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Energy loss straggling of $(0.5 < E_p < 2.0)$ MeV protons in formvar

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ABSTRACT

Energy loss distributions for $(0.5 < E_p < 2.0)$ MeV protons traversing polyvinyl formal have been measured in transmission. Then, they have been analyzed in order to determine energy loss straggling variance data. For avoiding non-stochastic broadenings and single collision events, only energy loss fractions within the range $2\% \leqslant \frac{\Delta E}{E} \leqslant 20\%$ have been considered. The inferred energy loss straggling data are compared to values derived by several theories of the collisional energy straggling and by Yang et al. empirical formula with assuming the validity of the Bragg–Kleeman additivity rule for compounds in all the performed calculations. The obtained results are discussed with distinguishing two projectile velocity regimes delimited by the proton energy $E_p \sim 1.2$ MeV. Over the high proton velocity regime, our data are in very consistent with the classical Bohr theory and the Yang et al. empirical formula predicting constant collisional energy loss straggling. It clearly appears that over the low proton velocity regime, our energy loss straggling data are in best overall quantitative agreement with values predicted by the Sigmund–Schinner binary collision stopping theory (the BCAS) involving both the shell and Barkas–Anderson corrections. Besides, the slight low energy-dependent behavior of experimental data shows to be consistent with the predictions of the Bethe–Livingston theory and the Yang et al. empirical formula.

1. Introduction

The slowing down of swift charged particles in matter is dominated by electronic collisions implying ionization and excitation processes of the stopping medium. The best understanding of these processes is required both in fundamental research and in many applications of charged particle beams, such as in medical and health physics [1–3], in ion beam modification of materials [4,5], in materials science and engineering [6–8]. Two main fundamental quantities used to quantify the energy deposition process are the stopping force and the energy loss-straggling characterizing, respectively, the ion mean energy loss per travelled path-length and the associated variance. While the former quantity has been extensively investigated [9–13] for a large variety of ion-target combinations, the latter received rather much less attention, so that energy loss straggling data are scarce or uncertain, in particular for compound and composite targets. Indeed, conversely to the mean energy loss by incident ions which can be reliably measured, energy loss straggling measurements are much more sensitive to target non-uniformities and non-homogeneities that may introduce substantial additional broadening to the real energy loss straggling variance. Experimentally, these spurious contributions are difficult to evaluate and, then, special precautions must be taken for reducing their effect to a negligible level for performing reliable energy loss straggling measurements. In the present paper, we report on energy loss straggling measurements for ∼(0.5–2.0) MeV protons traversing a thin polyvinyl formal foil. The derived energy loss straggling data are compared to the predictions of Bohr [14] and Bethe–Livingston [15] classical theories, and to the empirical formula of Yang and O'Connor [16]. Besides, they are discussed in comparison to values generated by the code PASS [17] supporting the Binary Collision Approximation Scheme (BCAS) of Sigmund and Schinner [18,19]. All calculated energy loss straggling values have been performed with assuming the validity of the Bragg–Kleeman additivity rule [20] for compound materials.

2. Experimental set up and procedure

The experimental set up and procedure are as those used previously [21] for measuring the stopping power of polyvinyl formal for ∼(0.2–3.4) MeV/amu $^1H^+$ and $^4He^+$ ions. For avoiding non-stochastic broadening and deriving reliable energy loss straggling

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data, only energy loss spectra corresponding to energy loss fractions, $\Delta E/E$, in the range ~2% up to ~20% have been considered here. Therefore, only a brief report on main experimental aspects is given below in this section. Further experimental details can also be found elsewhere [22–24].

2.1. Set up and measurements

Incident H^+, H_2^+ and H_3^+ ion beams were produced by the Algiers 3.75-MV Van de Graaff accelerator with energy resolution of ~0.1% (see reference [25]) and mean current intensity value of ~30 nA. The primary ions were first backscattered off a very thin Au–Si target placed under high vacuum at the center of the scattering chamber, then they passed through the scanned formvar foil before being detected by a 500 µm-thick ULTRA ion implanted Si detector set at a laboratory angle of 165° relative to the primary incident direction. The analyzed ion beams were collimated by two slits of 1.5 mm and 3.0 mm in diameters placed, respectively, at the entrance of the chamber and in front of the detector. In each experimental run, ion energy loss distributions obtained with (straggled ions) and without the polymer target sample in place were successively recorded under the same pressure of $\sim 3 \times 10^{-6}$ mbar using a special target holder, as detailed in Ref. [22]. Typical such energy loss distributions taken at energy $E = 1176$ keV of the secondary proton beam are shown in Fig. 1 ((a) for un-straggled protons and (b) with for straggled protons). Curves of Gaussian shapes were fitted to experimental energy spectra over energy intervals of ~3.5 times the standard deviations around the particle peaks and are also reported in this figure.

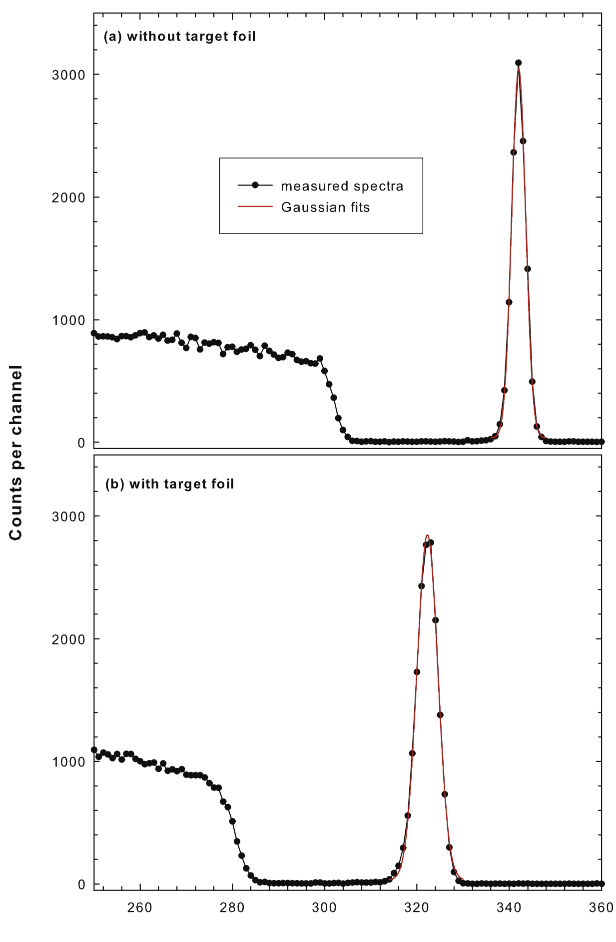

Fig. 1. Typical energy loss spectra (solid circles) and their corresponding Gaussian fits (red solid curves) recorded with and without the formvar sample in place for 1.2 MeV incident protons using a 2048-channel portion of an Ortec MCB Card. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

2.2. Target thickness determination

The used formvar target foil was supplied by the Chemistry Laboratory of the InESS/CRNS of Strasbourg (France) with stated thickness of ~2.5 µm. It is a single-bonded polymer compound of chemical formula $(C_5H_8O_2)_n$ and density value of 1.31 g/cm^3. Its elemental composition, expressed in weight fraction, is as follows: C: 0.5998, H: 0.0805, O: 0.3197. The film thickness and non-uniformity of the target sample have been investigated in a previous contribution [21] through systematic energy loss measurements for alpha particles [22,24,26] delivered by a very thin mixed ^{241}Am–^{239}Pu–^{233}U radioactive source. Since the foil thickness and non-uniformity are the main factors affecting the measurement of energy loss straggling widths, a special attention has been focused on their determination. These measurements were performed under the same experimental conditions as in proton energy loss measurements with the mixed radioactive source placed in front of the Au-scattering target and the accelerator beam switched off. This procedure ensured that the same formvar foil thickness was crossed by the ion beam both in alpha particle and proton energy loss measurements. Fig. 2 reports typical energy loss spectra for alpha particles from the mixed radioactive source that have been recorded over an advanced 4096-channel pulse height analyzer without (a) and with (b) the formvar target foil being exposed to the secondary proton beam. Multiple Gaussian shapes were fitted to the experimental energy spectra, thus allowing a precise determination of the alpha particle peak positions and are also shown in Fig. 2. The *R*-squared values for all multi-Gaussian fits (red curves in Fig. 2) were better than 99%. In the energy calibration of the detection system, all adjusted alpha particle peaks in Fig. 2(a) (solid and dashed blue curves) were considered while only the peaks of highest energies (solid blue curves in Fig. 2) produced with branching ratios higher than 80% from the ^{241}Am ($E_\alpha = 5485$ keV) and ^{233}U ($E_\alpha = 4824$ keV) radio-isotopes were considered for the determination of the formvar target foil thickness. The positions of these two alpha particle peaks were then deduced from the Gaussian fits with a relative uncertainty better than 0.01%. The formvar foil thickness was determined from the measured ΔE alpha particle energy loss data and corresponding $S(\bar{E})$ stopping power values computed by the SRIM-2008 code assuming the validity of the Bragg–Kleeman additivity rule at mean energy, $\bar{E} = E - \frac{\Delta E}{2}$ (with E being the energy of incident alpha particles). As a result, a mean areal thickness value, $\bar{x} = 277.5$ µg/cm^2, was derived with a relative uncertainty of ~2.5%. This latter uncertainty mainly results from that affecting the computed SRIM stopping power values assumed here to be of ~2%. Indeed, for $E > 1$ MeV/amu where the Bethe stopping theory is very reliable, the scatter of the measured data relative to the SRIM predictions for alpha particles in C, O and H elements is low [11], which supports the assumed SRIM accuracy. The non-uniformity of the formvar target foil was first investigated on a macroscopic scale through systematic energy loss measurements for 5.485 MeV alpha particles from the ^{241}Am radio-isotope over the formvar sample area. Consequently, the obtained ΔE energy loss data lie within a standard deviation better than 1.5%. In addition, the formvar sample was investigated at a microscopic level following the procedure [27] based on the use of a single ion specy to evaluate thickness inhomogeneity in thin films. Thus, in the same manner as in Ref. [27], an upper limit of the formvar foil roughness coefficient, $\rho = \sigma/\bar{x}$ (with σ being the foil thickness