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## Carbon 14 Distribution in Irradiated BWR Fuel Cladding and Released Carbon 14 after Aqueous Immersion of 6.5 Years

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### Abstract

Spent fuel cladding which is highly activated and strongly contaminated is expected to be disposed of in an underground repository. A typical activation product in the activated metal waste is carbon 14 ( $^{14}\text{C}$ ), which is mainly generated by the  $^{14}\text{N}(\text{n,p})^{14}\text{C}$  reaction and produces a significant exposure dose due to the large inventory, long half-life (5730 years), rapid release rate, and the speciation and consequent migration parameters. In the preliminary Japanese safety case, the release of radionuclides from the metal matrix is regarded as the corrosion-related congruent release, and the cladding oxide layer is regarded as a source of instant release fraction (IRF).

In the present work, specific activity of  $^{14}\text{C}$  was measured using an irradiated BWR fuel cladding (Zircaloy-2, average rod burnup of 41.6 GWd/tU) which has an external oxide film having a thickness of 25.3  $\mu\text{m}$ . The  $^{14}\text{C}$  specific activity of the base metal was  $1.49 \times 10^4$  Bq/g, which in the corresponding burnup is comparable to values in the existing literature, which were obtained from various irradiated claddings. Although the specific activity in oxide was 2.8 times the base metal activity due to the additive generation by the  $^{17}\text{O}(\text{n},\alpha)^{14}\text{C}$  reaction, the  $^{14}\text{C}$  abundance in oxide was less than 10% of total inventory. A static leaching test using the cladding tube was carried out in an air-tight vessel filled with a deoxygenated dilute NaOH solution (pH of 12.5) at room temperature. After 6.5 years,  $^{14}\text{C}$  was found in each leachate fraction of gas phase and dissolved organics and inorganics, the total of which was less than 0.01% of the  $^{14}\text{C}$  inventory of the immersed cladding tube. A simple calculation based on the congruent release with Zircaloy corrosion has suggested that the 96.7% of released  $^{14}\text{C}$  was from the external oxide layer and 3.3% was from the base Zircaloy metal. However, both the  $^{14}\text{C}$  abundance and the low leaching rate suggests that  $^{14}\text{C}$  in oxide does not have a significant impact on the IRF in the safety case.

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## 1. Introduction

One challenging issue in the nuclear fuel cycle is to implement safe and rational management of waste generated from fuel reprocessing. After nuclear fuel is reprocessed, spent fuel cladding which is highly activated and strongly contaminated is expected to be disposed of in an underground repository. A typical activation product in activated metal waste is carbon-14 ( $^{14}\text{C}$ ) which is mainly generated by a  $^{14}\text{N}(\text{n,p})^{14}\text{C}$  reaction in the nuclear reactor. In a preliminary performance assessment in Japan,  $^{14}\text{C}$  produces a significant exposure dose due to the large inventory, long half-life (5730 years), rapid release rate, and the speciation and consequent migration parameters. The release of radionuclides from the metal matrix is regarded as the corrosion-related congruent release. Conversely, the oxide film formed on the Zircaloy cladding surface is regarded as a source of the instant release fraction (IRF). The Zircaloy corrosion rate of  $0.02\ \mu\text{m}/\text{y}$  and the 20% IRF of the spent cladding waste have been suggested as the source term parameters in the preliminary Japanese safety case.<sup>1</sup> The speciation of  $^{14}\text{C}$  has been assumed to be an aqueous organic in this safety case.<sup>1</sup>

The data for specific activity of  $^{14}\text{C}$  were summarized by Van Konynenburg<sup>2</sup> from measurements of commercial spent claddings in the U.S.<sup>3-9</sup> These data show activity estimates on the order of  $10^4\ \text{Bq}/\text{g-metal}$  which increased in correspondence with fuel burnup. The approximately same level of activity for  $^{14}\text{C}$  were reported for the claddings of Japanese pressure water reactors (PWRs) (burnup of  $47.9\ \text{GWd}/\text{tU}$ )<sup>10</sup> and boiling water reactors (BWRs) (STEP III type,  $39.7\ \text{GWd}/\text{tU}$ )<sup>11</sup> and for the claddings of German PWRs and BWRs<sup>12</sup>. Using specific activity measurements, Guenther et al.<sup>13</sup> and Yamaguchi et al.<sup>10</sup> studied the external oxide layer formed by in-pile corrosion of Zircaloy fuel cladding and estimated that the total  $^{14}\text{C}$  contained in the layer is about 15% (Guenther) or 17% (Yamaguchi). Sakuragi et al.<sup>14</sup> have employed the ORIGEN calculation using the chemical composition and oxide thickness of Japanese cladding to estimate the  $^{14}\text{C}$  inventory of the oxide layer as approximately 9% for PWRs and 0.8% for BWRs.

Radionuclide release from the spent cladding has been reported by several studies that investigate the release of volatile  $^{14}\text{C}$  in air at temperatures above  $373\ \text{K}$ <sup>15,16</sup> and aqueous leaching under caustic conditions (using several boiling, acid, and alkaline aqueous solutions).<sup>17</sup> For the deep repository condition, immersion tests in aqueous media of pH 12.5 at room temperature were performed using cladding from Japanese PWRs<sup>10</sup> and BWRs.<sup>11</sup> However, the 2-year immersion periods for both PWR and BWR tests were too short for an assessment of long-term evolution.

In the corrosion behavior of Zircaloy, the pioneering studies using electrochemical techniques suggest that zirconium alloy has an extremely low or negligible corrosion rate.<sup>18,19</sup> Shoesmith and Zagidulin reviewed the relevant literature and estimated that the corrosion rate of zirconium alloys under geological repository conditions would be less than  $1\ \text{nm}/\text{year}$ .<sup>20</sup> However, Kreis pointed out that an electrochemical study is not well suited due to the sensitivity limits of the apparatus.<sup>21</sup> A recent sensitivity analysis conducted by measuring the hydrogen evolution from a corrosion reaction has demonstrated that the corrosion rate of Zircaloy decreased to  $5\ \text{nm}/\text{y}$  after 1500 days.<sup>22</sup> In addition, the corrosion behavior of zirconium base alloys at high temperatures in in-pile conditions has been studied for many years. Owing to the results of these studies, it is widely accepted that the corrosion kinetics of Zircaloy follows a cubic rate law before the transition point (“breakaway”) is reached, and an Arrhenius type corrosion rate constant has been suggested.<sup>23</sup> Maeda et al. have discussed the low temperature corrosion kinetics through extrapolation on the high temperature Arrhenius plot.<sup>24</sup>

Despite the presence of these many studies, the basic information is still sparse on radionuclide inventory, release behavior of radionuclides, and corrosion. On the source term issues mentioned above, we have prepared an irradiated BWR fuel cladding (Zircaloy-2, average rod burnup of  $41.6\ \text{GWd}/\text{tU}$ ) which has an external oxide film with a thickness of  $25.3\ \mu\text{m}$ . The content of  $^{14}\text{C}$  was determined by a wet chemical treatment and liquid scintillation counting of  $^{14}\text{C}$  activity using different parts of the cladding such as the base Zircaloy metal, oxide, and metal with and without oxide film. In the next step, a static leaching test using cladding tube which internal oxide was removed was carried out under simulated geological disposal conditions for an exposure period of 6.5 years. This report then uses the results on Zircaloy corrosion rates to discuss the  $^{14}\text{C}$  leaching behavior and which part of cladding provides the major leaching source.

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