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Phase quantification in UAl_x-Al dispersion targets for Mo-99 production

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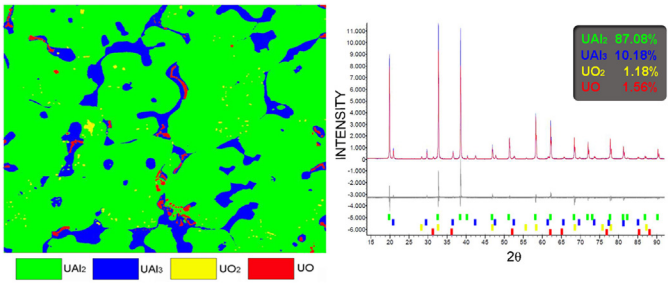
HIGHLIGHTS

- Image analysis and X-ray diffraction methods for phase quantification of UAl_x phases.
- Mapping UAl_x phase composition during target fabrication.
- Following UAl_2 transformation during rolling UAl_x-Al dispersion targets.

GRAPHICAL ABSTRACT



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ABSTRACT

Uranium aluminide (UAl_x) is a mixture of three distinct intermetallic compounds comprised of UAl_2, UAl_3 and UAl_4, where the "x" is used to denote a mixture of those phases. Usually UAl_x is formed during the target fabrication process by means of a solid state reaction between the uranium aluminide and aluminum. Quantitative techniques such as image analysis and X-ray diffraction using the Rietveld method were compared for their applicability in the determination of the UAl_2, UAl_3 and UAl_4 concentrations, both in the UAl_2 primary ingot and in the UAl_x-Al dispersion. The UAl_x composition was quantified in all stages of the target manufacturing. The image analysis method was shown to be useful for UAl_x phase quantification in the primary UAl_2 ingot, but was not applicable in the case of UAl_x-Al dispersions. The X-ray diffraction method allowed the quantification of the existing UAl_x phases in both the primary ingot and UAl_x-Al dispersions. Possible sources of error are discussed. The method of quantification based on X-ray diffraction was shown to be appropriate to monitor the evolution of UAl_x phases during the manufacturing process.

1. Introduction

Every year the world demands more than 30 million medical imaging procedures that use the technetium-99 m radioisotope (Tc^{99m}), which correspond to approximately 80% of all nuclear medicine diagnoses [1]. This radiopharmaceutical product stems from the radioactive decay of molybdenum-99 (Mo^{99}), which is commercially produced in research reactors by irradiating targets that contain uranium-235. However, continuous supply of Mo^{99} has decreased over the last decade, mainly due to shutdowns that have occurred in the main research reactors that produce radioisotopes [1]. To deal with this scenario, Brazil has decided to build a

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multipurpose reactor which among other functions will irradiate uranium targets to produce enough Mo-99 to meet domestic demand [2,3].

There are currently two technologies available to produce uranium targets. One is based on a uranium-aluminum compound dispersed in an aluminum matrix [4–9] and the other one is based on metallic uranium thin foils [10–14]. The dispersed targets are the most used worldwide for commercial production of molybdenum-99 b y nuclear fission. The targets use low enriched uranium (LEU) and are fabricated according to traditional technology based on the picture-frame technique [15–17], which is adopted for commercial production of fuel elements for nuclear research reactors. Because of the experience acquired over the years in the manufacturing of dispersion fuel elements, it was decided to implement this technology to fabricate UAl_x-Al dispersion targets for future molybdenum-99 production in Brazil.

The binary system, uranium and aluminum, forms a phase diagram which shows the existence of intermetallic compounds consisting of three phases, namely UAl_2, UAl_3, and UAl_4. The mixture of these phases is known in the literature as UAl_x [18]. It has been reported that UAl_3 and UAl_4 are more easily dissolved in alkaline solutions than UAl_2, which ultimately defines the radiochemical processing yield after the irradiation [19]. Therefore it is desirable for UAl_3 and UAl_4 phases to be present in the final target as they show good basic dissolution behavior during the subsequent radiochemical processing.

UAl_2 has been used as starting material for the fabrication of the irradiation targets [4–9]. The use of UAl_2 instead of other aluminides offers advantages in terms of its synthesis since the UAl_2 has a congruent melting point and as a result it can be synthesized in a single step, requiring no post-synthesis annealing. UAl_3 and UAl_4 have incongruent melting points and thus are formed through peritetic reactions, requiring long thermal treatments to complete synthesis [18,20]. Moreover, UAl_2 has higher U-content (81.52 wt% U) and density (8.14 g/cm^3) than the other uranium aluminides and therefore it maximizes the uranium-235 content in the target.

UAl_x-Al dispersion targets are fabricated by hot-rolling according to the traditional picture-frame technique [15–17] as small aluminum plates containing a UAl_x-Al dispersion meat. During fabrication UAl_2 reacts with aluminum to form UAl_3 and UAl_4 during the thermal-mechanical and annealing processes [5,9].

An important requirement for the target is the limitation of the UAl_2 content in the finished target, which should preferably to be zero. As already mentioned, this limitation is due to the fact that the uranium aluminides have different chemical properties with respect to their dissolutions in basic medium for the extraction of molybdenum-99 after irradiation. For example, Cols et al. [19]. Reported that UAl_3 and UAl_4 dissolve more easily in alkaline medium than UAl_2, and this defines the yield of radiochemical processing for Mo-99 extraction.

On the one hand, UAl_2 is used as starting material to fabricate the target. On the other hand, it must be totally consumed at the end of thermal mechanical processing. Therefore the UAl_2 assay must be quantified before and after thermal processing to ensure that all UAl_2 has been consumed. Thus, characterizing the phase composition in the starting material and UAl_x dispersions is important to assess the UAl_2 transformation during target fabrication, so that a thermo-mechanical treatment can be developed to ensure that all UAl_2 is consumed during manufacturing and will not be present in the final target.

This work aims to investigate methods to quantify the phases present in UAl_2 ingots and also in UAl_x-Al dispersion targets. Image analysis and X-ray diffraction were assessed as two possible methods.

2. Experimental procedures

The uranium dialuminide (UAl_2) was prepared by induction melting method and the uranium and aluminum were weighed-in stoichiometrically (81.5 wt% U) [20]. Then, the metals were charged into a zirconium crucible and melted using a 15 kW induction furnace. Prior the melting, the furnace was purged with argon after vacuum of 2.6×10^{-3} mbar. The UAl_x ingot was ground in a mortar under argon atmosphere. The grinding product was sieved in 8-in. (204 mm)-diameter stainless steel sieves. Three particles sizes were separated: +170 mesh (>88 μm), −170 + 325 mesh (<88 μm and >44 μm), and −325 mesh (<44 μm). The fuel powders used in the fabrication of targets were 80%−170 + 325 mesh and 20% −325 mesh. The density of the UAl_2 powder was determined to be 8.13 ± 0.01 g/cm^3 (triplicate) by helium pycnometry. The uranium content was determined to be 80.74 ± 0.02 wt% (triplicate) by chemical analysis [21,22].

Mixtures of aluminum and UAl_2 powders corresponding to 50 and 45% in volume respectively were homogenized for 1 h in a blender (Turbula T2F) and then compacted under pressure of 490 MPa to produce briquettes with porosity around 5 vol%. After compacting, the briquettes were degassed at 250 °C for 3 h under vacuum (5.10^{-3} Pa). Table 1 presents the typical characteristics of briquettes.

The briquettes were assembled into aluminum picture frames (4.20 mm thick) which were clad with two aluminum plates (2.86 mm tick). The assemblies were TIG (Tungsten Inert Gas) welded together and then rolled to form the targets, according to the picture-frame technique [15–17]. The assemblies were hot-rolled in six rolling passes. The final thickness for the target was reached through the cold-rolling pass. Fig. 1 illustrates the main

Table 1
Main typical characteristics of briquettes.

	Dimensions (mm)	22.16 × 22.15 X 4.20 (thickness) (with rounded corners/R = 3.0 mm)
Briquette	Mass (g)	9.67
	Volume (cm^3)	1.94
	Mass (g)	2.65
Aluminum	Mass fraction (%)	27.4
	Volume (cm^3)	0.98
	Volume fraction (%)	50.5
	Mass (g)	7.02
UAl_2	Mass fraction (%)	72.6
	Volume (cm^3)	0.86
	Volume fraction (%)	44.3
Pores	Volume (cm^3)	0.10
	Volume fraction (%)	5.2