Available online at www.sciencedirect.com

SCIENCE DIRECT®

Surface & Coatings Technology 196 (2005) 63–68

SURFACE & COATINGS TECHNOLOGY

www.elsevier.com/locate/surfcoat

On the use of ion beams for the selection of radioactive waste matrices

Frédérico Garrido*, Aurélie Gentils, Lionel Thomé

Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse, CNRS-IN2P3-Université Paris-Sud, Bâtiments 104-108, Orsay Campus 91405, France

Available online 27 October 2004

Abstract

The safe and long-term control of radioactive waste arising from nuclear power plants is one of the major challenges to the future use of nuclear energy. The evaluation of nuclear waste matrices relies on numerous conditions regarding the physico-chemical properties of the selected materials, such as resistance against oxidation or aqueous corrosion, stability in a radioactive environment, and ability to confine radioactive elements. Ion beams provide efficient tools for the evaluation of radwaste matrices since they allow to address three major issues: (i) the simulation of ion irradiation; (ii) the doping of the matrix with stable elements simulating the nuclei to be confined; and (iii) the characterization of the material by the use of nuclear microanalysis techniques. Illustrative examples are provided in the case of urania and zirconia.

Keywords: nuclear waste; ceramic; ion irradiation; retention capacity; ion-beam analysis

1. Introduction

An important issue conditioning the development of nuclear energy is the safe disposal of high-level nuclear waste. One of the current strategies is based on the so-called multibarrier concept, which consists in encapsulating the waste in a solid form (first barrier) and burying it in a deep geological repository (the host rock constitutes the geological barrier). Due to the presence of long-lived radiotoxic nuclides, mostly constituted of alpha-emitting transuranium elements, the contamination of the biosphere should be hindered for periods exceeding several thousands of years. The contamination vector is the underground water and the first step towards the dissemination of radionuclides is the aqueous corrosion of the waste solid form. As the disposal matrix is subjected to an intense self-irradiation, the possible deleterious effects of radiation damage on the chemical durability of the waste form are of prime importance.

Based on the main objective of insuring the long-term confinement of radiotoxic elements, several classes of solid matrices were identified as potential candidates for the incorporation of nuclear waste, depending on management strategies [1–4]. In countries like France and Japan, which rely on fuel reprocessing, the high-level waste is mixed with a borosilicate glass. In others countries like Sweden and the United States, the spent nuclear fuel is intended for direct disposal and it acts as the first barrier towards the release of radiotoxic elements. Recently, the emergence of alternative strategies with the ultimate goal to reduce the amount of high-level waste that is produced, such as the burning of actinides produced during the reactor operation in a so-called "inert fuel matrix," gave a new impetus to the search of advanced waste forms (mostly crystalline ceramics).

In this general framework, ion beams provide efficient tools for the selection of radioactive waste matrices and their qualification as disposal or transmutation matrices. Indeed, they allow to address three major issues: (i) the simulation by external ion irradiation of the radiation damage, which alters the crystalline structure of the matrix; (ii) the doping of the material with stable or radioactive elements, which simulates the species to be confined; and (iii) the characterization of the material via nuclear microanalysis techniques. The purpose of the present article is to give illustrative examples related to the first two items in relation with ion beam characterization techniques.

* Corresponding author. Tel.: +33 1 69 15 52 57; fax: +33 1 69 15 52 68.

E-mail address: garrido@csnsm.in2p3.fr (F. Garrido).

0257-8972/$ - see front matter © 2004 Elsevier B.V. All rights reserved.
doi:10.1016/j.surfcoat.2004.08.097

2. Simulation of radiative environment

The radiation stability of the material is one of the most important criteria for the selection of specific nuclear waste matrices. Energetic ion beams simulate: (i) the alpha decay of actinide elements (i.e., the production of an alpha particle of 5 MeV typical energy and the concomitant generation of a heavy recoil nucleus with an energy of ~100 keV); and (ii) the slowing down of fission fragments [i.e., medium and heavy elements (mass number ranging from 80 to 155) energy in the range 70–100 MeV]. Fig. 1 sketches the typical ranges and defect densities of the above-mentioned sources of radiation in zirconia (zirconium dioxide ZrO_2) calculated with Monte Carlo simulations [5,6]. Alpha particle irradiation produces mainly isolated defects at the end of their ~15-μm range. Heavy recoil nuclei create dense collision cascades on a very short distance (up to ~20 nm). Fission fragments lead to the formation of tracks in the major part of their trajectory and dense collision cascades at their end of range.

Energetic ions lose their energy via two nearly independent mechanisms: (i) a direct transfer of kinetic energy to the target atoms by elastic collisions between the projectile nucleus and target nuclei, commonly denoted as nuclear energy loss; and (ii) the generation of excited and ionized atoms, referred to as electronic energy loss. Nuclear energy loss is the dominant process at low ion energy (below 10 keV amu^{-1}), while electronic energy loss contributes the most to the slowing down of fast ions (above 1 MeV amu^{-1}). Therefore, the various sources of radiation occurring in nuclear waste matrices involve both nuclear and electronic energy loss processes. The partitioning of the energy loss into nuclear and electronic stopping (i.e., the nature of the physical mechanisms) is decisive for the radiation damage production. Both processes are responsible for the formation a large variety of atomic rearrangements (e.g., topological and chemical disorder, phase transformations, removal of surface atoms, swelling, and amorphization), depending on the nature of the irradiated target and on irradiation conditions (energy, fluence, flux, and irradiation temperature).

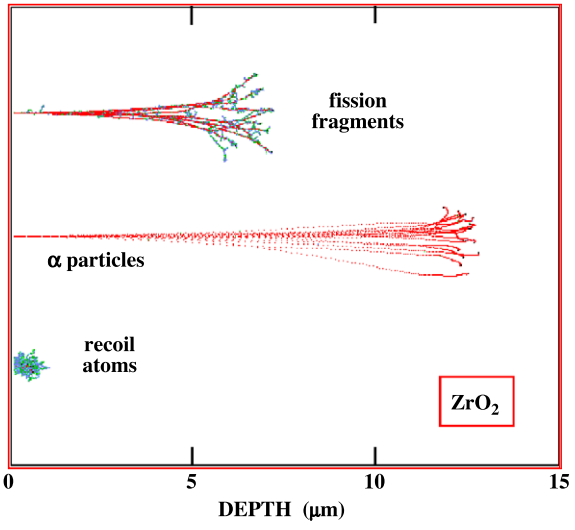

Fig. 1. Range and defect density for the main sources of radiation occurring in a nuclear waste matrix. Calculations are performed in zirconia with the SRIM code [5]. Note that the range of recoil atoms has been multiplied by a factor of 10 for the sake of legibility.

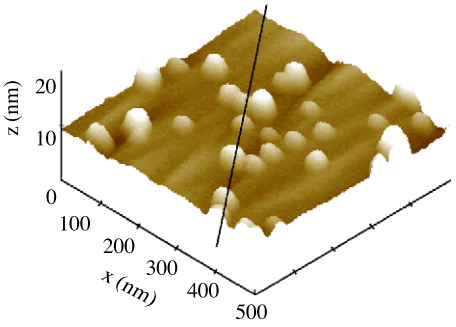

Fig. 2. Atomic force microscopy micrograph recorded on a UO_2 single crystal irradiated with 1-GeV Pb ions at a fluence of 10^{10} cm^{-2}.

The formation of radiation damage in nuclear ceramics by nuclear energy loss is studied from many years and the current state of knowledge is summarized in recent reviews [7,8]. Thus, the present paper is focused on the stability of waste matrices submitted to electronic energy loss for which the relaxation processes are not still fully understood.

The electronic energy deposition due to the passage of a swift ion induces the formation of an electrostatically unstable continuous cylinder of ionized atoms and the emission of electrons. Two main approaches, namely the thermal spike [9,10] and the Coulomb explosion [11] models, were developed to account for the observed atomic rearrangements. In the thermal spike model, excited electrons transfer their energy to the lattice through electron–atom interactions. The Coulomb explosion concept considers that the unstable cylinder of highly ionized matter induces radial impulses to atoms lying in the ion wake. Only a few studies were conducted on the stability of nuclear waste matrices subjected to swift heavy ion irradiation. The results presented in this section deal with the specific case of urania.

Damage creation in solids irradiated with swift heavy ion generally proceeds by the formation of tracks. At very low fluences, individual impacts of ions are observed at the surface of the target. As an example, Fig. 2 shows typical hillocks formed on the surface of a UO_2 single crystal bombarded with 1 GeV of Pb ions, recorded by atomic force microscopy experiments [12]. When the ion fluence is increased, individual ion impacts overlap and the microstructure of the material evolves depending on projectile and target parameters. A peculiar single crystal–polycrystal phase transformation (i.e., polygonization of the material) was recently evidenced in UO_2 single crystals irradiated with swift Xe ions by the combination of Rutherford backscattering spectroscopy (RBS) in channeling conditions and X-ray diffraction experiments [13,14]. The RBS spectra presented in Fig. 3 exhibit two main parts: (i) a high-energy