Carbon 14 distribution during spent fuel dissolution - Application to an UOx3 spent fuel

M. Crozet, G. Ferlay

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M. Crozet; G. Ferlay

CEA Nuclear Energy Division, Marcoule Center

Radiochemistry & Processes Department,

Laboratory of Dissolution Studies

F-30207 Bagnols sur Cèze Cedex, France

Carbon 14 (^{14}C) is a radionuclide formed during the irradiation of nuclear fuel, primarily from nitrogen 14, impurity present in the fuel, and from oxygen 17 present in oxide matrix. One of the objectives related to this radionuclide concerns the knowledge of its distribution in the different flows during the spent fuel dissolution, some of which contribute to the wastes of the plant. These flows are gases emitted during the dissolution, unclarified dissolution solution and hulls. In 2013-2014, an essay program offering quantification of ^{14}C in each of these flows during nitric dissolution was proposed and applied: around 51 grams of a 53.1 GWd/t_{HM} UOX3 spent fuel were involved in the essay. The block diagram of the conducted essay is shown in Figure 1.

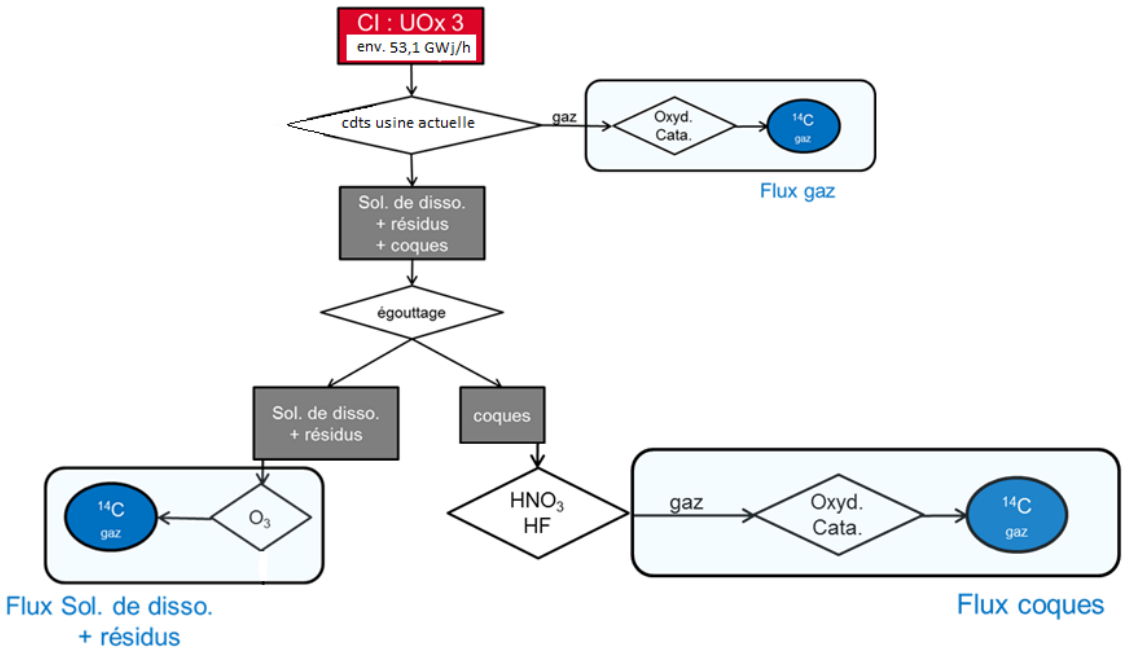

Figure 1 : Block diagram of the essay of ^{14}C measurement in the different flows generated during spent fuel dissolution

Measurement principle: Whatever the material of which one wants to determine the ^{14}C content, the principle of ^{14}C analysis method is currently almost always the same. This is to pass the ^{14}C in the gas form then converting it to carbon dioxide to trap as carbonate in a sodium hydroxide solution, which solution is then analyzed by β scintillation (Figure 2). Each step must be quantitative or with a known yield.

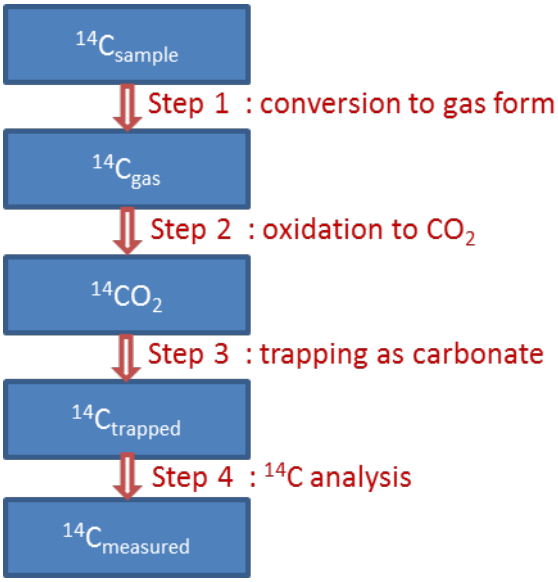

Figure 2 : Measurement principle of ^{14}C in a material

Gas flow: The ^{14}C activity measured in the gas flow emitted during this spent fuel dissolution (on conditions closed to the current plant ones and before trapping) is approximately 580,000 Bq that is to say 11,300 Bq /g of spent fuel (in this work, by spent fuel, it is understood the entire fuel section involved, oxide and clad), about 13,400 Bq / g of oxide.

Hulls flow: The amount of ^{14}C measured in hulls corresponds to an activity of 195 000 Bq for the overall essay and thus to 3,800 Bq / g of spent fuel.

Unclarified dissolution solution: The amount of ^{14}C measured in the unclarified spent fuel dissolution solution corresponds to an activity of 666,000 Bq for the essay, and thus to 12,900 Bq / g of spent fuel.

The total ^{14}C activity measured in the Cruas (53.1 GWd / t_{HM}) UOX3 fuel section dissolved in conditions closed to those of the current plant is 1,441,000 Bq corresponding to about 51 g of spent fuel dissolved that is 28,000 Bq / g of spent fuel (oxide and clad). Under these spent fuel dissolution conditions, 40% of ^{14}C present in the UOx3 dissolved section are measured in the emitted gas during the spent fuel dissolution, 46% are measured in the unclarified dissolution solution and 14% are measured in hulls. This essay is the first measurement of ^{14}C distribution in the different flows generated by spent fuel nitric dissolution..