Monte Carlo simulation of the electron and X-ray depth distribution for quantitative electron probe microanalysis of PWR spent fuels

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1. Introduction

Electron probe microanalysis requires several corrections to quantify an element of a specimen. The X-rays produced by the primary beam are created at some depth in the specimen. This distribution is usually represented as the function ϕ(ρz), and it is possible to calculate the correction factors for atomic number and absorption effects [1].

The electron and X-ray depth distributions for a quantitative electron probe micro analysis were simulated by the CASINO Monte Carlo program [2] to quantify some elements of the PWR spent fuel with 50 GWd/tU of burnup and 2 years of cooling time.

2. Methods and Results

In this section some of the theories and results of Monte Carlo simulation are described.

2.1 PWR Spent Fuel

Fresh nuclear fuel only has uranium and oxygen before the fission reaction. During the irradiation period of a nuclear fuel at a reactor, almost all the elements of the periodic table are produced as named in the fission product and trans-uranium. The Origen-S[3] code was used for time-dependent concentrations of spent fuels enriched to 4.5% U-235. Two years was selected for the decay time and its burnup was 50 GWd/tU. Table 1 shows major elements from the results of Origen-S code calculations.

Table I: Compositions of PWR spent fuels

Element	*wt %*
U	*83.64*
O	*10.61*
Pu	*1.04*
Xe	*0.72*
Nd	*0.54*
Zr	*0.48*
Mo	*0.45*
Cs	*0.37*
Enrichment	*4.5 wt% U-235*
Burnup	*50 GWd/tU*
Cooling time	*2 years*

2.2 Depth Distribution

The term ϕ(ρz) can be defined as the number of X-rays generated in a thin layer, dρz, of a specimen at a depth, ρz, from the surface, normalized by the intensity generated in an isolated layer of the same thickness. The depth distribution of the production of X-rays can be represented using the ϕ(ρz) versus ρz curves.

The ϕ(ρz) correction model combines atomic number and absorption effect and is usually expressed as a ratio 'k ratio' given by

$$k_a = C_a \frac{\int_0^\infty \Phi_a(\rho z)\exp(-\chi_a \rho z)\, d\rho z}{\int_0^\infty \Phi_s(\rho z)\exp(-\chi_s \rho z)\, d\rho z}$$

where χ is μ csc(ψ), which is defined as the fraction of X-rays that escape from the specimen at an angle, ψ. Subscript a is the sample and s is the standard. ρz is the mass depth.

2.3 Monte Carlo Simulation

Montel Carlo simulation was carried out for the PWR spent fuel, metallic U, UO_2, Nd and Al_2O_3 using 200,000 electrons of 25 keV.

Figures 1 and 2 show the maximum electron penetration depth and the energy of the backscattered electrons escaping the sample surface in the PWR spent fuel sample. The electron penetration depth in the sample was 1.2 μm, and the diameter of the interaction area was 1.8 μm as a result of the electron beam simulation.

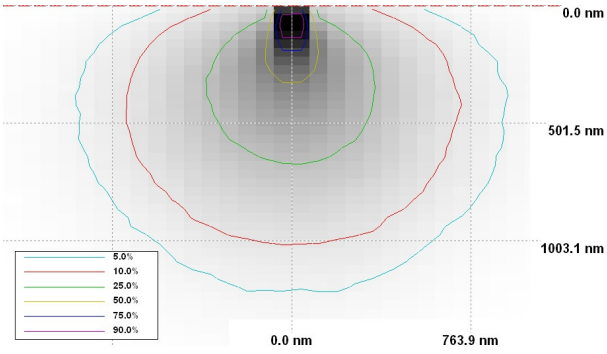

Fig. 1. Side view of absorbed energy in PWR spent fuel sample showing contour energy lines.

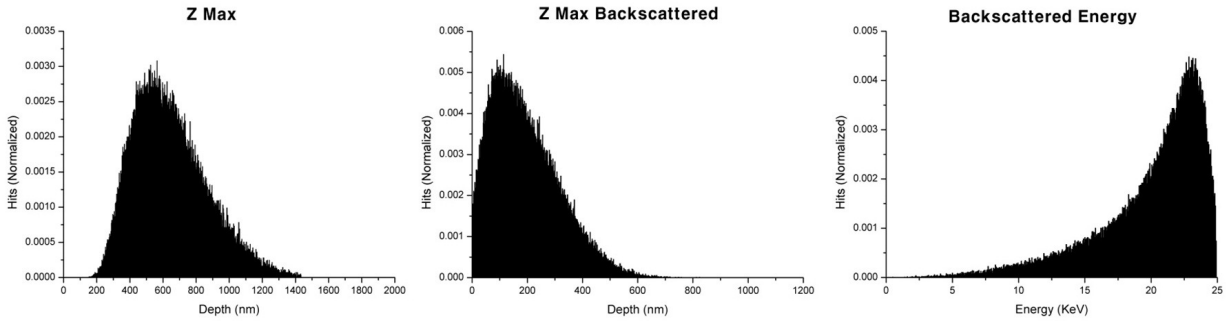

Fig. 2. Electron depth distribution: (a) maximum penetration depth in the spent fuel sample of the electron trajectories; (b) maximum penetration depth of electron trajectories that will escape the sample surface; (c) energy of Bes when escaping the surface of the sample.

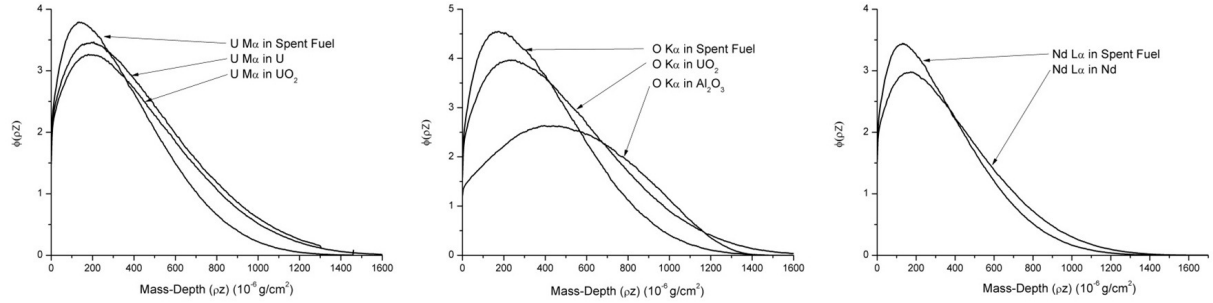

Fig. 3. X-ray depth distribution: (a) U Mα line in spent fuel, U, and UO_2; (b) O Kα line in spent fuel, UO_2 and Al_2O_3; (c) Nd Lα line in spent fuel and Nd

Table II: Generated X-ray intensity and correction factor

Sample	*X-ray line*	*$\phi(\rho z)$gen Area (g/cm^2)*	*$\phi(\rho z)$emit Area (g/cm^2)*	*$\phi(0)$*	*$\phi(max)$*	*k ratio*	*ZA*
Spent Fuel	*U Ma*	*2.05E-03*	*1.47E-03*	*1.57*	*3.79*		
U	*U Ma*	*2.36E-03*	*1.55E-03*	*1.48*	*9.48*	*0.798*	*9.54E-01*
UO2	*U Ma*	*2.19E-03*	*2.19E-03*	*1.50*	*3.27*	*0.563*	*7.62E-01*
Spent Fuel	*Nd La*	*1.77E-03*	*1.22E-03*	*1.52*	*3.42*		
Nd	*Nd La*	*1.79E-03*	*1.52E-03*	*1.52*	*2.98*	*0.0044*	*8.04E-01*
Spent Fuel	*O Ka*	*2.76E-03*	*2.24E-04*	*1.65*	*4.55*		
UO2	*O Ka*	*2.96E-03*	*1.92E-04*	*1.58*	*3.97*	*0.124*	*1.17E+00*
Al2O3	*O Ka*	*2.30E-03*	*2.84E-04*	*1.21*	*2.63*	*0.084*	*7.89E-01*

Figure 3 is the $\phi(\rho z)$ curves, which give information about the X-ray generation depth of each chemical element in the sample and data to calculate the correction factors

The different atomic number matrices cause a change in $\phi(0)$ and the height of the $\phi(\rho z)$ curves. Correction factor ZA was calculated by taking the ratio of the $\phi(\rho z)_{gen}$ area for the standard to the $\phi(\rho z)_{gen}$ area for an element as shown in table II.

3. Conclusions

Electron and X-ray depth distribution in PWR spent fuel with 50 GWd/tU of burnup were simulated by a CASINO Monte Carlo program. Finally, by using the $\phi(\rho z)$ curves obtained by the simulation, the correction factors of U Ma, Nd La, and O Ka in the spent fuel were determined.

REFERENCES

[1] C. Merlet, An Accurate Computer Correction Program for Quantitative Electron Probe Microanalysis, Mikrochimica Acta, Vol.114/115, p. 363, 1994.
[2] D.Drouin et al., CASINO V2.42 – A Fast and Easy-to-use Modeling Tool for Scanning Election Microscopy and Microanalysis Users, Scanning, Vol.29, p.92, 2007.
[3] I.C. Gauld et al., ORIGEN-S: Scale System Module Calculate Fuel Depletion, Actinide Transmutation, Fission Product Buildup and Decay, and Associated Radiation Source Terms, Vol II, Section F7, NUREG/CR-0200, 2004.