Nuclear Instruments and Methods in Physics Research B 299 (2013) 51–53

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Comments on recent measurements of the stopping power of liquid water

Rafael Garcia-Molina [a], Isabel Abril [b], Pablo de Vera [b], Helmut Paul [c,*]

[a] Departamento de Física – Centro de Investigación en Óptica y Nanofísica, Universidad de Murcia, E-30100 Murcia, Spain
[b] Departament de Física, Universitat d'Alacant, E-03080 Alacant, Spain
[c] Atomic and Surface Science, University of Linz, Altenbergerstrasse 69, A 4040 Linz, Austria

ARTICLE INFO

Article history:
Received 16 November 2012
Received in revised form 2 January 2013
Available online 8 February 2013

Keywords:
Stopping power
Stopping force
Energy loss of particles
Liquid water
Protons

ABSTRACT

Two experiments about the stopping power of liquid water for proton beams have recently been reported. The one by the Jyväskylä group (4.8–15.2 MeV) agrees nicely with the Bethe theory and other recent theoretical calculations, whereas the other one by the Kyoto group (0.3–2 MeV) appears to be about 10% low. In this comment we show that the Kyoto energy spectra can be interpreted differently, so that the deduced stopping power also agrees with the Bethe stopping power.

For medical physics, liquid water is one of the most important substances. There are two recent experimental stopping power results of liquid water for swift protons: measurements by Shimizu et al. [1,2] at Kyoto (Japan), and by Siiskonen et al. [3] at Jyväskylä (Finland). In the Kyoto method, the stopping power was not measured directly, but proton energy spectra after traversing a liquid water jet target (diameter $D = 50$ μm) were measured at various scattering angles, and the energy distributions were compared to distributions calculated by means of Monte Carlo simulations using the GEANT4 toolkit [4]; the diameter D of the liquid water target and a correction factor α multiplying SRIM stopping [5] were used as fitting parameters. At Jyväskylä, transmission measurements were employed using a thin liquid water target (enclosed within two thin copper sheets). Absolute values of both sets of experimental results, compared to various theories, can be seen in the collection of stopping data graphs by one of us [6]. Fig. 1 shows the results relative to the proton stopping table of ICRU Report 49 [7], so as to make small differences more visible. For the curves in Fig. 1, the value of the mean ionization potential I is given where known.

According to the relativistic Bethe theory (without corrections) [8,7], the mass stopping power for ions is given by

$$\frac{S}{\rho} = (0.307075\ \text{MeV cm}^2\ g^{-1})\frac{Z_1^2}{\beta^2}\frac{Z_2}{A_2}L(\beta), \tag{1}$$

where S is the linear stopping power; Z_2, A_2 and ρ are the atomic number, mass number and density of the target; Z_1 and v are the atomic number and velocity of the projectile; $\beta = v/c$ where c is the speed of light; and the stopping number L is given by

$$L(\beta) = \ln\frac{2mv^2}{(1-\beta^2)} - \ln I - \beta^2, \tag{2}$$

where m is the mass of the electron and I is the mean ionization potential of the material. Eq. (2) is generally reliable [9] at energies high enough (but not so high that the density correction [7] becomes appreciable). To extend the validity to lower energy, one customarily adds shell, Barkas–Andersen and Bloch corrections [7] to Eq. (2), thus obtaining the "corrected Bethe equation". The choice of the proper I value for liquid water is an ongoing problem, with I between 78 and 79 eV being probably best (see, e. g., Ref. [10]).

The characteristics of the other curves shown in Fig. 1 are briefly described in what follows. The program PASS is based on the binary theory of electronic stopping [11]. The semi empirical SRIM code [5] is based on the parameterization and interpolation of a large collection of available experimental data. Calculations based on the dielectric formalism using optical data models for the energy loss function of liquid water based on the most recent measurements [12] are represented by Emf06 [13] and GarM09 [14], whose main differences lie in the procedure to extend the optical data to the whole momentum and energy excitation spectrum of the target.

On the average, above 10,000 keV the corrected Bethe curve in Fig. 1 (with $I = 78$ eV) is 0.52% below unity (the ICRU 49 curve with $I = 75$ eV), as one expects comparing the stopping numbers $L(\beta)$, Eq.

* Corresponding author. Tel.: +43 732 2468 8514.
E-mail address: helmut.paul@jku.at (H. Paul).

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http://dx.doi.org/10.1016/j.nimb.2013.01.038

(2), for the two different I-values; the uncorrected Bethe equation (Eqs. (1), (2)) is also shown in Fig. 1 to indicate the size of the corrections to that equation.

Inspection of Fig. 1 shows a problem [10]: the Bethe equation with corrections is generally reliable and depends essentially only on I and on the shell correction [7] in the region above 2000 keV, and the corrections are quite small here. The GarM09 [14] curve (above 800 keV) and the PASS [11,15] curve (above 1500 keV) are quite close to the corrected Bethe curve, and therefore they all seem quite reliable in this energy range. The Emf06 [13] and the SRIM [5] curves both oscillate about the ICRU values in the range of interest of this work. The resemblance of the SRIM curve at low energies to the experimental data reported by the Kyoto group is because these authors use SRIM data as input for their fitting procedure, as will be discussed later on.

Hence it appears that the Kyoto measurements may be too low by about 10%. This is surprising since Shimizu et al. checked the accuracy [16] of the method by using He ions on their water jet target, and by measuring proton energy loss and scattering from an Al wire; in both cases, the results agreed with the data from ICRU Report 49 [7]. In addition, the recent measurements of the Kyoto group for He ions on liquid ethanol [17], by the same method, agree very well [6] with SRIM [5] and with BEST [18].

Recently, new light has been shed on this discrepancy by using the SEICS code (Simulation of Energetic Ions and Clusters through Solids) [19], which has been employed to simulate a 2 MeV proton beam scattered off a cylindrical water jet at a detection angle of 10 mrad. The obtained results are compared in Fig. 2 with the corresponding energy distribution measured by Shimizu et al. [2]. Our calculated distribution agrees very nicely with the measurements (evidently better than the GEANT4 simulation); to get this agreement we used the unchanged GarM09 stopping power [14] and only had to adjust the thickness of the water jet to $D = 48.25$ μm. This should be compared to the Kyoto group simulation [2], represented by a dashed curve in Fig. 2, which was obtained using the GEANT4 code by varying two quantities: the diameter D of the liquid water jet and the stopping power. The former was taken to be $D = 51$ μm, whereas the latter was 0.89 times the SRIM2008 stopping power for water in the vapor phase. It is worth to remark that the result from the SEICS code only required a reduction of D by 3.5% from the nominal value $D = 50$ μm, which does not seem unreasonable due to possible evaporation or narrowing of the liquid jet downstream. Such a good agreement was also obtained using the same value of D for simulating the proton energy distribution at 30 and 50 mrad [20].

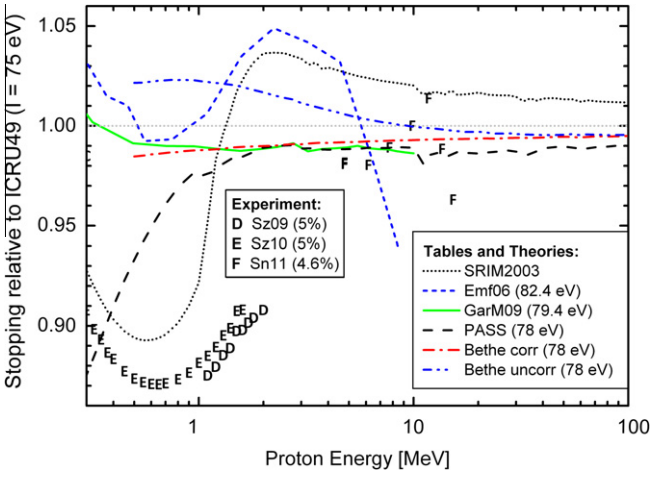

Fig. 1. Electronic stopping power of liquid water for protons above 300 keV, normalized by the data from the proton table of ICRU Report 49 [7]. The designations Sz09 and Sz10 refer to the liquid jet measurements by Shimizu et al. [1,2]; Sn11 refers to the transmission measurement by Siiskonen et al. [3]; experimental uncertainties are given in parentheses. The references for the curves are as follows: SRIM [5], Emf06 [13], GarM09 [14], and PASS [11,15]. "Bethe corr" is the Bethe theory with shell, Barkas and Bloch corrections, calculated using BEST [18]. To indicate the size of the corrections, the Bethe theory without any corrections ("Bethe uncorr") is depicted, also calculated by means of BEST. I values are shown in parentheses, where known.

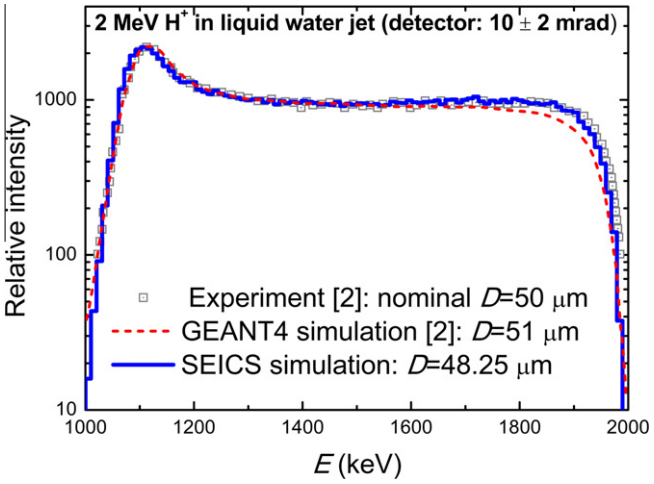

Fig. 2. Energy distribution at 10 mrad of 2 MeV protons interacting with a liquid water jet exiting a nozzle with a (nominal) $D = 50$ μm diameter. Symbols correspond to experimental measurements [2]; the dashed line is the distribution obtained [2] with GEANT4 by fitting the nozzle diameter to $D = 51$ μm and the stopping power of liquid water to 0.89 times the value provided by SRIM2008 for water vapor. The solid line represents the results of the SEICS code [19] after fitting only the value $D = 48.25$ μm for the diameter of the water jet, without adjustment of the stopping power of liquid water, which is provided by the MELF-GOS method [14].

From the above discussion we conclude that the experimental stopping power of liquid water in the range of 0.3–2 MeV deduced by the Kyoto group is too small by about 10% since it disagrees with several theories and since the measured energy distributions can also be explained on the basis of the GarM09 stopping power which agrees closely with the Bethe stopping power. Clearly, this statement does not invalidate the measured energy spectra [2].

Acknowledgements

We acknowledge financial support from the Spanish Ministerio de Economia y Competitivad and from the European Regional Deveolpment Fund (Project FIS2010-17225). PdV thanks the Conselleria d'Educació de la Generalitat Valenciana for a grant within the VALi+d program. Support from the COST Action MP1002 NanoIBCT is gratefully acknowledged. HP is grateful to P. Sigmund for calculating the PASS curve and to Dr. Shimizu for many instructive email discussions.

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