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DEPTH PROFILING OF OXIDISED a-C:D LAYERS ON BE - A COMPARISON OF [4]He RBS AND [28]Si ERD ANALYSIS

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Abstract:

Depth profiling by means of RBS is often difficult due to the overlap of the backscattering intensities of different constituents from different depths. The erosion and reaction of deposited amorphous deuterated carbon (a-C:D) films with a Be substrate due to annealing in air poses an analyical challenge especially if simultaneously the exchange of hydrogen isotopes should be monitored. The analysis of the different recoiling atoms from collisions with heavy ions in Elastic Recoil Detection (ERD) can provide a tool which resolves all constituents in a single analysis.

In the present study the composition of intermixed layers on Be containing H, D, Be, C and O has been analysed using conventional [4]He RBS at 2.2 MeV together with 2.5 MeV [4]He ERD for hydrogen isotope analysis. At these energies, an overlap of signals from different constituents could be avoided in most cases, but the hight $Be(\alpha,n)C$ cross section sets stringent limits for analysing fluxes and fluences. Also, backscattering and recoil cross sections are often not Rutherford and not available for all energies and scattering angles.

As alternative method heavy ion ERD using Si^{7+} ions extracted from a 5 MeV Tandem Van de Graff accelerator was investigated. At a scattering angle of 30 deg Si ions could not be scattered into the detector and a solid state detector without protecting foil could be used. Even in the intermixed layers at terminal energies of 5 MeV the heavy constituents could be separated while signals from recoiling hydrogen and deuterium atoms could be resolved on top of the signal from the Be substrate.

For the analysis of the RBS and ERD data the newly developed spectra simulation program SIMNRA has been used which includes a large data bank for scattering and nuclear reaction cross sections. For heavy ion ERD all interactions are described by Rutherford cross sections. The depth profiles of all constituents extracted from the simulation. are compared for both methods. In the case of the [4]He analysis problems in the evaluation arise mainly from the combination of the two different analysing runs for the heavy constituents and the hydrogen isotopes. In the case of heavy ion ERD the simulation of the substrate signal at large depths suffers from the insufficient description of plural and multiple scattering processes.

DISCLAIMER

Introduction:

In applications dealing with the deposition of amorphous hydrogenated carbon (a-C:H) layers [1] or in the determination of the composition of deposited layers on the walls of nuclear fusion plasma experiments [2] the analysis of mixtures of light elements on heavy substrates is necessary. In the case of wall analysis in thermonuclear plasma experiments the typical deposited elements are Be, B, C, O, Si and Ti, often on Be or C substrates. In these layers the amount of incorporated hydrogen isotopes is often of crucial importance [3]. This poses an eminent task to both, ion beam analysis techniques and data evaluation programs.

Using conventional 2.5 MeV Van-de-Graff accelerators this task cannot be accomplished in a single analysis. In most cases several analyses on the same spot have to be performed, such as ^{4}He Rutherford Backscattering (RBS) for depth profiling of heavier constituents and nuclear reaction techniques such as ^{9}Be(p,α)^{6}Li, ^{11}B(p,α_o)^{8}Be or D(^{3}He,α)H or ERD using 2.6 MeV ^{4}He for the profiling of hydrogen isotopes and light constituents. ^{4}He RBS often leads to overlapping signals of different elements from different depths and the composition of different analysis techniques into one depth profile is rather difficult [3].

Heavy ion ERD on the other hand allows in principle the detection of a variety of elements in one single analysis and the separation of signals overlapping in energy by the mass analysis of the detected particles. The questions to be answered in the present investigation were: Can heavy ion ERD analysis reasonably be achieved already with small tandem accelerators with a terminal voltage of the order of 2 or 3 MeV, and what are the depth resolutions and the maximum analysable layer thicknesses at these energies.

Experimental:

As samples, different thermally treated a-C:D films on Be were used [4]. Throughout the paper examples are given for a system of 100 nm a-C:D on 200 nm Be$_2$C on a Be substrate. This

sample allowed the analysis using ^{4}He RBS without overlapping signals from Be, C and O constituents.

The sample was investigated after thermal treatment in-situ in the UHV chamber using 2.2 MeV ^{4}He RBS and 2.5 MeV ^{4}He ERD as described in detail in ref [4]. The detection angles were 150° and 30°, respectively. For the ERD analysis a 5 μm stainless steel foil was used to prevent scattered ^{4}He ions to enter into the detector. A movable solid state detector with a typical energy resolution of 25 keV for ^{4}He ions was used for both detection geometries.

For heavy ion ERD the samples were transferred through air to the analysis chamber at the 5 MV Tandem accelerator [5]. The modest vacuum conditions of 10^{-6} mbar were due to frequent venting because the 12 μm Mylar foil in front of the detector, preventing the detection of scattered analysing ions, was installed or taken out leading to some oxygen uptake during analysis. As analysing ions Si at energies from 12 MeV (2 MV Si^{5+}) to 40 MeV (5 MV Si^{7+}) was used. The scattering angle was 30° with typically 75° incidence angle and 75 ° detection angle. At this geometry the incident Si ions cannot be scattered into the detector even from the heaviest target element, O, and the use of a protection foil was not necessary. For the highest energies a clear separation of the Be, C and O signals could be achieved without protection foil. Subsequently also for lower energies, where the signals of different elements overlap in energy, no mass analysis was performed. The detector was a conventional surface barrier detector with an energy resolution of the order of 400 keV for the ejected heavy particles. The spectra were interpreted using the newly developed computer program SIMNRA.

Analysing computer program SIMNRA:

For data analysis and spectrum simulation the program SIMNRA was used [6]. SIMNRA is a Windows 95 program with full graphical user interface for the simulation of RBS, ERDA and nuclear reaction analysis spectra. The program was mainly developed for the simulation of non-Rutherford scattering, ERDA (including non-Rutherford ERDA) and nuclear

reaction analysis. It uses a data base of about 300 non-Rutherford scattering and nuclear reactions cross sections. It allows the calculation of any ion-target combination and the use of any foil in front of the detector. In the case of Rutherford scattering cross sections the angular dependent screening of the nucleus is taken into account [7]. The target may consist of up to 100 layers. The simulation of a spectrum lasts only several seconds on a 166 MHz Pentium processor even for complicated target structures. The program is available as Shareware at SigmaBase (http://ibaserver.physics.isu.edu/sigmabase/programs/programs.html).

SIMNRA uses the Andersen-Ziegler tables for the electronic stopping of hydrogen isotopes and helium in all elements [8,9]. The electronic stopping power of heavy ions is obtained from the electronic stopping of protons using the formalism of Ziegler et. al. [10]. In compounds Bragg's rule is applied. Nuclear stopping is calculated using the universal potential from [10]. For the evaluation of energy loss the same formalism as in RUMP is used [11]. The derivatives of the stopping power are obtained by numerical differentiation of the stopping power curves.

For the calculation of nuclear energy loss straggling Bohr's theory is used [12]. Electronic energy loss straggling is calculated by applying the Chu correction to Bohr electronic energy loss straggling [12]. The effect of nonstochastic broadening or squeezing of an energy distributed beam due to the differences in stopping power (Symon's [13], Payne's and Tschalär's theory [14-16]) is also taken into account. The convolution of the energy spectrum with the depth dependent energy loss straggling and the finite energy resolution of the detector is performed by using Gauss-Mehler integration.

Results and Discussion:

1. Analysis at the 2.5 MeV Van-de-Graff acellerator

The layer system chosen was specially prepared to allow the separation of the different elements within the film without overlapping signals using 2.2 MeV [4]He RBS. Hydrogen isotope anaysis

was performed using 2.5 MeV ^{4}He ERD. For both analyes Be(α,n) processes resulted in measurable neutron production posing limits to the analysing fluxes and ion energies. Fig 1 shows the data from RBS and ERD and the corresponding simulation of the spectra using SIMNRA.

Fig. 1a and b

For the simulation of the spectra a system of 100 nm a-C:D with a composition of $C_{0.65}D_{0.35}$ on 200 nm $Be_{0.6}C_{0.4}$ was used. Within the depth resolution of the method (about 60 nm, mainly due to the energy resolution of the detector), no diffusional profile gradients at the interfaces were necessary to fit the measured spectrum. The cross sections for backscattering from C and O are still Rutherford at this energy [17,18]. For Be the backscattering cross section is clearly not Rutherford and data from [19] were used for a scattering angle of 157°. A correction factor of 1.4 as recommended by Liu et al. [20] in the energy range of 2 to 2.2 MeV would, acctually, lead to a strongly improved agreement of experiment and simulation. No data for the actual scattering angle of 150° are available and the fit to the spectra of the Be substrate is poor. For the ERD analysis of D the cross section data from Besenbacher et al. [21] are used. The recoil cross section is clearly non-Rutherford. The good agreement of both spectra simulations using the same target compositon requires good knowledge of the detection geometries, the charge collection and the solid angle of the detector. The use of the same movable detector avoided errors due to different energy calibrations.

2. Analysis at the 5 MV Tandem accelerator

Starting at an energy of 12 MeV Si ions the energy was raised to 24, 32, and 40 MeV and comparisons were made with and without protection foil. At the lowest energies, the use of a 12 μm Mylar foil resulted in the separation of the hydrogen isotopes from the other recoils as all ejected particles except hydrogen were stopped in the foil. At 40 MeV the signals from O, C and Be could be separated without overlap in energy. At this energy, however, deviations of the simulated spectrum from the experimental data are most probably due to deviations from the

Rutherford cross sections used in the evaluation [22]. As an example fig 2 shows the data at 24 MeV (3MV Si^{7+}) with and without the foil including the simulations using the SIMNRA code.

Fig 2a and b

In fig. 2a the spectrum taken without protecting foil shows the a-C:D layer not overlapping with the Be substrate. The Be_2C phases for carbon and beryllium overlap forming an additional peak in the spectrum around 11.5 MeV. Without prior knowledge of the target composition a deconvolution of the spectrum is not possible. If the different elemental contributions were separated experimentally, e.g. by taking time of flight information in addition to the total energy [22], the Be and C spectra can be registered with a depth resolution of about 25 nm. In the present case the depth resolution results in almost equal parts from the stopping of the incident 24 MeV Si ions (304 $eV/10^{15}$ at/cm^2) and the ejected 15 MeV carbon atoms (317 $eV/10^{15}$ at/cm^2). The steep increase of the carbon surface edge is given by the energy resolution of the detector of about 400 keV for the ejected 16 MeV carbon atoms. The gradual rise of the Be contribution both at the interface to the a-C:D layer and to the Be substrate shows diffusional transition layers of 50 and 70 nm, respectively, which are not resolvable using ^{4}He RBS. The signal from the ejected D atoms on top of the Be substrate signal has a much poorer depth resolution as the ejected 4.5 MeV D atoms have a very small stopping power of the order of 1 $eV/10^{15}$ at/cm^2.

The addition of a stopper foil in front of the detector shifts the signals of the heavier elements Be and C to lower energies while the ejected hydrogen isotopes are only little influenced by the foil. As carbon loses more energy in the foil, the contribution with the highest energy now is the Be. Actually, the carbon atoms are so strongly stopped in the foil that the full depth of the 300 nm layer cannot be analysed completely. Heavier surface constituents, such as O, cannot be detected and are stopped in the foil. Due to the smaller background of the Be substrate now also a superficial layer of about 5×10^{16} adsorbed H atoms can be detected. Both spectra could be simulated using the same target concentrations, no loss of D was found due to the subsequent analysis. Small deviations from a perfect fit of the leading Be and D edges may be due to the fact

that the energy calibration of the detector is slightly different for different elemental species while in the evaluation of the spectra an average energy calibration was used.

In order to compare the depth information as function of incident ion energy the carbon signal is shown in fig 3 on the same energy scale for the four different energies used for analysis without stopper foil.

Fig. 3a-d

For the highest energy of 40 MeV the carbon recoils are nearly completely separated from the Be recoils. For decreasing energies the signal overlap increases. From the steepness of the edges it appears that the depth resolution degrades with increasing ion energy. This is, however, not supported by the evaluation of the stopping powers, both of the incident Si ions and the ejected C atoms, which vary by less than 5% in this energy range. Also, the leading carbon edge shows that the detector resolution is only a weak function of energy increasing from 400 keV for 7.5 MeV to 500 keV at 25 MeV carbon atoms. The degrading steepness of the edges of both, the carbon and the beryllium contributions is due to irradiation induced changes in the layer structure. Similarly, an increasing loss of deuterium in the a-C:D layer is clearly observed at the high analysing energies accompanied by an uptake of oxygen from the residual gas in the analysing chamber. The ion induced changes in the sample structure is demonstrated in fig. 4.

Fig. 4a and b

In fig 4 the deuterium loss and uptake of oxygen as well as the width of the interfacial transition layers are plotted versus the analysing ion fluence. With increasing analyzing fluence the total amount of D decreases and the thickness of the interface layers increases. However, the fluence alone is not the determining factor of the ion induced changes. While in the phases of 12 and 24 MeV analysis almost no changes in the target structure were observed, drastic changes occur at energies above 32 MeV. The loss of deuterium amounts to about 2×10^4 per incident ion at 32 MeV and increases to about 1.5×10^5 at 40 MeV. Large radiation induced losses of hydrogen from

a-C:H layers of the order of 1×10^6 were reported earlier for 200 MeV Iodine bombardment [23]. These losses are usually attributed to the energy deposition into the surface layer by electronic energy loss. However, the sharp onset of ion induced desorption at 32 MeV indicates that the desorption mechanism is not a simple linear function of electronic stopping. The deuterium depleted layer takes up oxygen from the residual gas or during venting similarly as reported for the same layers during thermal desorption of deuterium in air or oxygen atmosphere [1,4]. Also, a steep increase in interfacial transition width occurs only at energies above 32 MeV.

Conclusion:

Due to the different masses of the ejected particles the ERD technique can, in principle, separate the signals from different elements in the analysed surface layer. However, this has not been done in the present experiments, resulting in overlapping spectra. The newly developed program SIMNRA is a powerfull tool for data analysis, both for RBS and ERD. The strength of the ERD technique is demonstrated by the evaluation of the contributions of different elements to the spectra.

ERD provides strongly improved depth resolution compared to He-RBS amounting in standard geometry to 25 nm and 60 nm, respectively, for carbon layers. Films of the order of 300 nm can be analysed at energies of 12 MeV (2 MV Si^{5+}), if a stopper foil can be avoided, and at 24 MeV (3 MV Si^{7+}) with a 12 Om Mylar foil.

Si ion ERD has a poor depth resolution for hydrogen isotopes due to the small stopping power of the incident ions and negligable values for the high energetic ejected hydrogen. Good resolution can only be obtained for heavy ions, special target geometries and high resolution detectors [24]. A better depth resolution was achieved for ^{4}He ERD at 2.6 MeV.

Heavy ion ERD, especially at energies above 32 MeV, leads to drastic changes in the sample structure and composition. Large release rate for gaseous constituents ($> 10^4$ atoms/ion) and ion induced diffusional broadening of layer structures were observed.

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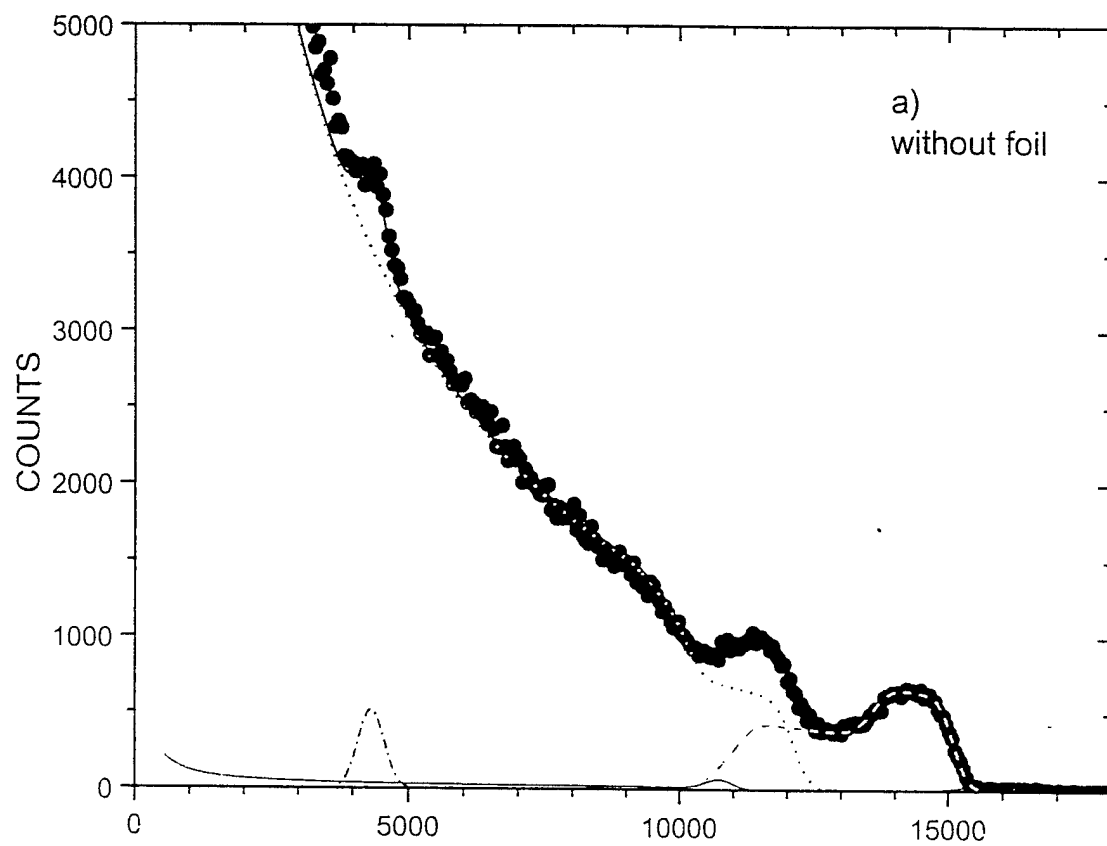
Figure captions:

Fig. 1 Analysis of the a-C:D/Be$_2$C/Be sample before Si ERD. a) RBS spectrum at 2.2 MeV ^{4}He. b) ERD spectrum using 2.6 MeV ^{4}He.
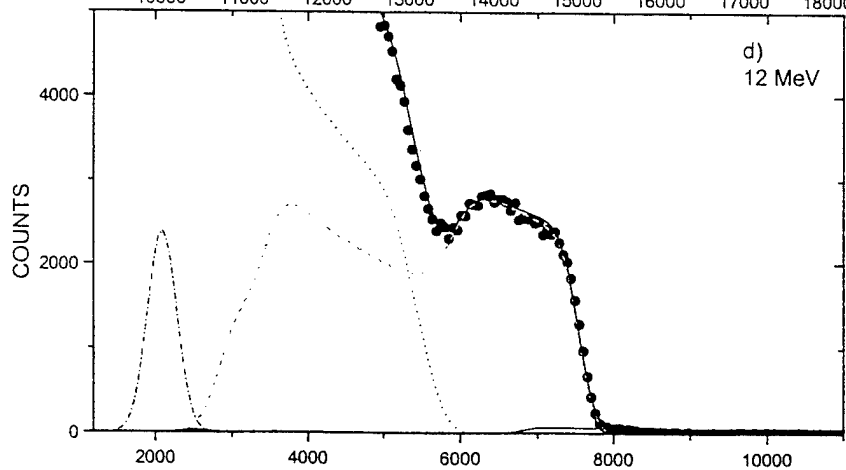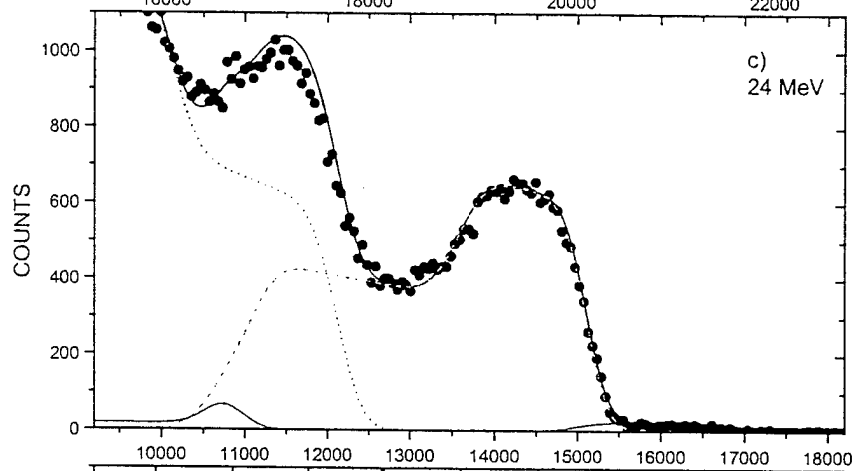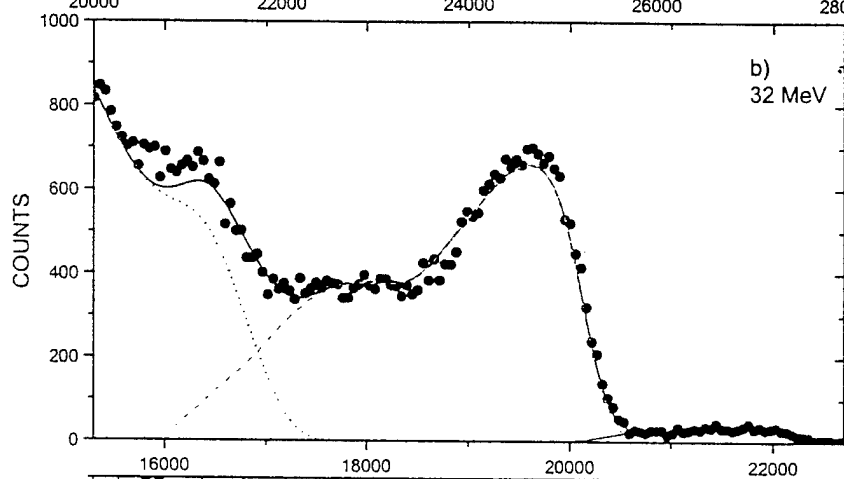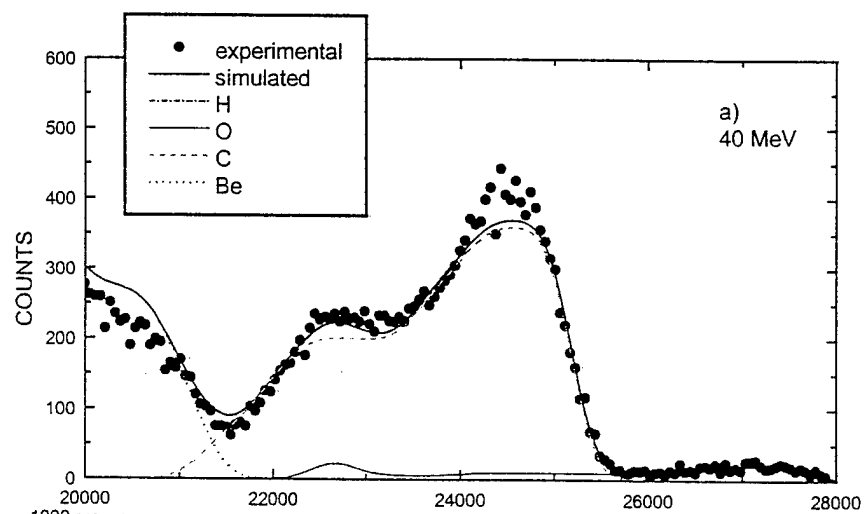
Fig. 2 Comparison of experimental dat and simulation for Si ERD of the a-C:D/Be$_2$C/Be sample. a) 24 MeV Si$^+$ ERD without stopping foil. b) 24 MeV Si$^+$ with a 12 μm Mylar foil infront of the detector.

Fig.3 Comparison of the carbon ERD signal for different Si ion energies as measured experimentally and simulyted using the SIMNRA code.
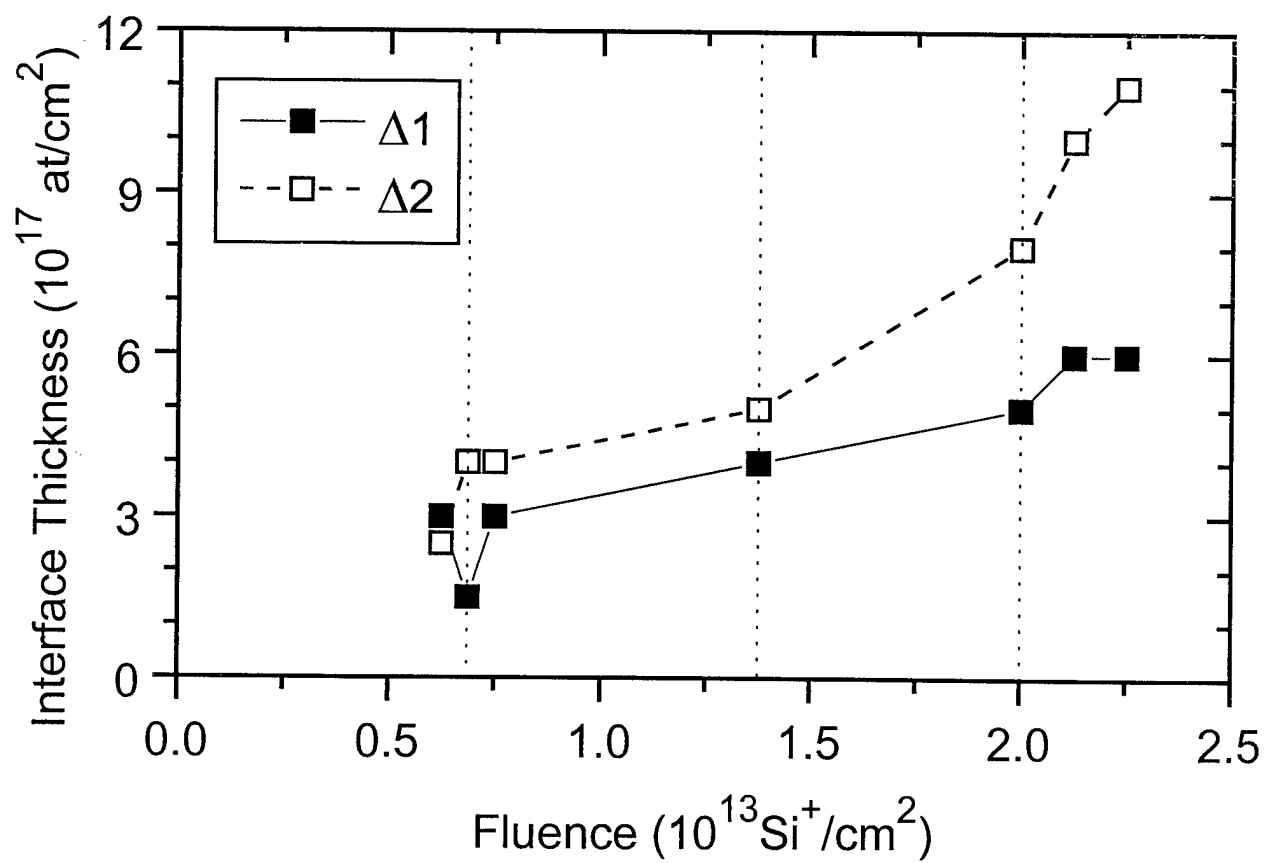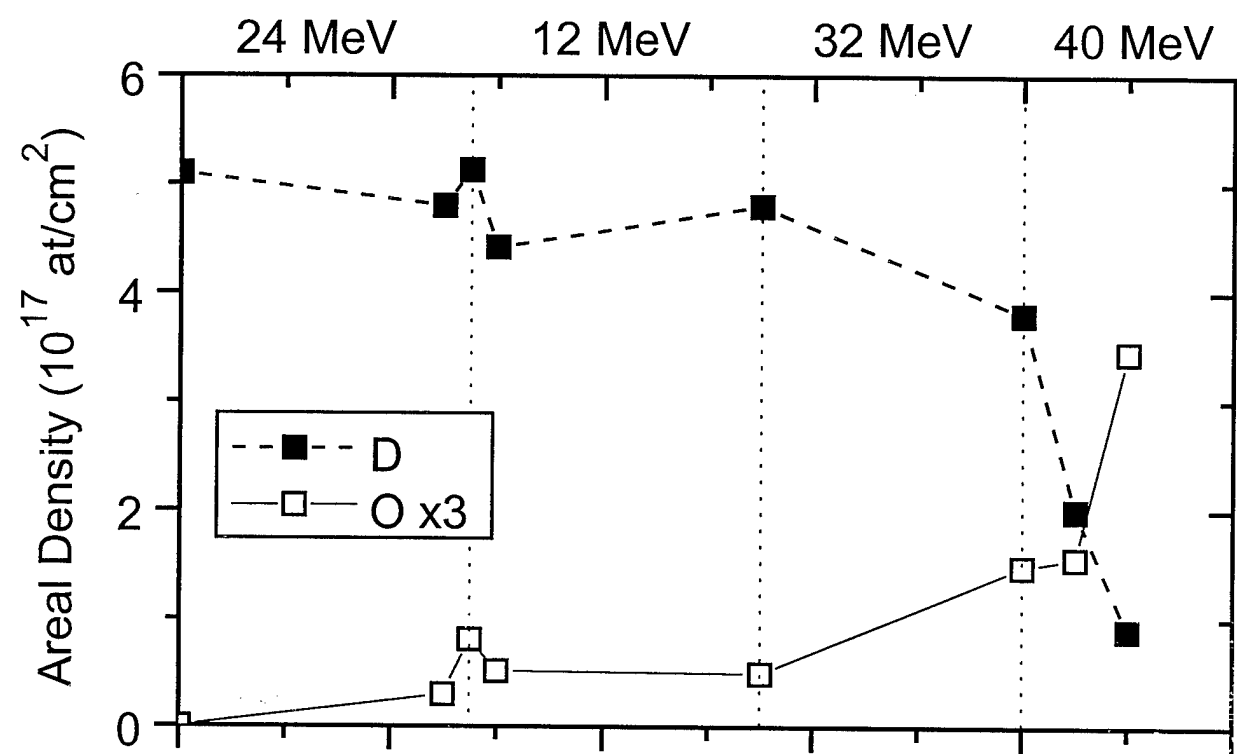
Fig. 4 Ion induced changes in the sample during Si ERD analysis top) Loss of deuterium and uptake of oxygen as function of analysing fluence. bottom) increase in interfacial transition layer thickness: Δ1 interface between a-C:D and Be$_2$C, Δ2 interface between Be$_2$C and Be sustrate

a) ^{4}He RBS

- experimental
- simulated
- C
- O
- Be

b) ^{4}He ERD

- experimental
- simulated, D

ENERGY (keV)

a) without foil

b) with foil

experimental
simulated
H
O
C
Be

COUNTS

ENERGY (keV)
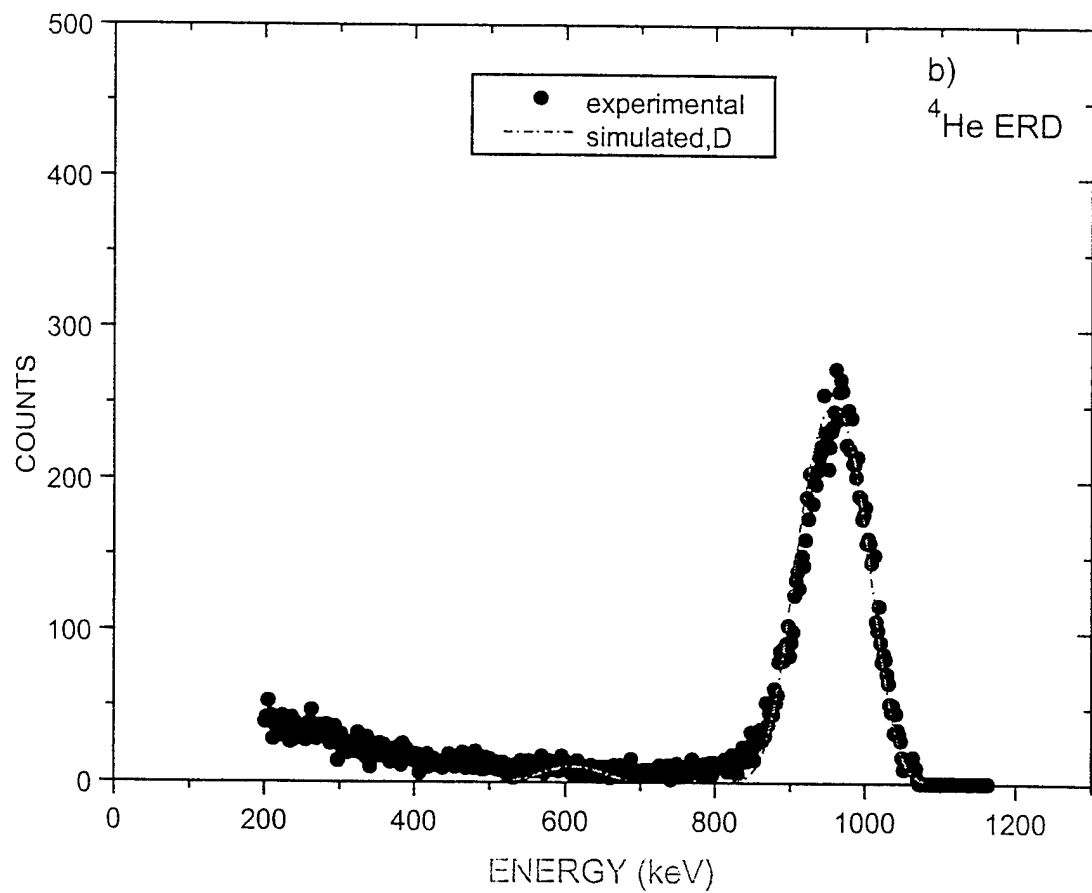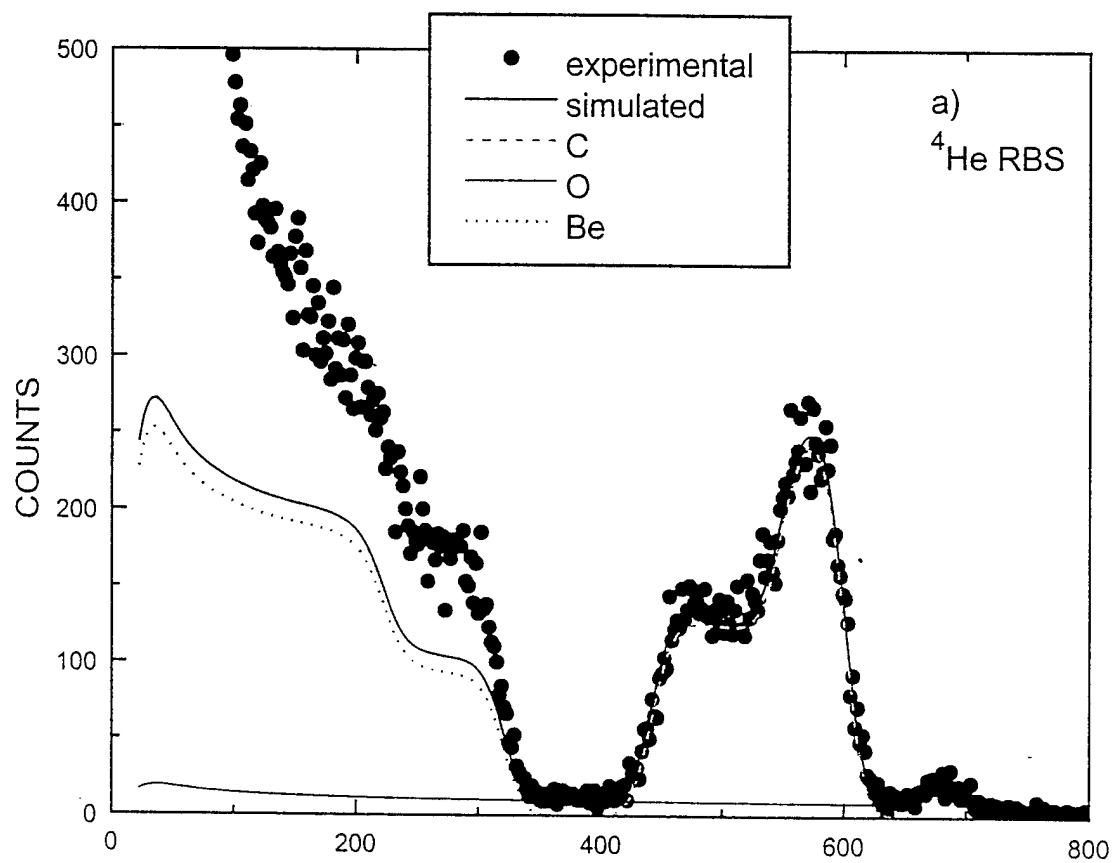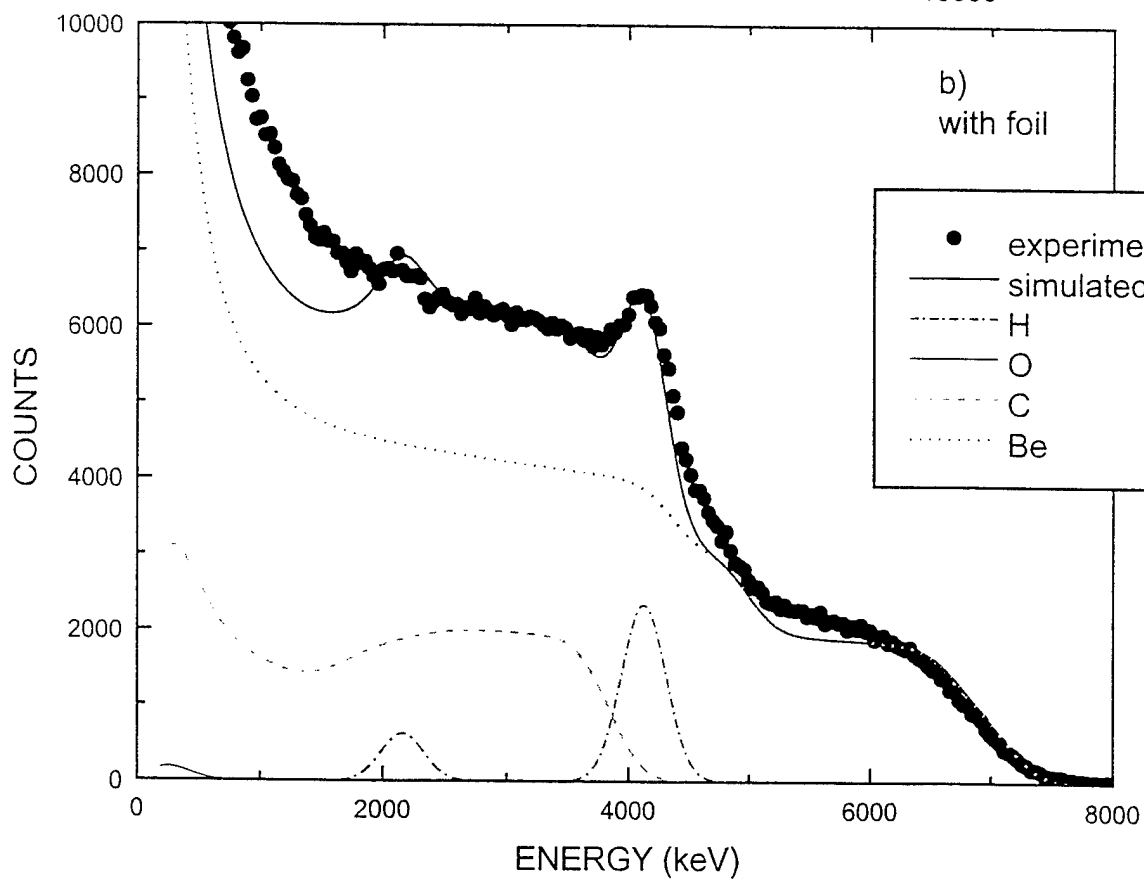
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