PRELIMINARY STUDIES ON THE DETERMINATION OF FISSION PRODUCT
INVENTORIES AT GRAIN BOUNDARIES IN USED CANDU FUEL

by

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ABSTRACT

Experimentally determined grain-boundary inventories for $^{137}$Cs and $^{99}$Tc are needed to verify a model that is currently used to calculate grain-boundary inventories for these fission products in used CANDU (CANada Deuterium Uranium) fuel. This report describes two methods that have been used to determine grain-boundary inventories for $^{137}$Cs, $^{99}$Tc, and also $^{90}$Sr. Both methods used air oxidation of the fuel in an attempt to make the grain-boundary inventories more accessible to leaching in aqueous solutions.

In the first method, oxidation was followed by leaching in dilute solutions of various compositions. The results obtained from these experiments were inconclusive because of over-oxidation of the fuel. This resulted in considerable exposure of the matrix, as well as the grain boundaries, to the leachants, and may also have caused preferential leaching as a result of severe matrix alteration.

In the second method, partially oxidized used fuel was stripped of its grain-boundary inventories by a brief exposure to strong acid. The resulting "bare" matrix was analyzed for fission-product inventories. Grain-boundary inventories for $^{137}$Cs, $^{90}$Sr and $^{99}$Tc were calculated by subtracting matrix and gap inventories from total fuel inventories. However, because of the propagation of 2a solution analyses errors, the results obtained from these experiments were also somewhat inconclusive. Scanning electron microscopy on pre- and post-acid-leached fuel suggested that the removal of grain boundaries by acid was either not complete or produced precipitates, giving rise to the suspicion that the matrix inventories determined after this leaching step were incorrect.

The results from measurements performed using the second method allowed the measured and calculated total inventories of $^{137}$Cs, $^{90}$Sr, and $^{99}$Tc to be compared for 13 fuel elements. The measured values for $^{137}$Cs and $^{90}$Sr were generally within 20% of the calculated values. For $^{99}$Tc, however, the comparison was poor, with most calculated and measured values differing by a factor of 2 to 5.