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Data Article

X-ray diffraction and thermoanalytical datasets of precursors of the $Gd_6UO_{12-\delta}$ phase processed by combined mechanochemical–thermal routes

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ABSTRACT

The datasets presented here are related to the research paper entitled *"Disordered* $Gd_6UO_{12-\delta}$ *with the cation antisite defects prepared by a combined mechanochemical–thermal method"* [1]. The datasets complement the findings [1] on the effect of the combined mechanochemical–thermal processing of the stoichiometric mixture of solid precursors ($3Gd_2O_3 + UO_2$) on the formation of $Gd_6UO_{12-\delta}$ phase. In this article, we provide (i) X-ray diffraction (XRD) data of the $3Gd_2O_3 + UO_2$ mixture milled for 12 h, (ii) the refined XRD data of the non-milled $3Gd_2O_3 + UO_2$ mixture after annealing at 1282 °C for 3 h in air, and (iii) the thermogravimetric and differential thermal analysis (TG-DTA) data for non-milled and mechanically

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preactivated $3Gd_2O_3 + UO_2$ mixture measured in air at a heat rate of 10 K/min.

Specifications Table

Subject	Materials Science, Materials Characterization
Specific subject area	Nuclear material, Material for nuclear fuel forms, Material for nuclear waste immobilization, Structural analysis, XRD, Thermal analysis
Type of data	Graph
How data were acquired	XRD TG-DTA
Data format	Raw Analyzed
Parameters for data collection	XRD patterns of powdered samples were taken in the range from 10° to 80° (2Θ) (with angular step of 0.02°) using Cu Kα radiation and a C1702 diffractometer (Shimadzu, Japan) operating in Bragg-Brentano configuration. The TG-DTA was performed in air in the temperature range from 20 °C to 1282 °C at a heat rate of 10 K/min.
Description of data collection	The Rietveld refinement of XRD data was performed using the *FullProf Suite* software [2]. The ICSD database [3] was utilized for phase identification.
Data source location	Institution: Nuclear and Energy Research Institute, IPEN/CNEN-SP City/Town/Region: São Paulo Country: Brazil
Data accessibility	With the article
Related research article	G. Darin, K. Imakuma, R.T. Santiago, K.L. Da Silva, L.F. Cótica, M. Fabián, J. Valíček, H. Hahn, V. Šepelák, Disordered $Gd_6UO_{12-\delta}$ with the cation antisite defects prepared by a combined mechanochemical–thermal method, J. Nucl. Mater. doi: 10.1016/j.jnucmat.2021.152895

Value of the Data

- The datasets are important for a non-conventional mechanochemical preparation of $Gd_6UO_{12-\delta}$, which is less elaborative, solvent-free, and high-yielded in comparison with the conventional (thermal) synthesis of this nuclear material.
- The data provide the basis for the development of novel nuclear fuel materials.
- The XRD and TG-DTA datasets can guide other materials scientists toward designing novel U-Gd-O systems for nuclear fuel forms and nuclear waste immobilization.

1. Data Description

Fig. 1 shows the XRD pattern of the $3Gd_2O_3 + UO_2$ mixture milled for 12 h before heat treatment. During the high-energy ball milling process, Gd_2O_3 and UO_2 are subjected to a continuous fragmentation accompanied by the reduction of their crystallite size to the nanometer range. The latter is documented by relatively broad XRD peaks shown in Fig. 1. The qualitative XRD analysis reveals that milling of the mixture of individual binary oxides also leads to the *partial formation* of the ternary Gd_6UO_{12} phase. The early stage of mechanosynthesis of the GdU_6O_{12} phase is indicated in Fig. 1 by the presence of a broad XRD peak at the diffraction angle of about 30.6° (2Θ). During the early stages of high-energy milling the reaction precursors are mixed at the atomic level and the new mechanosynthesized phase nucleates in interfacial regions between the solid reactants during the impact period. During the subsequent thermal treatment

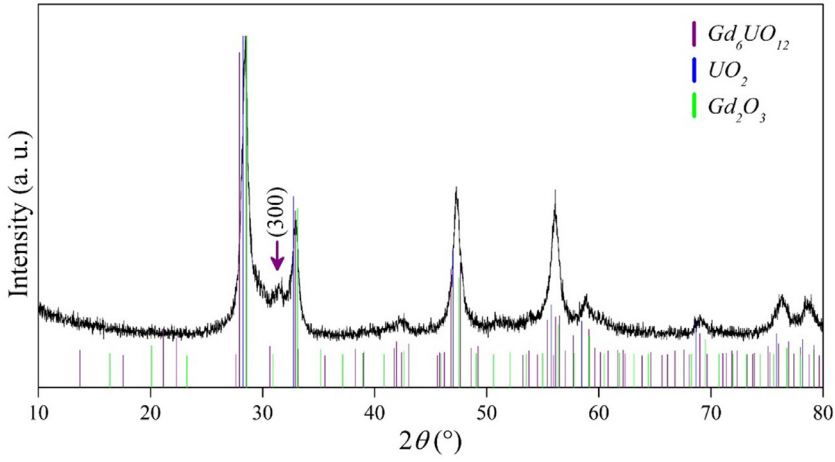

Fig. 1. XRD pattern of the $3Gd_2O_3 + UO_2$ mixture milled for 12 h before heat treatment. The *partial formation* of the Gd_6UO_{12} phase is evidenced by the presence of a broad XRD peak at the diffraction angle of about 30.6° (2Θ) indicated by arrow.

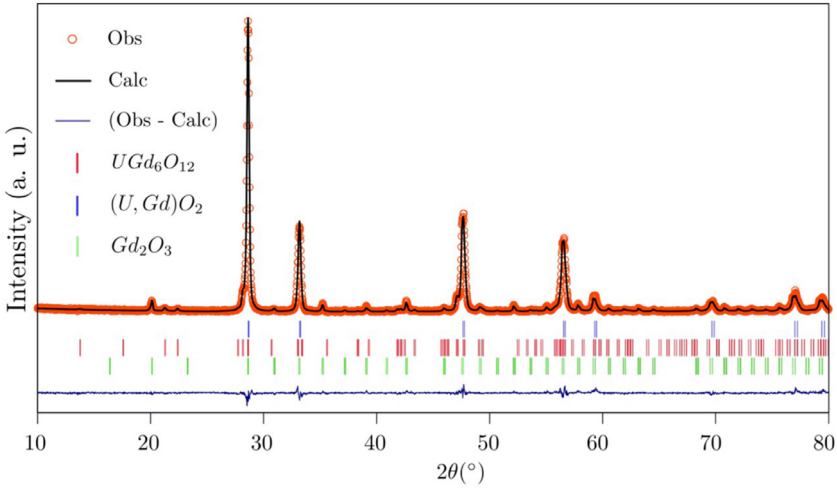

Fig. 2. The refined XRD pattern of the non-milled $3Gd_2O_3 + UO_2$ mixture after annealing at 1282 °C for 3 h in air. The following three phases are present in the sample: Gd_2O_3, $(Gd,U)O_2$, and Gd_6UO_{12}.

at 1573 K for 3 h, the *total conversion* of the precursors to the $Gd_6UO_{12-\delta}$ phase takes place [1]. Such a favorable formation of the uranate during the thermal treatment step is a consequence of an accelerated mass transfer and enhanced ionic diffusivity at contact zones between precursors due to reduced diffusion paths as a result of their mechanical preactivation and the partial mechanosynthesis of the target phase.

Fig. 2 presents the refined XRD pattern of the non-milled $3Gd_2O_3 + UO_2$ mixture after annealing at 1282 °C for 3 h in air. The data document that the solely thermal processing of the mixture does not lead to the complete transformation of educts to the final Gd_6UO_{12} phase. The quantitative analysis of XRD data reveals the presence of the following three phases in the sample: 23 wt.% of Gd_2O_3 (ICSD collection code: 230,768), 66 wt.% of $(Gd,U)O_2$ (236,072), and 11 wt.% of Gd_6UO_{12} (21,945) [3].

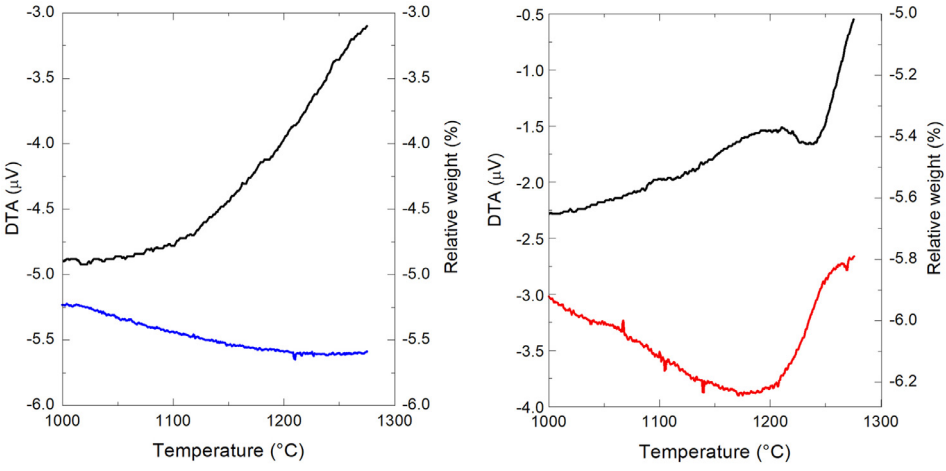

Fig. 3. (Bottom) TG and (top) DTA curves for (left) non-milled and (right) mechanically preactivated $3Gd_2O_3 + UO_2$ mixtures measured in air at a heat rate of 10 K/min.

Fig. 3 shows the TG-DTA curves for non-milled and mechanically preactivated $3Gd_2O_3 + UO_2$ mixture measured in air at a heat rate of 10 K/min. At about 1200 °C, a significant increase of the relative weight for the mechanically pre-treated mixture is attributed to its oxidation resulting in the complete formation of Gd_6UO_{12}; *i.e.*, the reaction $3Gd_2O_3 + UO_2 + ½O_2 \rightarrow Gd_6UO_{12}$ proceeds. This is in contrast to the TG curve of the non-milled mixture exhibiting only a slight increase at temperatures above 1250 °C. The latter is associated with the initial stages of the conventional (thermal) synthesis of Gd_6UO_{12}.

2. Experimental Design, Materials and Methods

The synthesis of the Gd_6UO_{12} solid phase requires the presence of two solid simple oxide precursors in the stoichiometric composition ($3Gd_2O_3$ and UO_2) and one gas phase (oxygen); *i.e.*, the overall heterogeneous chemical reaction leading to the formation of Gd_6UO_{12} can be written as $3Gd_2O_3 + UO_2 + ½O_2 \rightarrow Gd_6UO_{12}$. At first, the stoichiometric mixture of UO_2 and Gd_2O_3 was preactivated by ball-milling in a Pulverisette 6 mill (Fritsch, Germany) for 12 h at 300 rpm in N_2 atmosphere at ambient temperature. The milling process was interrupted every 1 h for 5 min (the total number of breaks: 11). A grinding chamber and balls made of stainless steel were used. The ball-to-powder mass ratio was 11:1. The UO_2 and Gd_2O_3 precursors used for the synthesis of Gd_6UO_{12} are products of Nuclear Materials Laboratory at the Industrial Nuclear Center of Aramar (São Paulo, Brazil) and the firm Alfa Aesar, respectively. The mixtures of non-milled and mechanically treated precursors were pressed into pellets at 390 MPa, which were subsequently heated for 3 h at 1573 K in air. XRD patterns of powdered samples were taken in the range from 10° to 80° (2Θ) (with angular step of 0.02°) using Cu Kα radiation and a C1702 diffractometer (Shimadzu, Japan) operating in Bragg-Brentano configuration. The Rietveld refinement of XRD data was performed using the *FullProf Suite* software [2]. The ICSD database [3] was utilized for phase identification. The TG-DTA measurements were performed using a simultaneous thermal analysis apparatus STA 409 C/7/E (Netzsch, Germany). The TG-DTA was done in the temperature range from 20 °C to 1282 °C at a heat rate of 10 K/min. The pelletized samples (700 mg) of both non-milled and mechanically preactivated $3Gd_2O_3 + UO_2$ mixtures were thermally treated in open platinum crucibles in air.

CRediT Author Statement

Gaspar Darin: Conceptualization, Methodology, Software, Visualization; **Kengo Imakuma:** Investigation, Formal analysis; **Rafael Trautwein Santiago:** Visualization, Investigation; **Klebson Lucenildo Da Silva:** Conceptualization, Supervision, Formal analysis, Resources, Writing - Original Draft, Writing - Review & Editing; **Luiz Fernando Cótica:** Formal analysis, Supervision; **Martin Fabián:** Investigation, Formal analysis; **Jan Valíček:** Investigation, Visualization; **Horst Hahn:** Supervision; **Vladimír Šepelák:** Supervision, Writing - Original Draft, Writing - Review & Editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships which have or could be perceived to have influenced the work reported in this article.

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Supplementary Materials

Supplementary material associated with this article can be found in the online version at doi:10.1016/j.dib.2021.106972.

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