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Simultaneous detection of light elements by ERDA with gas-ionisation/Si ΔE–E detector telescope

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Elastic Recoil Detection Analysis (ERDA) using $\Delta E-E$ telescope with gaseous ΔE detector is carried out for the first time for simultaneous detection of H alongwith other light elements C, N, and O in Diamondlike Carbon (DLC) films. The experimental data indicated the detection limit of 0.002 at.% for H and 0.006 at.% for N and O with 10% statistical inaccuracy.

1. Introduction

Rutherford Backscattering Spectrometry (RBS) [1] and Elastic Recoil Detection Analysis (ERDA) [2] are two complementary, non-destructive, analytical nuclear techniques applied widely in materials science. ERDA is used for identification and depth profiling of light elements in thin films whilst RBS is ideal for the analysis of heavy elements in light substrates. High energy heavy ions provide powerful techniques which have been used so far only by a few groups [3–9]. It has been shown [10] that ERDA with high energy heavy ions gives excellent mass separation and even the isotopes ^{12}C and ^{13}C are separated in thin ($\sim$ 500 nm) self supporting foils. Because of overlapping recoil energies, it is difficult to identify more than one light element, particularly for thick samples. Even thin films on thick substrate are difficult to analyse for neighbouring light masses (e.g., C, N, O, etc.) in conventional ERDA. This is because of the use of a stopper foil in front of the detector to stop unwanted scattered projectiles and the recoils from thick substrate, which spoils the resolution of the detection system. The use of stopper foil eliminates the pulse pileup as well as radiation damage to detector also. If the interest is only in the lightest mass H, one can use a stopper foil of an appropriate thickness to stop unwanted heavier recoils. However, it is often desirable to get the information of all the elements in the sample. For instance, in case of Diamondlike Carbon (DLC) films, the determination of absolute H and C concentration as well as other elements present as impurities, if any, is important for better understanding of the observed properties and it helps in refining the method of preparation. Conventional ERDA, as discussed above, will not meet the requirement for multi elemental detection in DLC film deposited on thick substrate.

Simultaneous detection of different elements has been done by using (1) recoil energy and time of flight [8,11–14], (2) ΔE–E detector telescope with either a solid state transmission type detector [15–17] or a gaseous ionisation detector [18,19] as ΔE detector, (3) magnetic spectrograph [20] and (4) Bragg curve spectrometer [21]. However, only Groleau et al. [11] and Arnold Bik et al. [16,17] reported the determination of H alongwith other light elements. Groleau et al. used simultaneous measurement of energy and time of flight of recoils using surface barrier detector and a micro channel plate (MCP). The problem in the time-of-flight (TOF) experiments for H detection is its low detection efficiency. It is due to low secondary electron yield from C foil for H. Arnold Bik et al. [16,17] have demonstrated the utility of ΔE–E detector telescope to detect H, C, N and O simultaneously but no quantitative estimate of H was made. It could be due to merging of ΔE signal of H recoils with electronic noise. A transmission type surface barrier detector with a thicknes of 10 μm was used as ΔE detector. Detection of heavier recoils needs very thin ΔE detectors which are very fragile and expensive. In this work, therefore, a gaseous ionisation chamber is used as ΔE detector and a silicon surface barrier detector as E detector. Particle telescopes with gaseous ΔE detectors have considerable advantages in experiments with heavy ions, besides being inexpensive and rugged. However, light elements pose some problems because of the small size signal unless higher gas pressure or larger depth gaseous cell is used. In order to satisfactorily detect hydrogen, it is important that the height of the signal from ΔE detector lies well above the noise

pedestal. In the present experiment, we were able to get ΔE signal from H recoils well above the electronic noise. A vacuum compatible preamplifier was used inside the chamber close to the detector to optimize the signal to noise ratio. The bombarding ion and energy were selected so as to ensure that the recoils could be separated in the detector telescope over as wide energy (i.e., depth) range as possible.

The interest in DLC films emanates from their unique properties of thermal and electrical conductivity, high resistance to corrosion, high hardness etc., which have extensive industrial applications. The samples under study were prepared by two different methods. One of the samples was made by microwave discharge [22] using a 2.45 GHz generator and a plasma cavity with a Si substrate placed in a quartz tube at 10^{-1} Torr. An Acetylene and hydrogen gas mixture (1:10) was passed through the tube and discharge was established. The other sample of DLC was made by hot filament deposition technique [23]. Mixture of methane and hydrogen at a pressure of 60 Torr was used during deposition and Si substrate temperature was maintained at 900°C.

2. Experiment

Ni ions of 90 MeV from the 15 MV Pelletron [24] at Nuclear Science Centre (NSC) Delhi were utilized to analyse the DLC films deposited on Si substrate. The incident beam current was about 1 pnA. The samples were mounted in the 1.5 m diameter scattering chamber [25], where the sample mounting ladder can be moved up, down and rotated. Detector mounting tables can be rotated around the ladder with 0.05° precision. The ladder and detector tables can be rotated from outside the vacuum. The vacuum in the chamber during the experiment was 1×10^{-6} mbar. The sample was tilted at an angle of 30° with respect to the beam direction. The detector telescope was positioned to detect recoils at an angle of 37° to the beam direction, which is beyond the maximum scattering angle of 28.8° to avoid elastic scattering events from Si substrate. The detector telescope consisted of a gaseous cell of 20 mm length with 1.5 μm aluminized polypropylene foil at the entrance of the cell. Isobutane gas was used at a pressure of 200 Torr to obtain the H recoil energy loss well above the electronic noise level in ΔE gaseous detector. A charge sensitive preamplifier [26] fabricated at NSC was mounted close to the detector inside the chamber to improve the signal to noise ratio by reducing cable capacitance. The surface barrier detector with a depletion depth of 300 μm was used as E detector. It was kept inside the gaseous cell at its exit end. A tantalum collimator of 2×4 mm^2 size was placed in front of the telescope detector. It subtended

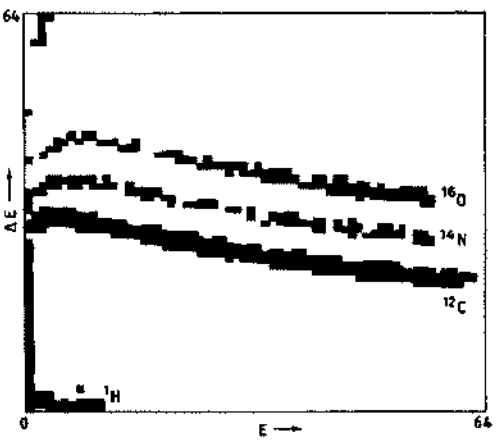

Fig. 1. Two-dimensional spectrum of recoil ions observed when 90 MeV ^{58}Ni ions were bombarded on diamondlike carbon film deposited on Si substrate. The scale of ΔE and E is in channels.

an angle of 0.2° and solid angle of 0.02 msr. The ΔE and E energy signals after processing with preamplifier and amplifier were simultaneously recorded in an eight channel NIM ADC. The strobe for ADC was derived from the stop E detector.

3. Results and discussions

3.1. Quantitative determination of constituents and contaminants

Fig. 1 shows a two-dimensional recoil spectrum observed when Ni ions were incident on the microwave plasma deposited DLC film. Recoils of H, N, C and O

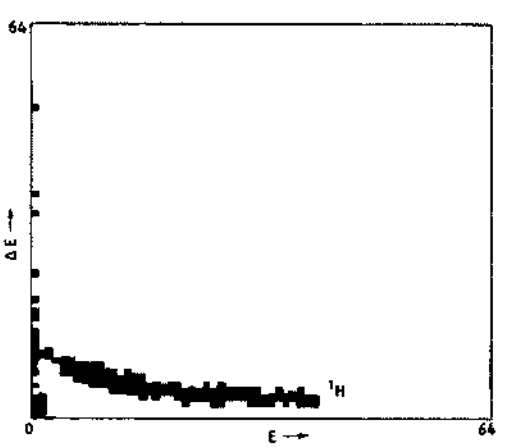

Fig. 2. Two-dimensional spectrum with increased amplifier gain of ΔE and E detectors. It clearly indicates that ΔE signal is well separated from electronic noise.

are clearly separated. Fig. 2 shows the two-dimensional spectrum of the same sample with larger gains of the amplifiers of ΔE and E detectors which indicates that H recoil band was well above the E axis. The energy loss ΔE_w and ΔE_g of the recoils (of energy E_r from the surface) in the detector window and isobutane gas respectively were calculated using TRIM. ΔE_g and $E_r - (\Delta E_g + \Delta E_w)$ thus calculated for different elements gave the calibration of ΔE and E channels respectively in the bi-dimensional plot. Total energy of the recoils were determined by $E_{total} = \Delta E + fE$, where f is dependent on the gains of the electronics in ΔE and E channels. It was calculated as suggested by Stoquert et al. [18]. New bi-dimensional (ΔE–E_{total}) plots were generated using the E_{total}. Channel to energy conversion for each recoil spectrum was done using the channel number corresponding to half the height of the edge and theoritically evaluated $E_r - \Delta E_w$. The recoil spectra of each elements were extracted by the projections of the corresponding bands of the bi dimensional (ΔE–E_{total}) spectrum. The conversion of energy to depth scale for each element was carried out using the respective recoil spectra with the help of computer code based on the formulism described by Arnold and Habraken [23]. Since the ions lose energy as they traverse the material and dE/dx is dependent on the energy, the film and stopper foil were divided in layers of thickness 1000 Å and the value of dE/dx were estimated at each layer for incident ion as well as recoil in the code. The area under recoil spectrum of each element up to a depth of 0.5 μm was taken to evaluate the elemental composition. The quantitative determination of the elements was done with the help of following relation.

$$\frac{N_X}{N_C} = \frac{Y_X}{Y_C} \frac{(d\sigma/d\Omega)_C}{(d\sigma/d\Omega)_X}, \quad (1)$$

where N is number of atoms/cm^2, Y is the area under recoil spectrum up to a fixed depth, and $(d\sigma/d\Omega)$ is Rutherford recoil cross section for elements X and C. The concentration of H, N, and O in DLC film made by microwave plasma deposition were found to be (6.3 ± 3.1%) at.%, (3.4 ± 6.9%) at.% and (8.1 ± 4.3%) at.% respectively with respect to carbon. The quoted errors are due to statistical inaccuracy. It does not include the error due to uncertainties in stopping powers. A large percentage of N and O found, could be either due to the impurities in the gases or improper flushing out of the residual gases before starting the deposition. Recoil spectra of the impurities O and N are given in Fig. 3. DLC films made by hot filament method did not have much N and O impurities. C and H recoil spectra are shown in Figs. 4 and 5 respectively. The concentration of H, N and O were found to (18.2 ± 0.7%) at.%, (0.3 ± 9.2%) at.% and (0.5 ± 6.6%) at.% respectively. The impurities of O and N are

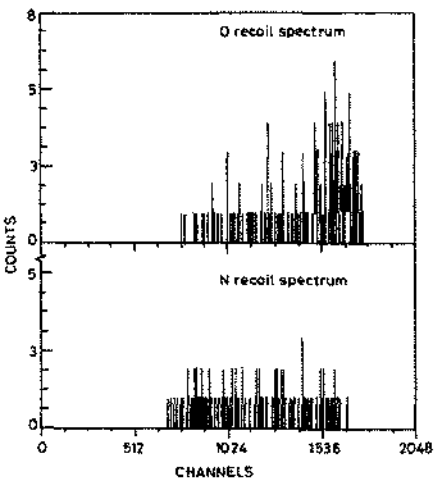

Fig. 3. Recoil spectra of impurities (N and O) of DLC film prepared by microwave plasma deposition technique.

believed to have been incorporated into the sample during the preparation as these are more or less uniformly distributed except for larger contents of O at the surface. If the impurities were due to the hydrocarbon deposition during ion irradiation, they should have been on the surface only.

It has been observed in our previous studies [27] that H content in the sample reduces under ion irradiation, which can be determined and corrected in precise H content measurement. In the present measurement, it is not accounted for.

3.2. Depth resolution and detection limit

The probing depth in the present experiment is ~ 1 μm for C and ~ 1.7 μm for H. The latter is limited by

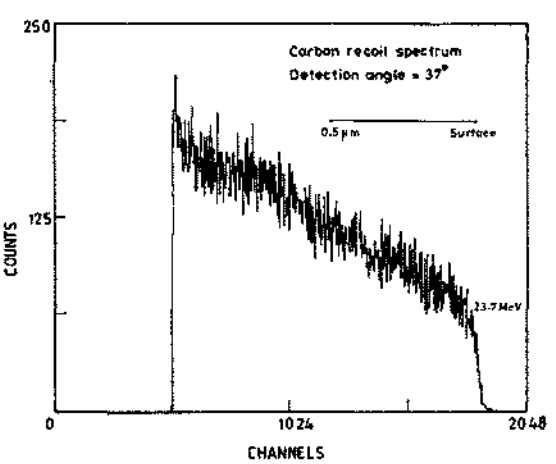

Fig. 4. Carbon recoil spectrum for the DLC film deposited by hot filament method. The depth scale is shown above the spectrum.

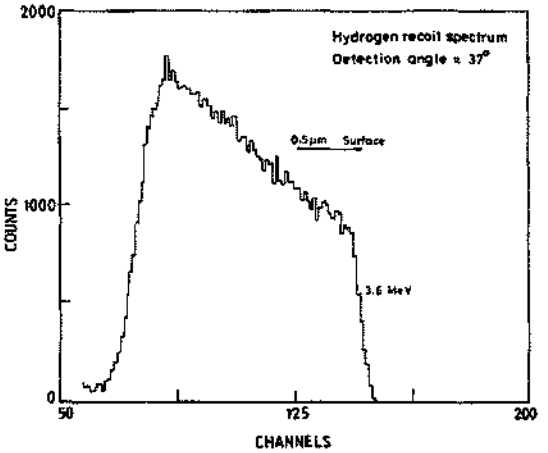

Fig. 5. Hydrogen recoil spectrum for the DLC film made by hot filament method. The depth scale is shown above the spectrum.

thickness of the film. Depth resolution at surface as 80 and 20 nm were obtained for H and C respectively. The present experiment indicates that for a moderate irradiation with integrated charge of 5 μC particle and small solid angle of 0.02 msr of the telescope detector, one can set the detection limit for H, N, O as 0.1 at.%, 0.3 at.% and 0.3 at.% respectively with a statistical inaccuracy of 10%. These can be easily improved to 0.002 at.% for H and 0.006 at.% for N and O by taking solid angle of 1 msr. However, the depth resolution will deteriorate with the increase in solid angle.

4. Conclusion

The application of ΔE gaseous and E solid state detector with ERDA technique is demonstrated to be a powerful technique for simultaneous determination of H and other light elements in thick DLC film on Si substrate for the first time. Further, it is possible to analyse even thick samples with this technique, which was otherwise not possible with conventional ERDA employing a stopper foil. On the basis of the present experimental data, it can be stated that the $\Delta E_{\text{gaseous}}$–$E_{\text{solid state}}$ detector telescope is capable of providing detection limit of 0.002 at.% for H and 0.006 at.% for O and N with a probing depth of about 1 μm. The ΔE gaseous detector has considerable advantage over the transmission-type surface barrier detector due to its being insensitive to radiation damage. Besides, it is not fragile and can be designed and fabricated according to the requirement. However, it requires a gas handling system with fine control needle valves to maintain a constant pressure in gaseous cell. The ability to vary the pressure in gas cell provides an additional advantage of giving a wide dynamic range and hence provides flexibity in use. Wide variety of high energy heavy ions available from 15 UD Pelletron [24] at NSC can produce desirable recoil energies suitable for this technique.

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