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Determination of the L- and M-Subshell X-Ray Production Cross Sections for Pb and U Using an Electron Microprobe.

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In the nuclear industry, electron probe microanalysis (EPMA) is used to quantitatively analyze the amount of actinides present in fresh and spent fuels and, therefore, reliable EPMA methods are needed to improve the manufacturing of mixed-oxide and next-generation fuels, the recycling of irradiated fuel, the waste disposal, and the analysis of severe accidents. However, quantitative EPMA is not always feasible for these materials because of the lack of suitable reference standards for the radionuclides. To overcome this difficulty, standardless methods of analysis that use ``virtual'' standards are employed. These virtual standards are generally obtained from empirical formulae based on experimental extrapolations or from theoretical calculations that require physical parameters which are poorly known. One such quantity is the cross section for x-ray production. Different theoretical models and predictive formulae are available to calculate x-ray production cross sections but the reliability of such calculations needs to be confirmed experimentally. Knowledge of x-ray production cross sections is also required in other fields such as radiation transport simulations, astrophysics, radiation shielding design, and fusion plasma physics.

In the present work, L- and M-shell x-ray production cross sections were measured for elements Pb (Z= 82) and U (Z=92) by electron impact using the experimental procedure described in Refs. [1, 2]. X-ray intensities were recorded for primary electron beams with energies ranging from the ionization threshold up to 38 keV, in steps of 0.5 keV (below 15 keV), 1 keV (from 15 keV up to 30 keV) and 2 keV (above 30 keV). The x-ray intensities were recorded with several wavelength-dispersive spectrometers on two different CAMECA SX-100 electron microprobes. For each electron incident energy, measurements were performed at least ten positions on different samples. In order to obtain a satisfactory signal-to-noise ratio, especially near the ionization threshold, ultrathin films of the considered elements deposited on self-supporting carbon backing films were used as targets, which were obtained by vacuum evaporation. The thickness of the active element films was in the range 0.2–8 nm, while the backing carbon films had a thickness in the range 5–25 nm.

Relative x-ray measurements, which give the shape of the cross-section curve as a function of beam energy, were performed on two sets of samples which had different thicknesses. The thinnest samples were used for the measurements at low energies in the attempt to minimize the effects of multiple scattering and energy loss within the active film, and of backscattering from the backing films. Random uncertainties were estimated to be of ~ 4% for the L lines and ~ 2% for the M lines. These uncertainties are mainly due to counting statistics, energy spread of the incident electron beam and instrumental instabilities. Relative x-ray production cross sections provide useful information for EPMA analysis at low overvoltage, for the characterization of highly-inhomogeneous samples and to confirm the reliability of predictive empirical cross-section formulae.

The conversion from the recorded signal into absolute x-ray production cross sections requires

knowledge of parameters such as the number of incident electrons, target thickness, solid angle of collection, and detector efficiency, which were determined by using different methodologies [1, 2]. For instance, the product of the solid angle of collection by the detector efficiency was determined by comparing measured and simulated bremsstrahlung intensities from samples of known composition (e.g. C for the M lines and Ni for the L lines). The simulations of bremsstrahlung were performed with the help of the Monte Carlo code PENELOPE [3]. Systematic uncertainties were estimated to be of ~ 5%, the main contributions being that of the detection efficiency and solid angle of collection. These systematic uncertainties do not alter the cross-section curve shape but cause a global shift of the curve. The global uncertainties were obtained by combining in quadrature random and systematic uncertainties and amounted up to ~ 8% for the L lines and ~ 7% for the M lines.

The measured cross sections have been compared, wherever possible, with experimental data available in the literature [4], with the predictions of analytical formulae widely used in practical applications such as the well-known formulae of Casnati or Gryzinsky, and with the more recent parameterization of Bote et al. [5]. These authors have given a useful parameterization of ionization cross sections obtained from an extensive database of cross sections, which had been calculated essentially with the distorted-wave Born approximation (DWBA). For the conversion of inner-shell ionization cross sections into x-ray production cross sections, we have extracted the necessary atomic relaxation parameters, namely the fluorescence and Coster-Kronig yields, vacancy transfer probabilities and x-ray emission rates from the Evaluated Atomic Data Library (EADL). Our measured cross sections agree satisfactorily, within the uncertainties of the measurements and of the relaxation parameters, with the predictions of the DWBA calculations of Bote et al. [5], and thus confirm the usefulness of Bote et al.'s predictive formula for the calculation of virtual standards in the EPMA analysis of actinide elements.

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