the fine UAl₂ powder sample are presented in Fig. 4 and Table 3. The three-dimensional diffusion model (D3) yielded inconsistent results at lower temperatures, whereas the two-dimensional diffusion model (D2) provided a closer fit to the data, yielding more reasonable activation energy values. In this scenario, the transition in the reaction mechanism is delayed to higher temperatures.

The jump method, while somewhat deviating from the model fitting values, shows alignment with the D2 model at 580 °C, with activation energies ranging from 97 to 98 kJ/mol. However, at higher temperatures, it begins to converge toward the behavior predicted by the D3 model, with values exceeding 200 kJ/mol. This suggests the operation of a mixed controlling mechanism involving both the D2 and D3 models, indicating that both diffusion processes may be occurring in parallel to some extent.

Activation energy values reported in the literature range from 130 to 180 kJ/mol for the UAl₂-Al system and from 200 to 220 kJ/mol for the

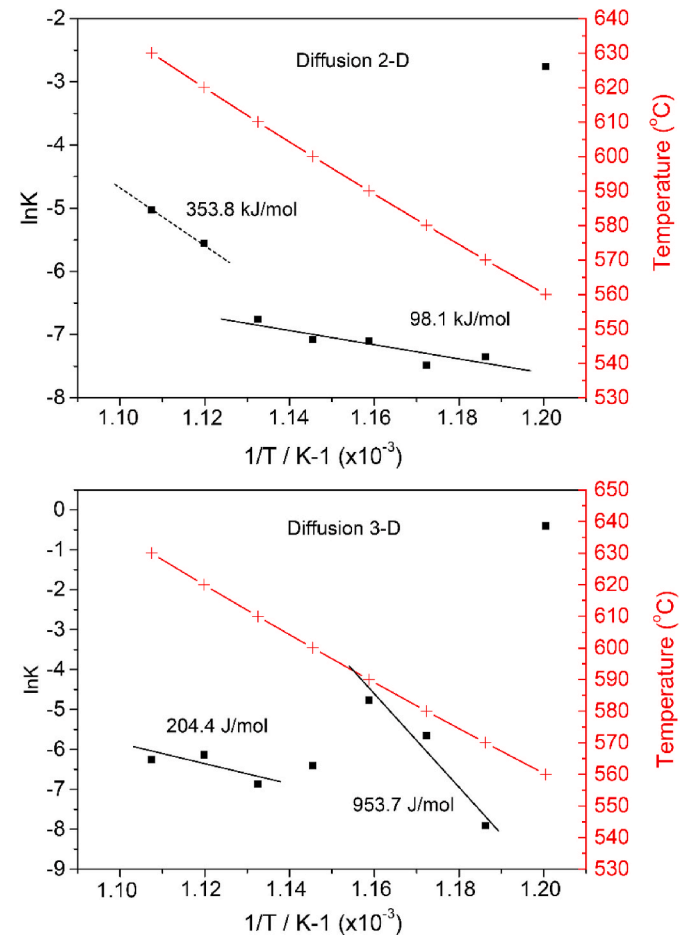


Fig. 4. Activation energies determined on 2-D and 3-D diffusion models of fine UAl₂ powder.

Table 3
Jump method derived activation energies of fine UAl₂ powder (<44 μm).

T1 (°C)	T2 (°C)	(dα/dt) ₁	(dα/dt) ₂	AE (kJ/mol)
560	570	1.71794E-4	3.21103E-4	365.2
570	580	5.6028E-4	7.52937E-4	176.7
580	590	8.95422E-5	1.04865E-4	96.7
590	600	5.45637E-5	8.44137E-5	273.3
600	610	5.14917E-5	7.40079E-5	232.5
610	620	4.67489E-5	6.92278E-5	257.4
620	630	3.66647E-5	5.68439E-5	294.0

UAl₃-Al system (Nazaré et al., 1975; Kniznik et al., 2011). In experiments utilizing traditionally sized powder, the activation energies derived from the D2 diffusion model are close to the first reaction range (130–180 kJ/mol), suggesting that the formation of UAl₃ is the rate-limiting step in the progression towards UAl₄. The results obtained by the jump method are even smaller. Low temperature values suggest the diffusion is easier and the UAl₃ growth is fast.

In the fine powder experiments, the consistently low activation energy values further support the notion that diffusion is enhanced by the increased surface area, interfacial contacts, and defects between particles, which serve to extend diffusion pathways. Overall, both experimental setups demonstrate more consistent results when applying the D2 diffusion model, particularly at lower temperatures. This suggests that the reaction rate at the interfaces significantly outpaces diffusion as the particles deviate from a spherical shape (Khawam and Flanagan, 2006).

4. Conclusions

The SID method can be employed for kinetic studies of solid-state reactions, specifically in the context of UAl₂-Al dispersed powder mixtures simulating nuclear fuel targets, by probing the reaction through linear expansion measurements in a dilatometer. The new method renders activation energy values close to literature information and describes the complex nature of Al diffusion into aluminides, where mixed control between D2 and D3 diffusion models is proposed.

The most accurate results were obtained using the D2 two-dimensional diffusion model, which indicates a rapid reaction rate and a non-spherical particle-matrix geometry. As the reaction progresses, the activation energy increases at higher temperatures, reflecting the near-completion of the UAl₃ formation and the subsequent slower reaction towards UAl₄. The inflection point observed in the traditionally sized powder experiments offers a critical basis for optimizing heat treatment protocols during target manufacturing.

CRedit authorship contribution statement

Thomaz Augusto Guisard Restivo: Writing – review & editing, Validation, Methodology, Formal analysis, Data curation, Conceptualization. **Giovanni de Lima Cabral Conturbia:** Validation, Methodology, Investigation. **Elita Fontenele Urano de Carvalho:** Visualization, Validation, Investigation. **Michelangelo Durazzo:** Writing – original draft, Validation, Supervision, Project administration, Formal analysis.

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.

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Data availability

Data will be made available on request.

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