Accurate automated non-resonant NRA depth profiling: Application to the low ^{3}He concentration detection in UO_2 and SiC

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Abstract

An automated method was developed to extract elemental depth profiles from non-resonant nuclear reaction analyses (NRA), which involves a two-stage procedure. The first stage enables the determination of the number of layers to be used in the final depth profile determination along with the thicknesses of each of the layers. To this end, the RESNRA program, which relies on the SIMNRA 5.0 simulation software to calculate a multilayer target, was designed at CERI. A definition of the depth resolution based on statistical considerations is proposed. In the second stage of the fitting process, a depth profile and corresponding error bars are extracted from the experimental spectrum by running a generalized reduced gradient (GRG2) algorithm using the previously calculated multilayer target. The one-to-one correspondence between the experimental spectrum and the depth profile demonstrates the objectivity of the method. The method is then applied to determining low concentration ^{3}He depth profiles in implanted UO_2 and SiC samples using the ^{3}He(^{2}H, ^{4}He)^{1}H non-resonant nuclear reaction. The results clearly demonstrate the relevance and potential of the method.
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1. Introduction

Nuclear reaction analysis (NRA) is a well known technique for depth profiling light elements in materials. As nuclear reactions are isotope specific, NRA is a powerful method in a wide range of fields requiring isotopic tracer measurements. In non-resonant NRA, the incident particles have a prescribed energy which they lose as they penetrate the material. The incoming particles are then liable to undergo a nuclear reaction with target nuclei. If reaction products have a high enough kinetic energy to recoil out of the target, they are detected at different energies depending on where they appeared in the sample. Non-resonant NRA

involves analysing the energy distribution of those reaction products, from which a depth profile is deduced.

Generally, the analysed target is modelled as a multilayer material where the layer thickness and the relative elemental concentrations are manually adjusted, so that the spectrum calculated using the SIMNRA 5.0 software [1] fits the experimental data available. Therefore, depth profiles extracted by different users may vary, despite all leading to acceptable simulations. This can cause some imprecision in the depth profile determination which in turn can induce a misinterpretation or over-interpretation of the experimental data.

The aim of this work is to develop an automated method to determine low concentration depth profiles in a one-to-one correspondence with experimental non-resonant NRA spectra. The analysis method relies on a two-stage process, the first of which involves determining the thickness of each

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of the material layers to be used in the final profile determination (second stage). To this end, the target is modelled as a multilayer material in which the analysed element concentration is constant and assumed to be low (as a consequence of this latter assumption, the stopping powers of incoming or outgoing particles are not altered). The thickness of each consecutive material layer is then chosen to correspond to the depth resolution at a given distance from the sample surface, which is derived from statistical criteria applied to RESNRA calculation results. This newly developed program relies on SIMNRA simulations to generate energy spectra corresponding to homogeneous distributions of ^{3}He atoms in the material.

In the second stage, the concentration of the probed isotope in each of the layers, the thickness of which is determined in the first stage of the analysis process, is evaluated using a generalized reduced gradient (GRG2) algorithm [2]. No constraints are assumed to fit the experimental energy spectrum. An automated method was previously proposed [3] to extract depth profiles from NRA spectra. Some aspects of that method differ from the present work: in this paper, the depth resolution is an essential part of the fitting process.

Our calculation procedure was used to extract ^{3}He depth profiles from α-spectra obtained from the ^{3}He(^{2}H, ^{4}He)^{1}H non-resonant nuclear reaction in UO$_2$ and SiC implanted samples. This method provides a single solution to the problem of the determination of the concentration profile and in addition provides error bars associated with the elemental concentrations determined in each of the modelled layers. This is obviously essential when analysing the experimental data and comparing two depth profiles.

2. Depth resolution and layer thickness determination

2.1. Statistical definition of the depth resolution

The depth resolution in backscattering analysis – e.g. NRA – is usually defined as the minimum detectable depth difference δx related to the minimum detectable scattered particle energy variation δE [4]. The energy difference between detected particles originating at different depths within the material is the result of the energy loss of incident and outgoing particles. Sources of the broadening of the measured energy distribution are detector resolution, initial energy spread of the incident beam, straggling of the outgoing and incoming particles, geometrical energy spread and energy spread coming from the multiple scattering [5]. Since energy straggling increases as the ion beam penetrates the sample, the depth resolution deteriorates at increasing distances from the sample surface. To calculate the depth resolution at any depth, the minimum detectable energy variation has to be defined. In non-resonant backscattering analysis, δE can be correlated with the help of a useful criterion based on statistical considerations.

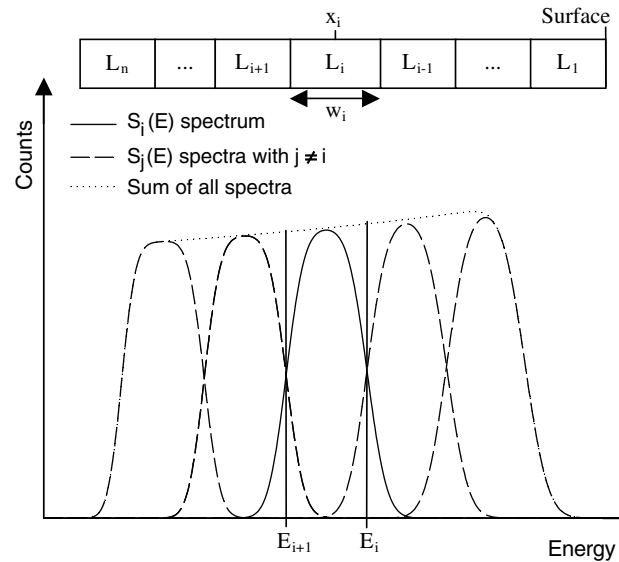For a given set of experimental conditions, we consider the NRA analysis of a thick material containing a homogeneous concentration of an element whose concentration one wants to determine. With the SIMNRA software, this material is modelled as a multilayer target and the spectral response of this analysis is the sum of n spectra generated from each of the modelled layers. A layer L_i of thickness w_i centred at depth of x_i corresponds to a detected energy distribution $S_i(E)$. Fig. 1 shows the contribution of each of the layers to the total energy spectrum of particles emitted from nuclear reactions in the material. To be clear in the understanding of Fig. 1, the cross section of the nuclear reaction is assumed to be constant in the laboratory frame and the thicknesses of the n layers identical. The $S_i(E)$ spectrum is intersected by other spectra at energies E_{i+1} and E_i.

The probability p_i that the particles detected in the $[E_{i+1}, E_i]$ energy range are emitted from the layer L_i is the counts in $S_i(E)$ divided by total number of counts in this energy range

$$p_i = \frac{\int_{E_{i+1}}^{E_i} S_i(E)\,\mathrm{d}E}{\int_{E_{i+1}}^{E_i} \left[\sum_j S_j(E)\right]\mathrm{d}E}. \qquad (1)$$

We now assume the only particles to contribute to the spectrum in the $[E_{i+1}, E_i]$ energy range are those produced in layers adjacent to layer L_i. Therefore, a first order estimate p_i can be calculated from Eq. (1) by considering three successive material layers L_{i-1}, L_i and L_{i+1} only, and their resulting consecutive spectra:

$$p_i \approx \frac{\int_{E_{i+1}}^{E_i} S_i(E)\,\mathrm{d}E}{\int_{E_{i+1}}^{E_i} [S_{i-1}(E) + S_i(E) + S_{i+1}(E)]\,\mathrm{d}E}, \qquad (2)$$

p_i takes into account the variations of the cross section in the vicinity of x_i, along the primary ion trajectory. Probability p_i is directly related to the $[E_{i+1}, E_i]$ energy range and

Fig. 1. Representation of spectra from the analysis using a non-resonant reaction of a multilayer material.