



Stopping power and energy loss straggling of thin Formvar foil for 0.3–2.7 MeV protons and alpha particles

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H I G H L I G H T S

- ▶ Stopping power and straggling data of H⁺ and He⁺ ions in Formvar were reported.
- ▶ The data were measured by using the indirect transmission technique.
- ▶ The target mean excitation and ionization potential was deduced from proton data.
- ▶ Straggling data were corrected from target thickness inhomogeneity via AFM (Atomic Force Microscopy) analysis.

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Stopping power and energy loss straggling data for protons (¹H⁺) and alpha particles (⁴He⁺) crossing Formvar thin polymeric foils (thickness of ~0.3 μm) have been measured in the energy range (0.3–2.7) MeV by using the indirect transmission technique. The determined stopping power data were compared to SRIM-2010, PSTAR or ASTAR calculation codes and then analyzed in term of the modified Bethe–Bloch theory to extract the target mean excitation and ionization potential $\langle I \rangle$. A resulting value of $\langle I \rangle \approx (69.2 \pm 1.8)$ eV was deduced from proton stopping data. The measured straggling data were corrected from surface roughness effects due to target thickness inhomogeneity observed by the atomic force microscopy (AFM) technique. The obtained data were then compared to derived straggling values by Bohr's and Bethe–Livingston's classical theories or by Yang's empirical formula. A deviation of ~40%–80% from the Bohr's straggling value has been observed for all reported energies, suggesting that the Bohr theory cannot be correctly applied to describe the electronic energy loss straggling process with the used low thickness of Formvar foil. The inner-shell contribution of target electrons to energy loss process is also advanced to explain the observed deviation from experiment in case of He⁺ ions. Finally, the reliability of Bragg's additivity rule was discussed in case of stopping power and straggling results.

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1. Introduction

As a part of our continuous work on the slowing down of light and heavy ions in matter and more specifically in polymers (Ammi et al., 1997, 2002, 2011), the present study has once again enriched the existing database of main physical parameters governing the ion energy loss process in compound materials. Such process occurs when energetic ions penetrate into the target material and lose their

kinetic energy mainly by inelastic electronic collisions, leading to the excitation or ionization of target atoms and to the energy spreading of the emerging ions due to statistical nature of the induced collisions. The basic physical quantities used to describe this process are the mean energy loss per length unit or stopping power parameter and the variance of energy spreading around the mean energy loss or energy straggling. Over several decades, a large number of theoretical and experimental works (Ziegler, 1980, 1999; Paul, 1999; Besenbacher et al., 1980; Yang et al., 1991; Sigmund, 2006; and references therein) have been performed on the stopping power and energy straggling parameters for a great variety of ions and target materials of different natures. In particular,

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the stopping power and straggling of swift light ions in compound materials have been received special attention in recent years (Shiomi-Tsuda et al., 1995; Ammi et al., 2000, 2004, 2006; Raisanen et al. 2003; Yu et al., 2006; Diwan et al., 2006) due to their widespread use in materials science and ion beam applications. In case of protons crossing $(C_xH_yO_z)_n$ polymeric foils, the measured stopping power data have been widely analyzed (Ammi et al., 2005; Shiomi-Tsuda et al., 1995; Raisanen et al. 2003; Porter, 1989) within the framework of the modified Bethe–Bloch stopping theory (Ziegler, 1999) and the famous Bragg additivity rule (Bragg and Kleeman, 1905) in order to extract the target mean excitation and ionization potential $\langle I \rangle$. Concerning the energy straggling parameter, numerous experimental measurements have been performed in recent years (Ammi et al., 2006; Yu et al., 2006; Diwan et al., 2006, 2007; Pratibha et al., 2008; Neetu et al., 2010) for both light and heavy ions traversing polymeric foils. Also, there have been different theoretical models initially developed to describe the energy loss straggling in elemental targets and then extended to compound materials by using the Bragg additivity rule. Thus, a simple energy-independent formula proportional to the target thickness has been firstly proposed by Bohr (Bohr, 1948) in high energy region ($v > v_0 Z_1^{2/3}$ with $v, v_0 = c/137$ and c being the ion, Bohr and light velocities, respectively, and Z_1 the ion atomic number). Then, other theories and semi-empirical formulations have been also developed (e.g., Livingston and Bethe (1937), Lindhard and Scharff (1953), Yang et al. (1991)) and applied to experimental data where the corresponding calculated straggling tends asymptotically to Bohr's formula at high energies. Most of these approaches were often found to predict straggling values in varying level of agreement with a limited number of experimental data. Moreover, reliable experimental data for various ion-polymer combinations and incident energy ranges are still needed to improve the available theoretical formulations in order to describe well the energy loss process within all its aspects.

In the present study, we report on the stopping power and energy loss straggling measurements for H^+ and He^+ ions crossing thin Formvar foils (thickness of $\sim 0.3 \mu m$) over the ion incident energy range of (0.3–2.7) MeV. Notice that for this polymer only few experimental stopping power data are available (Munnik et al., 1997; Damache et al., 2010) in case of H^+ and He^+ ions over the MeV energy region, while no measured straggling values are found in the literature to our knowledge. Through this investigation, we would like also to provide useful information about the limited use of the Bohr classical theory to correctly describe the energy loss straggling process in the case of very thin foils. To carry out stopping power and energy straggling measurements, we have adopted a previously improved experimental method (Ammi et al., 2002, 2006) based on the Rutherford backscattering spectrometry and the transmission of the secondary produced ions for data analysis. Then, the target mean ionization and excitation potential of the studied Formvar foil has been also determined by analyzing the measured proton stopping data within the Bichsel formalism based on Bethe–Bloch's stopping theory (Ziegler, 1999; Ammi et al., 2005). Finally, the measured energy loss straggling data have been interpreted in terms of the Bohr and Bethe–Livingston classical theories or Yang et al.'s empirical formula by using the Bragg additivity rule. Notice that before comparison, the correlation effects due to the target thickness inhomogeneity have been incorporated as refinement in these theories.

2. Experimental

The experimental method used in this work to perform stopping power and energy straggling measurements has been

already well described in previous papers (Ammi et al., 2002, 2004, 2006). It consisted briefly to the combination of both Rutherford backscattering spectrometry and transmission techniques with and without thin Formvar foil covering a surface barrier detector placed at 165° relative to the primary ion beam direction. The energy resolution of the used detector is of ~ 12 keV. This indirect transmission technique allows to measure in one run the energy loss and energy straggling of the secondary ions, permitting, hence, to avoid uncertainties due to beam fluctuations and thin target degradation. The used Formvar foils were prepared from a polyvinyl formal powder of high purity (99.99%) purchased from SIGMA-Aldrich (South Africa) Company. It was diluted in chloroform at a concentration of 1.5% in solution. A drop of this solution was dropped on water surface and a thin foil was created. The thin foil was allowed to dry and then was scooped from the water and gently put in a clean aluminium holder. The target thickness was determined by using the energy loss of 5.485 MeV alpha particles from a very thin ^{241}Am radioactive source passing through the Formvar foil (Ammi et al. 2006). Referring to tabulated stopping power values (Ziegler et al., 1985), the areal mass density (in $\mu g/cm^2$ unit) of the used foil was determined. Its mean value is found of $(36.7 \pm 2.5) \mu g/cm^2$ that corresponds to a linear thickness of $(0.3 \pm 0.02) \mu m$ by assuming a mass density of $1.23 g/cm^3$ for polyvinyl formal. The latter has a chemical formula $(C_5H_8O_2)_n$ with (8 H–C, 4 C–C and 4 C–O) single bonds. Finally, the surface roughness and thickness homogeneity of the studied Formvar foil were investigated by the atomic force microscopy (AFM) analysis technique.

3. Data treatment, results and discussion

3.1. Stopping power data and analysis

As described in our previous papers (Ammi et al., 2002, 2004), two typical spectra were recorded for each incident energy showing two energy peaks corresponding to the backscattered ions detected with and without the Formvar foil. The experimental values of energy loss (ΔE) were deduced from the channel shift measurement of the energy peak. Thus, the stopping power $S(\bar{E})$ at a mean energy $\bar{E} = E_i - \Delta E/2$ was determined in $(MeV mg^{-1} cm^2)$ unit by dividing the energy loss on the target areal density, $S(\bar{E}) = \Delta E/\rho \Delta x$, where ρ and Δx are the mass density and linear thickness of the studied foil, E_i is the initial energy of the backscattered ions. It should be noted that all determined $S(\bar{E})$ data within this method are relative to a tabulated stopping power value for 4.485 MeV alpha particles in Formvar. Then, the absolute uncertainties in the determined stopping powers were estimated by considering the propagated errors originating from the measured energy losses ($\sim 4.6\%$, in average) and from the target areal density ($\sim 6.8\%$). The obtained stopping power values and their associated uncertainties are reported in Table 1 for both H^+ and He^+ ions in Formvar foil. These data are then plotted in Fig. 1 versus ion incident energy where they are compared to calculated stopping values from SRIM-2010 (Ziegler et al., 1985), PSTAR or ASTAR (Berger et al., 2005) computer codes as well as to other experimental data taken from literature. It should be noted that the calculation codes used here are mainly based on the compilation of extensive experimental stopping power data supplemented by Bragg's additivity rule in case of complex compound materials. As can be seen in Fig. 1, our $S(E)$ data show an overall agreement, within the experimental uncertainties, with the previous reported data from literature. In addition, our measured data for protons are in reasonably good agreement (within $\sim 8.9\%$ and $\sim 5.7\%$, in average, respectively) with SRIM and PSTAR calculated stopping values. However, those of alpha particles appear to be best described ($\sim 2.9\%$ and $\sim 2.5\%$, in average)

Table 1

Experimental stopping power data S_{exp} (in $\text{MeV mg}^{-1} \text{cm}^2$ unit) and energy straggling Ω_{exp}^2 (in keV^2 unit) versus incident kinetic energy for $^1\text{H}^+$ and $^4\text{He}^+$ ions crossing Formvar foil.

$^1\text{H}^+$ ions				$^4\text{He}^+$ ions			
$\bar{E}$ (MeV)	$\Delta E/E$ (%)	S_{exp} (MeV/ mg/cm^2)	Ω_{exp}^2 (keV^2)	$\bar{E}$ (MeV)	$\Delta E/E$ (%)	S_{exp} (MeV/ mg/cm^2)	Ω_{exp}^2 (keV^2)
0.32	6.7	0.58 ± 0.05	10.2 ± 2.7	0.88	9.0	2.17 ± 0.17	39.2 ± 3.4
0.38	4.9	0.51 ± 0.04	6.0 ± 1.5	1.07	7.1	2.07 ± 0.16	57.3 ± 6.2
0.48	3.3	0.43 ± 0.04	9.4 ± 2.3	1.26	5.7	1.96 ± 0.15	75.8 ± 9.4
0.58	2.4	0.38 ± 0.03	4.7 ± 0.5	1.44	4.8	1.87 ± 0.15	122.9 ± 12.0
0.68	1.9	0.35 ± 0.03	8.3 ± 1.5	1.63	3.9	1.74 ± 0.13	76.9 ± 7.8
0.78	1.5	0.32 ± 0.03	10.2 ± 1.2	1.82	3.3	1.64 ± 0.13	92.7 ± 10.3
0.88	1.2	0.29 ± 0.03	6.2 ± 0.7	2.00	2.8	1.53 ± 0.12	63.0 ± 9.4
0.98	1.0	0.27 ± 0.03	4.4 ± 0.6	2.19	2.6	1.52 ± 0.12	39.2 ± 4.1
1.07	0.9	0.26 ± 0.02	8.0 ± 1.0	2.38	2.1	1.38 ± 0.11	48.7 ± 5.4
1.17	0.8	0.25 ± 0.02	4.7 ± 0.6	2.56	1.9	1.30 ± 0.10	40.3 ± 5.1
1.27	0.6	0.21 ± 0.02	2.5 ± 0.3				
1.37	0.6	0.20 ± 0.02	3.3 ± 0.5				
1.56	0.5	0.19 ± 0.03	8.3 ± 1.7				
1.76	0.4	0.18 ± 0.03	2.5 ± 1.3				
1.96	0.3	0.16 ± 0.03	8.0 ± 1.3				
2.15	0.3	0.17 ± 0.02	5.8 ± 1.4				
2.35	0.2	0.14 ± 0.02	8.0 ± 1.8				
2.55	0.1	0.10 ± 0.02	8.7 ± 2.2				
2.74	0.2	0.12 ± 0.02	4.8 ± 0.9				

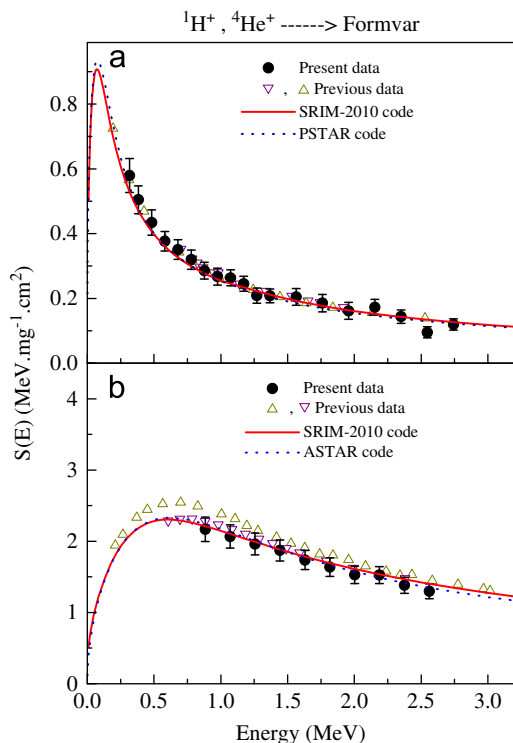


Fig. 1. Experimental stopping power values in ($\text{MeV mg}^{-1} \text{cm}^2$) unit: (a) for $^1\text{H}^+$ and (b) for $^4\text{He}^+$ ions (scatter) in Formvar foil, together with SRIM-2010 (solid lines), PSTAR or ASTAR (dash lines) codes and previous data (scatter) from literature (Munnik et al., 1997; Damache et al., 2010).

by the latter calculation codes (i.e., SRIM and ASTAR) over the whole investigated energy range.

Besides, the experimental proton stopping power data were then analyzed in the framework of Bethe–Bloch's stopping theory supplemented by the Bragg additivity rule in order to extract the target mean excitation and ionization potential, $\langle I \rangle$, for Formvar foil. As in our previous work (Ammi et al., 2005), we used a

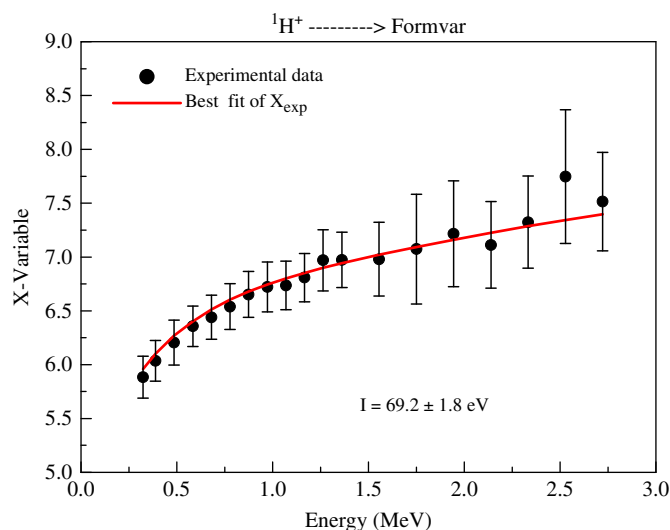


Fig. 2. The best fit of X_{the} variable to the X_{exp} curve that an $\langle I \rangle$ value of 69.2 ± 1.8 eV for protons in Formvar foil.

modification of Bichsel's X-variable obtained from the reduced experimental stopping power data:

$$X_{exp} = \ln \left(\frac{2m_e v^2}{1 - \beta^2} \right) - \beta^2 - \frac{\beta^2 S_{exp}(E)}{\kappa Z_1^2 Z_2} \quad (1)$$

or from the theoretical Bethe–Bloch formulation:

$$X_{the} = \ln(I) + \frac{C}{Z_2} + \frac{\delta}{2} - Z_1 L_1 - Z_1^2 L_2 \quad (2)$$

with $\beta = v/c$ being the ion to light velocity ratio, m_e the electron mass and κ a constant expressed in unit of (MeV mg cm^{-2}). The other quantities in Eq. (2), $\langle C/Z_2 \rangle$ and $\langle \delta \rangle$ are, respectively, the sum of target shell corrections that can be evaluated following the method of Ziegler (1999) and the density effect correction significant only for relativistic energies (Sternheimer et al., 1984). $Z_1 L_1(\beta)$ is a secondary correction term due to the Barkas effect estimated from Ziegler's empirical formulation (1999). $Z_1^2 L_2(\beta)$ is the Bloch correction term which has a small contribution to the stopping power (Ziegler, 1999) over this investigated energy range. The physical parameters defined in Eqs. (1) and (2) were evaluated for Formvar compound target by assuming the validity of Bragg's additivity rule in stopping power as in Ammi et al. (2005), Shiomi-Tsuda et al. (1995). Then, the value of the mean excitation energy $\langle I \rangle$ for Formvar foil was taken as an adjustable parameter (see Eq. (2)) and determined from X_{the} by the least squares fit of the X_{exp} (see Fig. 2), the obtained result is $\langle I \rangle = 69.2 \pm 1.8$ eV. No available experimental data were found in the literature to make a possible comparison with the present extracted value, to our knowledge. Finally, in regards to our recently reported $\langle I \rangle$ -values for Mylar, Polypropylene, Makrofol and LR-115 polymeric foils (Ammi et al., 2005), the present result of Formvar foil came once again to validate our previously adopted theoretical approach for the determination of the mean excitation energies from experimental stopping data.

3.2. Energy straggling data

The experimental energy loss straggling values have been also determined from the recorded energy loss spectra by fitting all measured energy peaks within a Gaussian distribution function as in our previous works (Ammi et al., 2002, 2004). All the measured energy loss distributions were practically of Gaussian shapes

(Besenbacher et al., 1980) and the energy peak width δE_{exp} at each mean energy $\bar{E} = E_i - \Delta E/2$, is thus determined by using the following relation:

$$\delta E_{\text{exp}}^2 = \delta E_{\text{with}}^2 - \delta E_{\text{without}}^2 \quad (3)$$

where δE_{with} and $\delta E_{\text{without}}$ are the full peak widths at half maximum (FWHM) corresponding to the backscattered ions detected with and without the Formvar foil, respectively. The energy loss straggling Ω in terms of standard deviation is obtained by dividing the measured energy peak widths δE_{exp} by the conventional factor $\sqrt{8 \ln 2} = 2.355$. Then, the measured energy loss straggling values have been corrected for the correlation effects due to foil roughness and thickness inhomogeneity by adopting the Besenbacher et al. approach (1980):

$$\Omega^2 = \Omega_{\text{exp}}^2 - \delta^2 \Delta E^2 \quad (4)$$

where Ω_{exp}^2 is the measured energy straggling and $\delta = \sigma/\Delta x$ is the standard deviation of the foil thickness relative to its mean thickness that can be estimated from the target surface roughness. Thus, a number of AFM scans were carried out on the surface of Formvar foil and the resulting images were analyzed within AFM treatment. An AFM image in 3D of the Formvar surface foil is shown in Fig. 3. The mean surface roughness (σ) value of the foil was estimated of about ~ 10 nm. This value has been then injected in our corrected straggling calculations (Eq. (4)). The present straggling corrections within AFM analysis are in agreement with those obtained from our previous method (Ammi et al., 2006) based on the statistical variations in energy loss measurements.

Finally, the absolute uncertainties associated to the measured energy loss straggling data are carefully evaluated by considering the propagated errors originating from the different quantities in Eqs. (3) and (4). The measured straggling data and the associated uncertainties for both H^+ and He^+ ions in Formvar foil are also reported in Table 1. These data are then plotted in Fig. 4 versus incident energy where they are compared to calculated straggling values derived from Bohr's and Bethe–Livingston's classical theories and Yang's empirical formula. It should be noted that both the experimental straggling values and the calculated ones were normalized to the Bohr straggling value in order to remove the thickness dependence and displaying only the beam energy variation. Also, the latter predicted theories were applied in case of Formvar polymeric compound similarly as proceeded

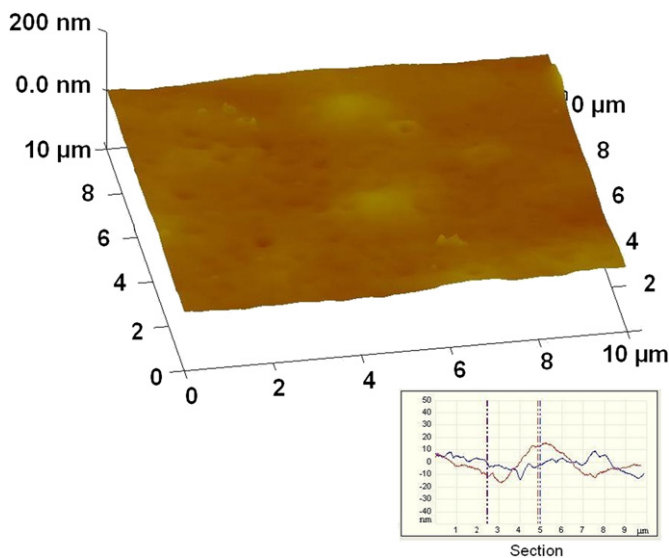


Fig. 3. AFM image (in 3D) of the surface of the used Formvar foil surface. An AFM section analysis is also included in order to check the thickness homogeneity of the studied foil.

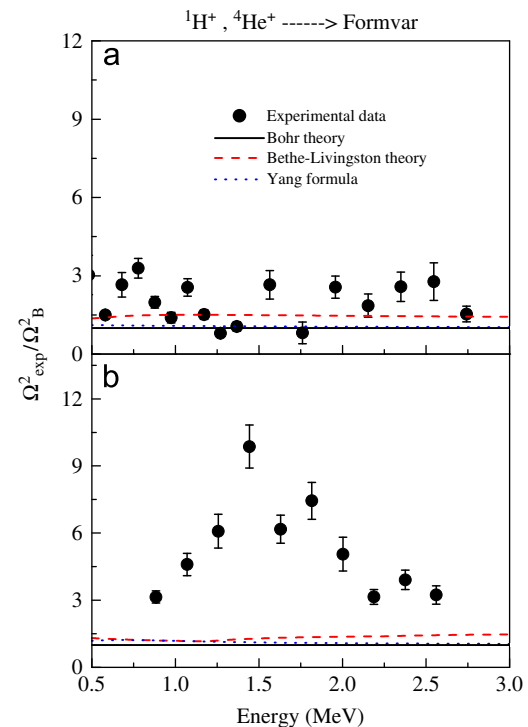


Fig. 4. Experimental reduced energy straggling data ($\Omega_{\text{exp}}^2/\Omega_{\text{B}}^2$) for (a) H^+ and (b) He^+ ions in Formvar foil versus ion energy, compared to straggling predicted values by Bohr, Bethe–Livingston theories and Yang's empirical formula.

previously (Ammi et al., 2002, 2004) and by using the Bragg additivity rule. First, examining the straggling variations versus incident energy shown in Fig. 4, one can observe the following trends. The measured data for protons are roughly independent of incident energy, whereas those of alpha particles increase up to an energy value of ~ 1.4 MeV and then decrease roughly closer to Bohr's theory at high energies. The latter trends also observed previously in case of light (H^+ , He^+) ions crossing solid metallic targets (Besenbacher et al., 1980; Arbó et al., 2002) and other polymeric foils (Ammi et al., 2006; Yu et al., 2006) are likely due to the fact that at high energies all target electrons are considered as free and contribute to the energy loss as suggested by Bohr's theory. As can be seen in Fig. 4, the calculated Bohr straggling value seems to exhibit only a limited consistency (to within $\sim 47.7\%$, in average) with the proton measured data and substantially deviates (to within $\sim 68.2\%$ at 0.9 MeV that increases to $\sim 89.9\%$ at 1.4 MeV then decreases to ~ 69.1 at 2.6 MeV) from those of alpha particles for all reported energies. However, these discrepancies can be reduced up to $\sim 39.5\%$ (for H^+ ions) and $\sim 71.3\%$ (for He^+ ions), in average, when using the Bethe–Livingston theory that takes into account the binding of target electrons within atomic shells. The Yang et al. empirical formula based on the Chu dielectric calculation (Chu, 1976) by including the charge exchange effects generates practically the same straggling values as Bohr's theory over the whole investigated energy range. The observed discrepancies in Fig. 4 between the measured data and the predicted theories are presumably due to the use of very thin Formvar foil (within a thickness of $\sim 0.3 \mu\text{m}$) as absorber in the present experiments, resulting in low fractional energy losses ($\Delta E/E < 10\%$, see Table 1) for all incident ion energies. In such conditions, Bohr's theory cannot be correctly applied to describe the energy loss straggling within the latter energy loss regime. This is in agreement with a previously reported work (Bichsel and Saxon, 1975) on electronic energy straggling in thin absorbers. Such explanation could be also supplemented by the assumption related

to the inner-shell contribution of target electrons to energy loss process not considered by Bohr's theory and significant in case of He^+ ions at low energies (Arbó et al., 2002; Araujo et al., 2002; Ammi et al., 2006). An evidence of such assumption is the substantial deviation (up to $\sim 90\%$) observed for He^+ ions at low energies compared to the better agreement (to within $\sim 47.7\%$, in average) obtained for the proton straggling data for all reported energies (see Fig. 4). Finally, in spite of the observed disagreement between the measured straggling data and the theoretical predictions, the Bragg additivity rule has been successfully applied in the present study both for stopping power and energy loss straggling parameters.

4. Conclusion

In the present study, we have summarized the results obtained by our group concerning the fundamental parameters characterizing the slowing down of light ions in Formvar polymeric foil, such as the stopping power $S(E)$, the mean excitation energy $\langle I \rangle$ and the energy loss straggling $\Omega(E)$. The experimental stopping power and energy loss straggling values have been measured by using the indirect transmission technique implemented at the CRNA Van de Graaff accelerator of Algiers. The obtained experimental results have been analyzed in terms of the modified Bethe–Bloch theory for stopping power parameter and with theories describing electronic energy loss straggling phenomena. Deviations of $\sim 40\%$ to $\sim 80\%$ have been observed between measured and calculated energy loss straggling values. This deviation should be attributed to the thin foil of Formvar ($0.3\ \mu\text{m}$) used as absorber in this work. The used thin Formvar foil induced a low energy loss regime ($\Delta E/E < 10\%$) where the Bohr theory cannot be correctly applied to describe electronic energy loss straggling. The inner-shell contribution of target electrons to energy loss process could be also associated with the latter suggestion to explain the significant discrepancies observed in case of He^+ ions. The AFM scans of Formvar surface foil have been used to correct foil roughness effect to straggling measurements.

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