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Evaluation of the feasibility of the powder injection moulding process for the fabrication of nuclear fuel and comparison of several formulations

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ABSTRACT

Six forming aid formulations were used to assess the nuclear fuel elaboration by means of the powder injection moulding (PIM) process. These formulations were especially designed for actinide powders (UO_2 and MOX) by taking into account the specificities of these powders, in particular any solutions to prevent radiolysis. Once the feedstocks with the UO_2 powder had been prepared, the thermal studies showed no significant endothermic or exothermic reaction, which demonstrates the quasi-independent behaviour of the polymer system with respect to the uranium dioxide powder. The mass of the carbon residues – a key criterion in selecting a formulation – was below 150 ppm for all six formulations, which is still sufficient for obtaining net-shaped pellets following the sintering process. The formulations to be recommended for the nuclear fuel application are those containing polystyrene as they achieve satisfactory deagglomeration combined with an adequate level of injectability (low shear viscosity of the system). Moreover, they are theoretically resistant to radiolysis phenomena due to the benzene rings of the polystyrene.

1. Introduction

Within the frame of the prospective emergence of Generation IV nuclear reactors, processes for fuel manufacturing are reconsidered. Indeed, the current fabrication process may hinder innovation in the field of future nuclear fuels if it is not modified or at least completely reviewed [1]. The expected improvements particularly concern 1) simplifying the current process to reduce the radiological impact on operators, 2) consolidating the robustness of the process with respect to shape requirements and to dimensional tolerances, and 3) enhancing the production rate. Using an injection moulding process to manufacture nuclear fuel is being considered in this specific context.

Outside the nuclear industry, the powder injection moulding (PIM) process is used for the mass production of net-shape parts at low cost [2]. This process also has the advantage of being able to produce metal [3] and ceramic parts [4] with complex, variable geometries with an accurate control of dimensional tolerances [5]. The PIM process involves hot filling a tool (mould) of the desired shape with a mixture of the forming aids and powder [6]. The mixture, referred to as 'feedstock', is cooled and then the forming aids are removed (debinding step) in order to start the densification process through the diffusion in solid state [7,8].

The main objective of using the injection moulding process is to simplify the current process. Firstly, the fact of introducing forming aids would make it possible, upstream, to restrict the powder preparation operations (crushing, mixing, etc.) and to eliminate the dispersal of contaminating and irradiating powder (reducing the radiological exposure of operators). Moreover, the possibility of directly manufacturing net-shaped pellets after the sintering step is a major advantage which would suppress the grinding step involving post-sintering abrasion, a very impacting process from a radiological viewpoint in the nuclear field. Lastly, the process offers a wide range of applications, whether in terms of the shape of the fuel or the chemical nature of the powders to be implemented. The PIM process therefore provides advantages as regards flexibility with respect to new designs, and therefore increases the number of innovation possibilities.

However, using actinide powders in the PIM process involves studying certain scientific subjects which represent a number of potential hurdles to overcome. The latter are directly related to the three main specificities of these powders. Firstly, the morphological characteristics of the actinide powders to be implemented are relatively different from the characteristics of the powders generally used in this process [8,9]. Moreover, the potential chemical interactions between the powder and the forming aids must be identified and their effects must be determined. Lastly, the impact of the radiolysis of the forming aids (modification of their properties under the effect of ionising radiation) must be precised. The purpose of the work described in this paper is therefore to determine the feasibility of the powder injection moulding process applied to the manufacture of nuclear fuel through the development of forming aid formulations adapted to the specificities of actinide powders.

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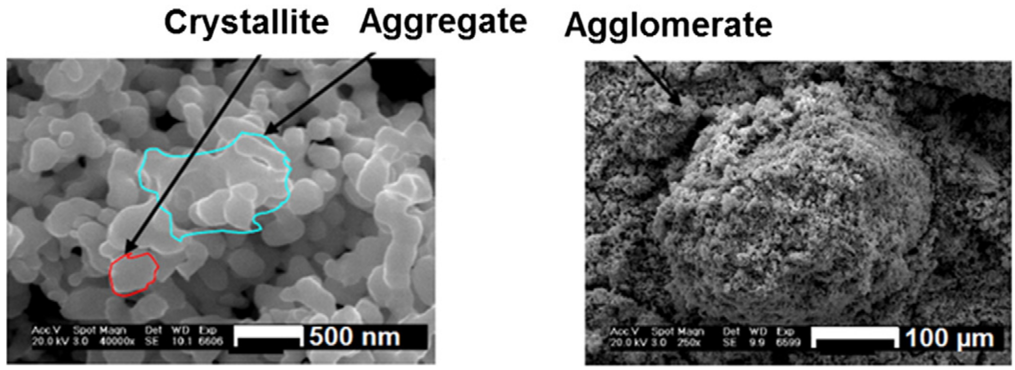

Fig. 1. SEM micrographs of the UO_2 used in the study.

2. Methodology — materials

2.1. Methodology

The issue at hand calls for a methodology based on an accurate scientific approach making it possible to process the three specificities of the actinide powders mentioned below.

2.1.1. Specificity 1: morphology of actinide powders

At present, the characteristics of actinide powders intended for future fuels have not been defined in a definite manner as fabrication processes may change. However, the characteristics of powders used for preparing the feedstock remain a key decisive element. Therefore, understanding how they affect the process is a major issue for PIM.

To do so, simulating alumina powders broadly covering the morphological characteristics of actinide powders have been implemented with a reference forming aid formulation. Results obtained by J. Bricout [10] revealed their suitable behaviour with respect to the injection moulding process on the following levels: preparation of highly loaded mixtures (>50 vol.%), rheological behaviour (shear-thinning) and debinding/sintering capacity.

This study aims to validate these conclusive results with actinide powders. To do so, uranium dioxide powder has been used, since it is the main component of the various oxide fuels currently employed. The possibility of preparing homogeneous highly loaded feedstocks and the rheological behaviour of the forming aid/UO_2 mixtures are analysed herein.

2.1.2. Specificity 2: chemical interactions between UO_2 and the forming aids

The actinide powder was in contact with organic products from the mixing step up to the debinding step. The mixing and injection steps were performed at a temperature of around 180 °C, while the debinding step was performed at around 600 °C. Several types of interactions can occur between the actinide powder and the forming aid throughout the process.

An indirect assessment based on a comparative approach was implemented to understand these interactions. For this purpose, each component was studied independently and then in the feedstock. We could therefore i) determine the influence of UO_2 on the behaviour of the feedstock at the temperatures implemented during the process (potential reactions), ii) characterise the UO_2 powder following the debinding step (observed phases, alterations of the fissile phase), and iii) characterise the material following the sintering step (density, carbon residues, etc.).

2.1.3. Specificity 3: radiolysis

Radiolysis is defined as the decomposition of organic matter under the effect of ionising radiation from the actinide powders (in our case). According to the chemical nature of the forming aids, such ionising radiation has a different impact.

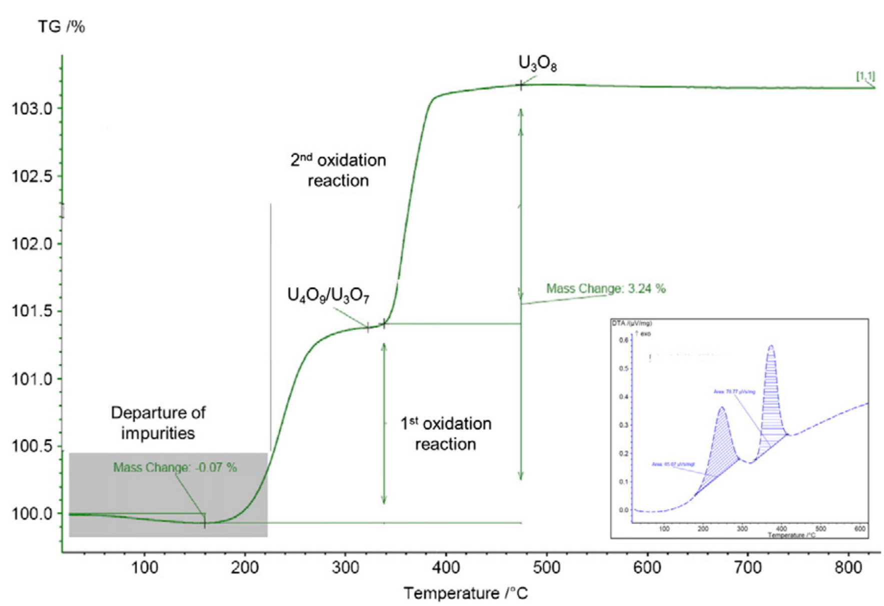

Fig. 2. Thermogramme and differential thermal analysis of UO_2 in a reconstructed air atmosphere (ramp: 10 °C/min up to 900 °C).