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Investigation of the mechanical properties of ZrO_2-doped UO_2 ceramic pellets by indentation technique

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HIGHLIGHTS

- Formation of solid solution between ZrO_2 and UO_2 up to 20 wt% of ZrO_2 content.
- The average grain sizes of $(U, Zr)O_2$ ceramics declined with the addition of ZrO_2.
- Hardness and elastic modulus of $(U, Zr)O_2$ increased with the addition of ZrO_2.
- Fracture toughness of UO_2 was considerably decreased by the addition of ZrO_2.

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ABSTRACT

ZrO_2-doped UO_2 ceramic pellets have been identified as a high performance nuclear fuel in LWR due to its desirable properties. Mechanical properties such as hardness, elastic modulus and fracture toughness, as well as microstructure of ZrO_2-doped UO_2 ceramics containing 0, 10, 15 and 20 wt% ZrO_2 were investigated by Vickers micro-indentation technique, X-ray diffraction and scanning electron microscopy, respectively. The lattice parameter of the ZrO_2-doped UO_2 ceramics linearly decreases as a function of the ZrO_2 content and follows Vegard's law, indicating the formation of a complete solid solution between the ZrO_2 and UO_2 up to 20 wt% of the ZrO_2 content. The average grain size was typically between 20 and 30 μm with no dopant additions and gradually decreased to about 10 μm for additions of 20 wt% ZrO_2. Additionally, the addition of ZrO_2 will significantly increase the hardness and elastic modulus of UO_2 ceramic by approximately 16.4% and 14.5% with the ZrO_2 contents increasing from 0 to 20 wt%, respectively. Furthermore, the fracture toughness of UO_2 ceramic was considerably decreased by the addition of ZrO_2 dopant, which is evidently contrary to the hardness and elastic modulus.
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1. Introduction

As one of the most widely used materials for nuclear fuel in light water reactor (LWR), uranium dioxide (UO_2) has been the subject of numerous theoretical as well as experimental studies. To improve the thermo-physical, mechanical and chemical properties of fuel matrix and reduce fission gas release as well as swelling during irradiation of the fuel pellets, many different dopants such as oxides of titanium [1–3], niobium [4–6], magnesium [7,8], chromium [9–12], and zirconium [13–18] have been investigated in the past few decades. The solid solutions of ZrO_2-doped UO_2 ceramic pellets have been identified as a high performance nuclear fuel in LWR due to its desirable properties which include [13–18]: (1) the corrosion resistance of the ZrO_2-doped UO_2 fuel pellets in the high-temperature water or steam is very excellent, (2) the thermal conductivity of ZrO_2-doped UO_2 fuel pellet remains relatively unchanged during irradiation, even though it is somewhat lower compared to the pure UO_2 fuel pellet, and (3) the swelling rate of ZrO_2-doped UO_2 fuel is lower compared to the pure UO_2 fuel during the in-pile irradiation.

Knowledge of mechanical properties of nuclear fuel is of importance for fuel design and manufacturing, as well as for the understanding of the fuel behavior during irradiation [19–23]. For example, fractures develop in fuel pellets during the in-pile irradiation due to the radial temperature gradients, which induce high

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thermal stresses [24,25]. The occurrence of fractures affects the dimensional stability, especially the fission gas release in fuel pellets. Hence, a fundamental understanding of the mechanical properties of a potential nuclear fuel is very important for the safe and efficient utilization of nuclear energy. However, to the best of our knowledge, although the microstructure, thermal properties and irradiation behavior have been successfully studied by a number of techniques over a wide range of compositions and temperatures in the past few decades [13–17,26,27], available literature data on mechanical properties of ZrO_2-doped UO_2 solid solution fuel is very scarce.

The micro-indentation technique has been developed in some decades and is used worldwide for the mechanical properties assessment of material since it can yield much more useful information of mechanical properties than the common hardness testing [28–33]. In particular, this technique is very useful in the field of nuclear materials when the sizes of available samples are limited since it needs only a small portion of smooth surface. However, the Vickers indentation fracture method, which has been widely used for determining fracture toughness of ceramics, has been subject of debate in these years [34], although it has been adopted in the American standard specification for ceramic bearing balls [35]. They claimed that the Vickers indentation fracture technique no longer be acceptable for the fracture toughness measurement of ceramic or other brittle materials since in between-laboratory consistency was poor [34,36], which was revealed by round robin tests on silicon carbide conducted in order to standardize the indentation fracture toughness test for ceramics [37,38]. However, almost three decades have passed since the last round robin tests. Performance of the ceramic materials has made a grade progress during the past few years and the indentation fracture measurement equipments have been also refined. It is likely that the accuracy and effectiveness of the indentation fracture test are improved as compared with those reports in the literature. Additionally, reproducibility of the indentation fracture test on the silicon nitride ceramics has been checked in recent years, and suggested that the Vickers indentation fracture test could provide reliable results if the magnification of microscope is suitable for the indentation load [39].

Therefore, the objective of this study is to identify the effect of the ZrO_2 content on the mechanical properties, such as hardness (H), elastic modulus (E) and fracture toughness (K_{IC}) of UO_2 fuel using the Vickers micro-indentation technique with the ZrO_2 contents ranging from 0 to 20 wt%. This study will help for studying the mechanical properties of ZrO_2-doped UO_2 ceramic fuel in detail and supplement the available experimental data for the fuel design.

2. Experimental procedures

In the present work, UO_2 powder produced by the conventional ammonium diuranate (ADU) conversion process was used. The O/U ratio of UO_2 powder was determined by Thermo Gravimetric Analysis (TGA) and was 2.04, the purity of the powder was 99.99%, with the major impurities being C and F elements. As a dopant ZrO_2 powder was used, and the purity of the powder was 99.99%. Four compositions of the ZrO_2-doped UO_2 fuel pellet samples with various ZrO_2 contents (0, 10 wt%, 15 wt%, and 20 wt%, respectively) were prepared by mixing of the UO_2 and ZrO_2 powders. To enhance the sinterability, the powder mixtures were further milled for 8 h using a planetary mill. The milled powder mixtures were dried in a baking box and then pressed into green pellets under a duplex axial pressure of about 300 MPa, followed by reactive sintering at 1973 K in a H_2-Ar gas flow atmosphere for 4 h.

To examine the purity and identify the constituent phases as well as determine the lattice parameters of the sintered ZrO_2-doped UO_2 fuel samples, X-ray diffraction (XRD) analysis using a powder X-ray diffractometer (Brucker, AXS, Germany) using Cu-Kα radiation in air at room temperature was performed. The density and porosity of the sintered samples were determined by the Archimedes method. After ceramographic preparation, sintered ZrO_2-doped UO_2 fuel samples were chemically etched in a solution of 50 g NH_4HF_2, 50 ml H_2O for 50 s in air atmosphere to reveal grain boundaries and grain size. The etched surfaces of the sintered samples were examined using a scanning electron microscope (SEM, JSM-6700 F). The grain sizes of the sintered ZrO_2-doped UO_2 fuel samples were statistical analyzed by a linear intercept method.

The mechanical properties of the sintered ZrO_2-doped UO_2 fuel samples were evaluated by Vickers micro-indentation testing. An MHT micro-indenter (CSM Instruments SA, Peseux, Switzerland) with a Vickers diamond indenter was used. To avoid the influence of the chemical etching on the indentation test results, the ZrO_2-doped UO_2 ceramic samples were re-grinded and polished before measuring the mechanical properties. Afterwards, the indentation load versus displacement curves were measured, and the data on hardness (H) and elastic moduli (E) were analyzed using the method proposed by Oliver and Pharr [30], which is widely used to determine the mechanical properties of ceramic materials.

The indentation fracture toughness (K_{IC}) of samples was calculated according to the equation proposed by Kapoor [40] and Lawn [41,42]. Kapoor [40] had identified the type of formed cracks in UO_2 ceramic is the half-penny crack configuration and proposed that the fracture toughness values can be calculated using Eq. (1).

$$K_{IC} = \delta \left(\frac{E}{H}\right)^{1/2} \frac{P}{c^{3/2}} \tag{1}$$

where, δ is an empirical calibration constant equal to 0.016, E is the elastic modulus (GPa), H is the hardness of sample (GPa), P is the applied load (N) and c is the half-length of the crack (m).

In addition, before the actual Vickers indentation tests were performed, the stability of the measurements was checked by applying a series of variable loads (range from 10 to 500 mN) using a continuous stiffness measurement (CSM) module. Considerations of that about 60 mN and 300 mN were shown to be suitable for determining the elastic modulus (contain hardness) and fracture toughness, respectively. During the indentation testing, the loading and unloading rates were maintained at 2 mN s^{-1} and a maximum load of 60 mN was used with a hold time at maximum load of 10 s when determining the hardness and elastic modulus, while the maximum load was replaced by 300 mN and maintaining the other parameters the same when assessing the fracture toughness of ZrO_2-doped UO_2 ceramic samples.

3. Results and discussion

The densities of ZrO_2-doped UO_2 ceramic samples were measured by the Archimedes' method and calculated as a percentage of the theoretical density are summarized in Table 1. All density measurements were performed in triplicate and the standard errors are calculated at the same time. It is indicated that the dense pellets with the various ZrO_2 contents were obtained after sintering at 1973 K for 4 h with relative density values in between 96 and 98%.

The X-ray diffraction patterns of all pure and ZrO_2-doped UO_2 ceramic samples are shown in Fig. 1, together with the peak positions of pure UO_2 [10]. In all the XRD patterns, ZrO_2 or other impurities peaks are not observed, indicating the Zr completely dissolved in UO_2 and formed solid solutions of (U, Zr)O_2. It was observed that the peak positions regularly shifted to higher angles

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