Monte Carlo Simulation of Characteristic Secondary Fluorescence in Electron Probe Microanalysis of Homogeneous Samples Using the Splitting Technique

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Abstract: Electron probe microanalysis (EPMA) is based on the comparison of characteristic intensities induced by monoenergetic electrons. When the electron beam ionizes inner atomic shells and these ionizations cause the emission of characteristic X-rays, secondary fluorescence can occur, originating from ionizations induced by X-ray photons produced by the primary electron interactions. As detectors are unable to distinguish the origin of these characteristic X-rays, Monte Carlo simulation of radiation transport becomes a determinant tool in the study of this fluorescence enhancement. In this work, characteristic secondary fluorescence enhancement in EPMA has been studied by using the splitting routines offered by PENELOPE 2008 as a variance reduction alternative. This approach is controlled by a single parameter NSPLIT, which represents the desired number of X-ray photon replicas. The dependence of the uncertainties associated with secondary intensities on NSPLIT was studied as a function of the accelerating voltage and the sample composition in a simple binary alloy in which this effect becomes relevant. The achieved efficiencies for the simulated secondary intensities bear a remarkable improvement when increasing the NSPLIT parameter; although in most cases an NSPLIT value of 100 is sufficient, some less likely enhancements may require stronger splitting in order to increase the efficiency associated with the simulation of secondary intensities.

Key words: EPMA, characteristic fluorescence enhancement, Monte Carlo simulation, variance reduction

INTRODUCTION

Electron probe microanalysis (EPMA) is a powerful analytical tool, which allows chemical quantification of the elements present in samples of different characteristics, along a wide range of specimen compositions. When a finely collimated electron beam impinges on a flat material, the characteristic intensity emitted by each element composing the sample can be used not only to identify these elements, but they may be recorded and then compared with the corresponding intensities emitted from standards of known composition. This comparison originates in the assumption that the emitted characteristic intensities proportionally relate to the mass concentrations of the corresponding elements, which allows to eliminate geometrical and physical factors that are very difficult to determine (Reed, 1993; Scott et al., 1995). With adequate procedures for data reduction, the different matrix effects are taken into account, usually referred to as "*ZAF* correction" (Goldstein et al., 2003); also called "matrix corrections," they were originally separated into factors accounting for differences in the generation of X-rays and scattering of the incident beam (*Z*, for atomic number

correction), absorption effects (*A*), and secondary fluorescence enhancement (*F*). These matrix correction factors strongly depend on the experimental conditions, mainly on the incident beam energy, X-ray take-off angle, and differences in composition of the standards used to compare with the unknown samples.

When the electron beam ionizes inner atomic shells and these ionizations cause the emission of characteristic X-rays, secondary fluorescence can occur, originating from ionizations induced by X-ray photons produced by the primary electron interactions. This fluorescence enhancement effect occurs when an atom species present in the target has an inner-shell ionization energy lower than the energy of other characteristic X-rays or bremsstrahlung photons that originate within the sample. In such a case, the measured X-ray intensity from the fluoresced element will include both the direct electron-excited intensity as well as the additional intensity generated by such enhancement. Despite there exist many approaches available for the assessment of fluorescence enhancement (Reed, 1965; Ugarte et al., 1987), it is impossible to compare these predictions with experimental data as the photons produced by fluoresced atoms cannot be discriminated from the total recorded radiation. Therefore, Monte Carlo simulations constitute a very important tool for estimating the fluorescence enhancement and a number of