A critical overview of recent stopping power programs for positive ions in solid elements

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A critical overview of recent stopping power programs for positive ions in solid elements.

Helmut Paul[1,]* and Daniel Sánchez-Parcerisa[2]

Abstract

Several tables exist in the literature containing stopping power data for different targets and projectile ions. Moreover, Monte Carlo transport codes often incorporate their own implementation of electronic stopping power. In this article, we compare recent stopping power publications to our large collection of experimental stopping data for positive ions, in order to test their validity. We consider stopping codes by Barbui et al., Javanainen, and Diwan et al., and also the stopping power descriptions built into Monte Carlo transport codes: "Bethe" in libdEdx / SHIELD-HIT, Geant4-DNA, FLUKA, and MNCPX. We use graphical comparisons for significant single ion/target combinations, and we use our statistical program "Judge" to analyse larger data sets for those ions and targets where many experimental results are available. Only solid elemental targets are considered.
The performance of the codes is evaluated in terms of how well they describe the available experimental data, in comparison with previously analyzed programs. Although interesting new approaches have been used in some of the codes, both SRIM and MSTAR still seem to be slightly better at reproducing the available data.

Keywords: stopping power, stopping force, energy loss of particles

1. Introduction

In spite of much work over many years, an accurate theoretical description of the electronic stopping power of matter for positive ions, over the whole energy range of practical interest, is still missing. A number of semi-empirical approaches have appeared and are still appearing, but their reliability is not always evident. In this paper, we attempt to test the validity of recent approaches by means of statistical comparisons to our large collection [1] of experimental data.
We first give a brief overview of stopping tables and programs that have been publicly[3] available since 1982 and that are useful for an extended range of targets and energies (see Table 1, based on ref. [2]). We have omitted the program by Konac et al. [3] since it applies only to two target elements, the program by Weijers et al. [4] since it is only for a restricted energy range, and the program LET [5,6] since it based on outdated (pre-1999) versions of TRIM/SRIM [7].
At high ion energies, an important and accurate reference is provided by the relativistic Bethe theory [8,9], according to which the mass stopping power is given by

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

where $S = -dE/dx$ is the linear stopping power (or stopping force), x is the distance, ρ is the density of the material; Z_1, E and v are atomic number, energy and velocity of the ion; Z_2 and A_2 are atomic number and mass number of the target; $\beta = v/c$, c is the speed of light; and the

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[3] Except for BEST which the author has obtained from M.J. Berger

stopping number L is given by

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

where m is the mass of the electron and I is the mean ionization potential. Eq. (2) is reliable at energies high enough (but not so high that the density correction becomes appreciable [9]). To extend the validity to lower energy, one customarily adds shell, Barkas-Andersen and Bloch corrections [9] to eq. (2).

The reliability of some of these programs has been investigated in earlier papers ([2,10,11, 12], see also "Statistical Analysis" in ref. [1]). Briefly, the results were as follows: for H and He ions, both ICRU Report 49 and SRIM describe the data best. For ions from $_3$Li to $_{18}$Ar, MSTAR and SRIM describe the data equally well; ICRU 73 is often too high, especially at low energy, and particularly for gases. For ions from $_{19}$K to $_{92}$U, the non-perturbational Lindhard-Sørensen theory must be used for accurate results above 100 MeV/u; between 30 and 100 MeV/u, SRIM and Hubert are slightly better. Below 30 MeV/u, SRIM is 6 % high in heavy targets and 5 % low in light targets, on the average. In that range, Hubert is better (but Hubert is valid only above 2.5 MeV/u). For gases near the maximum, SRIM is too high. But SRIM describes all ions at all energies in all targets, and is best in most cases.

In this article, we investigate the reliability of recent stopping programs and of the stopping power built into Monte Carlo transport codes, in terms of how well they describe the available experimental data.

2. Statistical analysis

The comparison between experimental and calculated stopping powers is done by both graphical comparisons (for some relevant cases) and by statistical analysis where enough experimental data are available. For the latter, we use our program "Judge" [13]. This program calculates the normalized differences

$$\delta = (S_{\text{exp}} - S_{\text{tab}})/S_{\text{exp}} \qquad (3)$$

for every data point. Here, S_{exp} is the experimental value from [1], and S_{tab} the corresponding table value for the same ion, same target and same energy. Then, in every range of specific energy (i.e., energy per nucleon), it determines the average normalized difference

$$\Delta = \langle\delta\rangle \qquad (4)$$

and its standard deviation

$$\sigma = \sqrt{\langle\delta^2\rangle - \langle\delta\rangle^2}\,. \qquad (5)$$

The averages designated by $\langle\ \rangle$ are unweighted, except that we reject experimental data that appear to be in conflict with the majority (see ref. [1]). The calculated average normalized difference (and its standard deviation) is then a reliable indicator of how well the programs reproduce the experimental data.

3. Analysis of recent programs

3.1 Barbui et al.

M. Barbui et al. [14] have recently presented a stopping power parameterization for heavy ions from 0.1 to 15 MeV/nucleon, using experimental heavy ion stopping data and the useful concept of effective charge with 6 parameters, separately for solids and for gases. This

concept has been introduced by Northcliffe [15] and critically discussed by Sigmund [10]. The basis of this concept is the empirical scaling law

$$\frac{S}{(\gamma Z_1)^2} = \frac{S_{ref}}{Z_{ref}^2} \qquad (6)$$

where Z_1 is the atomic number of the heavy ion, γZ_1 is its effective charge, and S_{ref} is the stopping power of the same medium for a reference ion of the same velocity v and of effective charge Z_{ref}.
Barbui used proton data from SRIM [7] for reference, after a slight renormalization.
The program is evidently meant for ion atomic numbers $Z_1 \geq 18$ (including extrapolation beyond $Z_1 = 92$), and for target atomic numbers $Z_2 \geq 6$, it is for elements and for compounds.
As an example, Table 2 shows a statistical analysis of both the Barbui parameterization and the SRIM program [7], using the stopping data collection from ref. [1] for ions from $_{13}$Al to $_{92}$U in Ni and Cu targets and our program "Judge" [13]. The accuracy of the two fits in Table 2 is comparable except at low energy, where the Barbui fit is too low (Δ too large). As an example, Fig. 1 shows a graph for uranium ions on aluminum where this discrepancy is clearly seen: unrealistically, the Barbui curve approaches zero at finite energy. But it is nice to see that the curve follows the unpublished Geissel (Gs98) data very well, even though Barbui et al. did not have these data in their data base [16]. In Fig. 2 for Ar on Al also, the Barbui curve is good at high energy, but becomes too low at 0.1 MeV/nucleon. A similar Barbui curve is also seen in Fig. 3 below. One can see this tendency also in Barbui's Fig. 2 [14], but our logarithmic abscissa shows it more clearly.

3.2 Javanainen

A. Javanainen has recently proposed a simple expression for the stopping power of heavy ions in solids [17], using an adaptation of Bohr's classical theory [10,18]. Without using the concept of effective charge, data for 20 different ion-target combinations in the range $2 \leq Z_1 \leq 54$, $6 \leq Z_2 \leq 79$, taken from the compilation [1], are fitted directly, using only three parameters. Only data below the stopping maximum ($v < 2Z_1 v_0$, where v_0 is the Bohr velocity) are used for the fit, and the simple Bloch [19] relation $I = Z_2 \cdot (10\ \text{eV})$ is employed for the mean ionization energy I.
Examples are shown in Figs. 2 - 6. In spite of the small number of parameters, the fits are surprisingly good. This can also be seen from Table 3 which shows a statistical analysis for ions C, N, and O in all solid elements for which we have data. The value of σ is smaller than for Barbui, even though the energy goes down to much smaller values. For comparison, Table 3 also analyzes the fits of three other tables: SRIM and MSTAR are clearly better than Jav12. In general, the accuracy increases with increasing energy, since measurements are more accurate at higher energies. Only in the case of Jav12, it decreases, since points beyond the maximum were not used for the fits.
Fig. 4 shows the case for Au ions in gold: the Jav12 curve is quite good even though these data were not used in fitting. In general, statistical analysis for heavy ions shows much larger discrepancies for Jav12 than Table 3, but this may be partly due to unreliable measurements.

3.3 Diwan et al.

Diwan et al. [20] have produced a fit for heavy ions in solids which is an extension of the Hubert table [21] to lower energies. Both tables are based on the useful concept of "effective charge"

Using helium as a reference ion, the parameter γ is empirically determined by Hubert as a function of Z_1, Z_2 and v. The Hubert formulation is valid for 2.5 MeV ≤ E/A_1 ≤ 500 MeV. In this range of specific energy, the He reference ions are fully stripped, i.e., $Z_{ref} = 2$.

Diwan et al., again using He ions for reference, have extended the Hubert fit to the range 0.5 MeV ≤ E/A_1 ≤ 2.5 MeV, for 3 ≤ Z_1 ≤ 35 and for solid targets within 6 ≤ Z_2 ≤ 79 by revising the Hubert fitting parameters; they did not need a dependence upon Z_2, but they had $Z_{ref} = Z^*_{He} < 2$. They took S_{ref} and Z^*_{He} from SRIM-98 [22] for their fit [23]. Fig. 5 shows an example for Cu ions in aluminum, where the Diwan fit nicely extends the Hubert table to lower energies. But since the data from SRIM-98 are hard to obtain nowadays, it is difficult to use the Diwan fit.

4. Analysis of stopping power in some Monte Carlo transport codes

4.1 "Bethe" in libdEdx / BETHE_EXT00 in SHIELD-HIT

An internet library [24] which provides electronic stopping power data for a wide range of ion/material combinations has recently been created by the Aarhus Particle Therapy Group [25]. A convenient online calculator is available [26] that reproduces several published stopping tables, and also a function called "Bethe". The latter is an implementation of the Bethe-equation (eqs. 1, 2) expanded to low energy; the algorithm agrees closely [27] to the algorithm BETHE_EXT00 used in the development version 12A of the Monte Carlo particle transport code SHIELD-HIT [28], the Heavy-Ion-Therapy version of the hadron transport code SHIELD [29].
The aim of the library is to provide easy access by exact reproduction of the published tables, without comparison with experimental results.
Here, we are only interested in investigating the "Bethe" program in its version of November, 2012. Examples can be seen in Fig. 6 for C ions in carbon and in Fig. 7 for N ions in silver where the "Bethe" curve appears too low left of the maximum. The double logarithmic plot shows that in this (as in all) cases, the libdEdx curve is proportional to velocity left of the maximum. Various degrees of agreement with the data can be seen in Figs. 1 – 5 and 10. Table 4 shows a statistical comparison between "Bethe" and experimental data, for Li, C, N and O ions[4]. Only in the highest energy range, the agreement is good; otherwise, "Bethe" is too low. For H and He ions, statistical comparisons show even greater discrepancies. An example can be seen in Fig. 8 for H ions in aluminum, where the disagreement is striking.

4.2 Geant4 and Geant4-DNA package

Geant4 (acronym for GEometry ANd Tracking) [30] is a platform for "the simulation of the passage of particles through matter," using Monte Carlo methods, applicable in many parts of physics. The general Geant4 platform has several options for published stopping powers, and the possibility of scaling proton stopping powers for use with heavier ions. Recent developments have come from the fields of radiation biophysics and hadron therapy. For instance, Lechner et al. [31] have shown that, by using the revised ICRU73 stopping table [32], Geant4 accurately predicts absolute Bragg peak positions in water at carbon beam energies of therapeutic interest. Moreover, a package called Geant4-DNA [33] has been developed for the modeling of early biological damages induced by ionizing radiation.

[4] For these ions in C, Al, Ni, Ag, and Au targets we have the greatest number of data

Recently, the latter package has been used to calculate the stopping power for electrons, protons and alpha particles in liquid water [34]. Fig. 9 shows the stopping power of water for protons, normalized by the data from the proton table of ICRU Report 49 [9] in order to make small differences visible. It shows recent experimental data due to M. Shimizu et al. [35,36] and Siiskonen et al. [37]. In addition, it shows the relativistic Bethe theory with shell, Barkas and Bloch corrections [9] (here calculated using BEST [38]) and the theories PASS [39,40], Garcia-Molina et al. [41], and Emfietzoglou et al. [42]. We have also added a Geant4-DNA curve corresponding to the high energy part of Fig. 3 from Francis[5] et al. [34].

Since the correction to the pure Bethe theory eq. (1) is less than 1 % at 10 MeV, the corrected Bethe theory can be expected to be quite accurate here. Indeed, the GarM09 curve and the PASS curve are very close to BEST, about 1 % below ICRU 49 (i.e., unity), and therefore, they all seem reliable[6]. In contrast, it appears that the Geant4-DNA curve is about 6 – 8 % too high in this energy region. It matches ICRU 49 at lower energies, but evidently, it has not been made to match Bethe stopping at high energy.

4.3 FLUKA

The modern FLUKA [43,44] is a general purpose Monte Carlo particle transport and interaction code originally developed for shielding and high energy physics applications. It is currently widely used for basic research, as well as in applied physics, including dosimetry, medical physics, radiation protection and space radiation.

The calculation of stopping powers for high energy is based on the Bethe formula (eqs. 1, 2) with shell, Barkas and Bloch corrections [45,46]. The average ionization potentials (I-values) and density effect parameters of pure elements are taken from the compilation of Sternheimer, Seltzer and Berger [47]. For the I values of compounds, data from ICRU 49 [9] are used if available; if not, Bragg's additivity rule is taken with I values from the constituting elements. In any case, the user can explicitly override the default I-values. In order to account for partially dressed ions, the effective charge correction by Hubert et al. [48] is implemented [49]. Shell corrections are derived from a parameterization by Andersen and Ziegler [50], fitting available experimental data for protons.

In the context of this work, FLUKA stopping power tables for several ions and elements were extracted and compared with the available experimental data. However, according to the FLUKA user license [45], the *"publication of any results of comparisons of specific internal physics models extracted from FLUKA (...) with data or with other codes or models is subject to prior written permission"*. Since, unfortunately, this permission was neither given nor denied within six months (which appears to be a sufficiently long time), we decided not to show the various comparisons obtained, results which were neither particularly favorable nor unfavorable to FLUKA.

4.4 MNCPX

MNCPX (Monte Carlo N-Particle eXtended) [51] is a Monte Carlo radiation transport computer code that transports nearly all particles at nearly all energies for nearly all applications. MNCPX stopping curves (calculated with version 2.7.0) are shown in Figs. 5 – 7 and in Fig. 10; the data are analyzed statistically in Table 5. At the view of these comparisons, MNCPX stopping power is generally 5 – 6 % too low, except at high energy, especially for heavy targets.

5. Conclusions

[5] This was calculated using numerical data kindly provided by Dr. Francis.

[6] BEST, GarM09 and PASS lie below unity because of their larger I values compared to that of ICRU 49.

We have compared various recent fits to experimental stopping data both graphically and by means of statistical analyses. In abbreviated form, these fits compare as follows:

Barbui et al. [14]	Good at high energy, but too low at low energy
Javanainen [17]	Surprisingly good agreement in view of the small number of parameters, but not as good as SRIM or MSTAR
Diwan et al. [20]	Extension of the Hubert table to lower energies, but hard to use in practice
libdEdx "Bethe" [26] (SHIELD-HIT BETHE_EXT00)	Generally too low at low energy
GEANT4 DNA package [34]	6 – 8 % too high in the range 500 – 5000 keV for protons in liquid water; agrees with ICRU49 at lower energy
FLUKA [43]	See section 4.3 above
MNCPX [51]	5 – 6 % too low in general, except at high energy.

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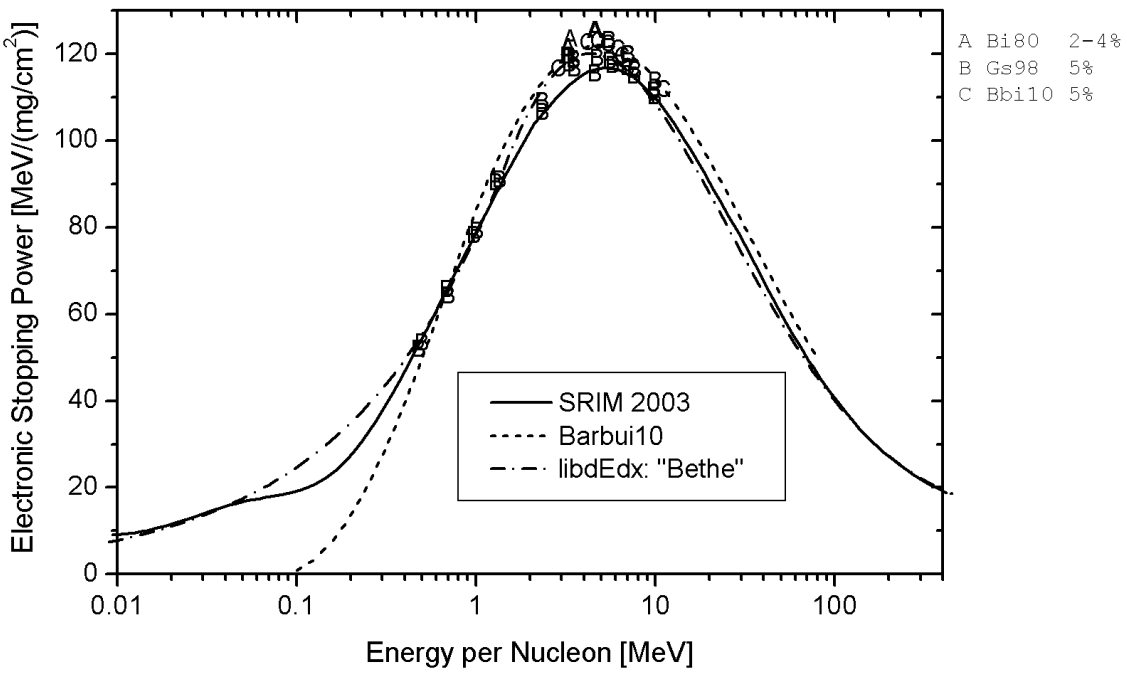

Fig. 1. The electronic stopping power of aluminum for U ions. Experimental data designated by letters are taken from the compilation [1]. The curves correspond to SRIM [7], Barbui [14], and "Bethe" [26].

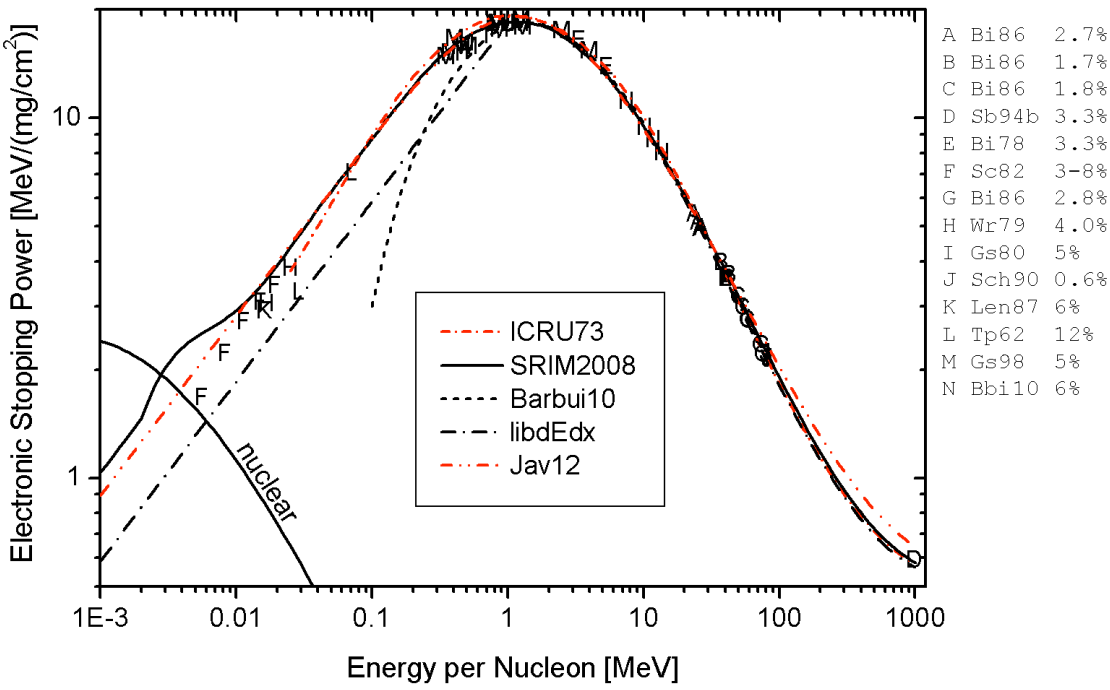

Fig. 2. Like Fig. 1, for Ar ions in aluminum. Additional curve designations are as follows: ICRU73 [10] and Jav12 [17]. The nuclear stopping power, calculated using SRIM, is also shown.

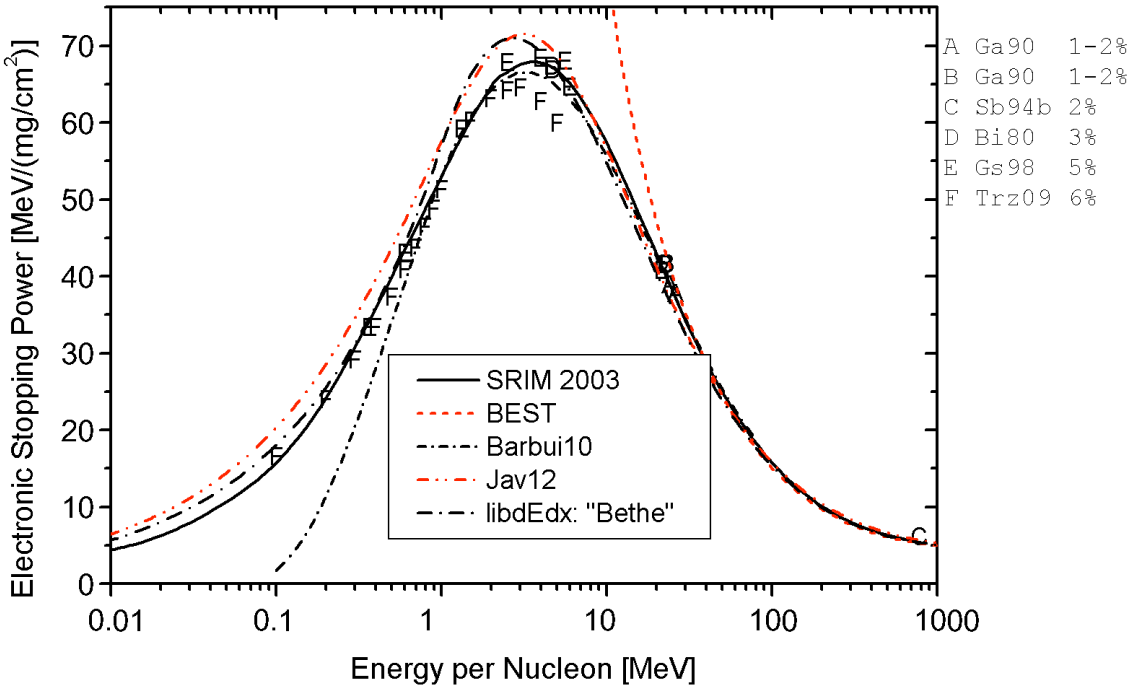

Fig. 3. Like Fig.2, for Xe ions in aluminum. The BEST (Bethe) program [38] assumes unclad nuclei.

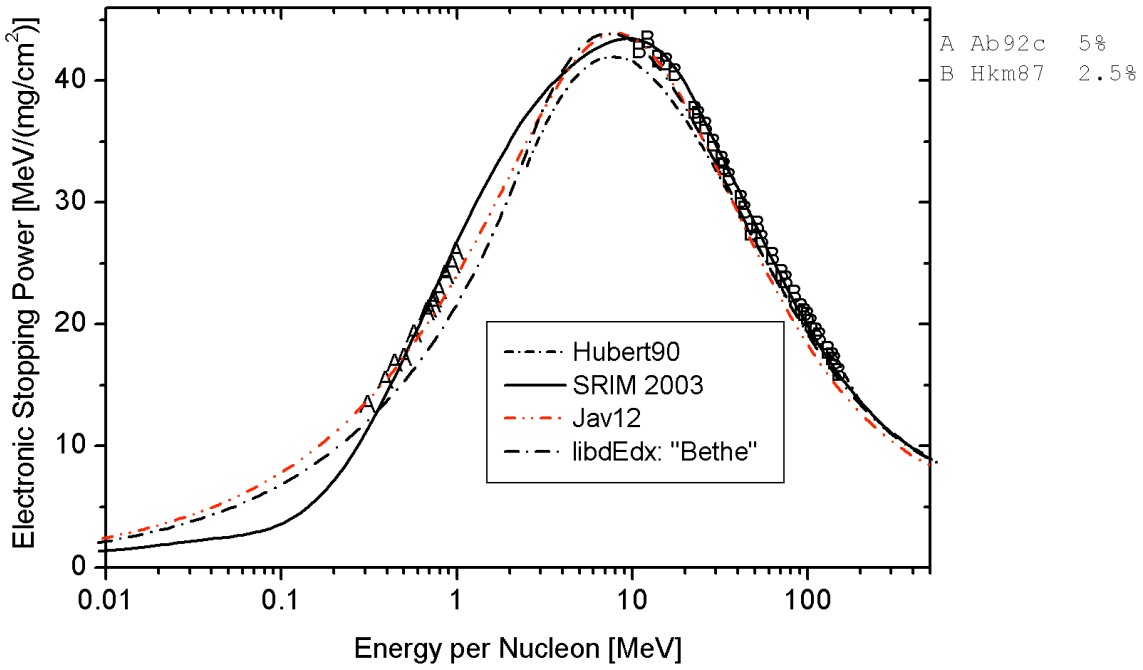

Fig. 4. Like Fig. 3, for Au ions in gold. The Hubert [48] curve is also shown.

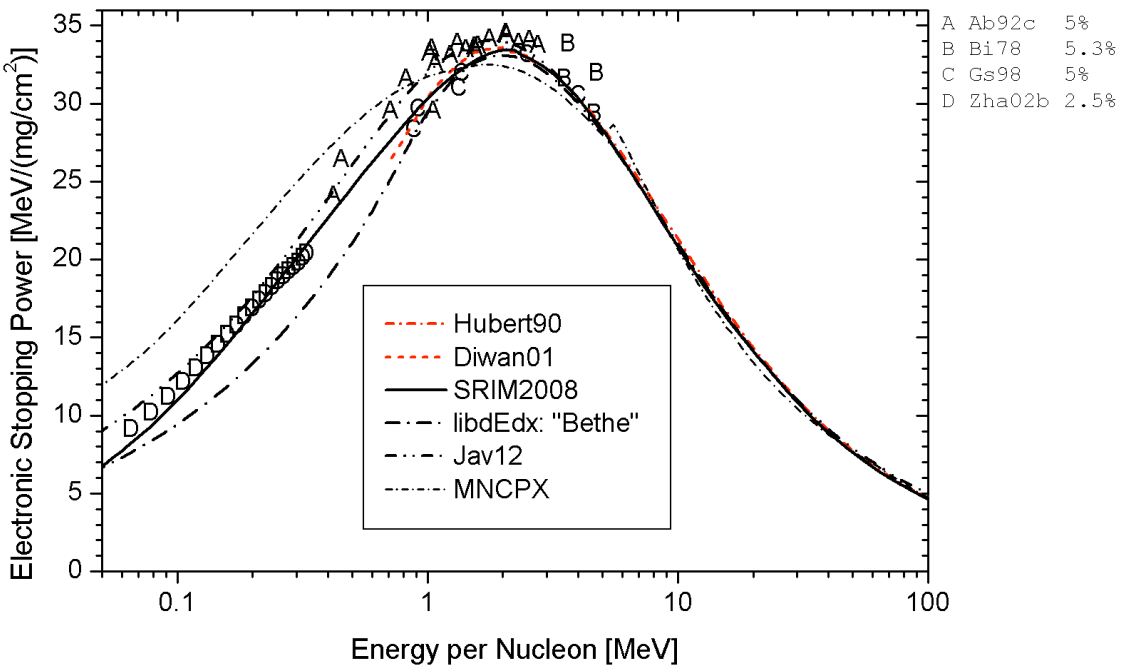

Fig. 5. Like Fig. 4, for Cu ions in aluminum. The Diwan fit [20] extends the Hubert table [48] to lower energies. A MNCPX curve [51] is also shown.

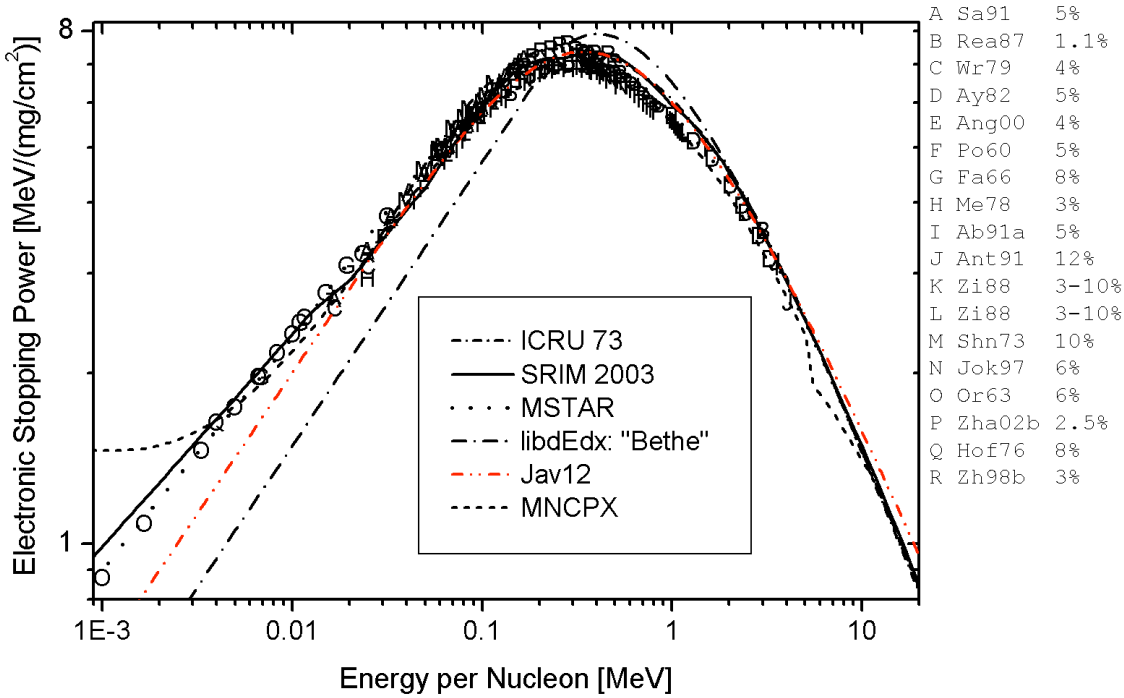

Fig. 6. Like Fig. 1, for C ions in carbon. Curve designations are as follows: ICRU 73 [10], SRIM [7], MSTAR [56], libdEdx [26], Jav12 [17], MNCPX [51]

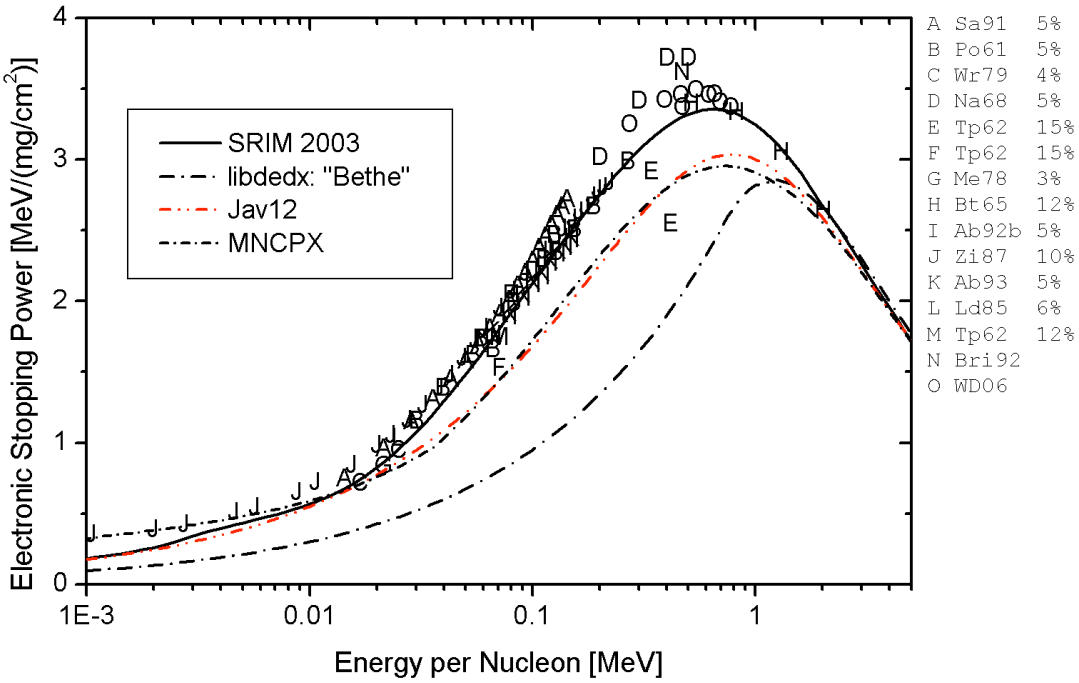

Fig. 7. Like Fig. 6, for ^{14}N and ^{15}N ions in silver.

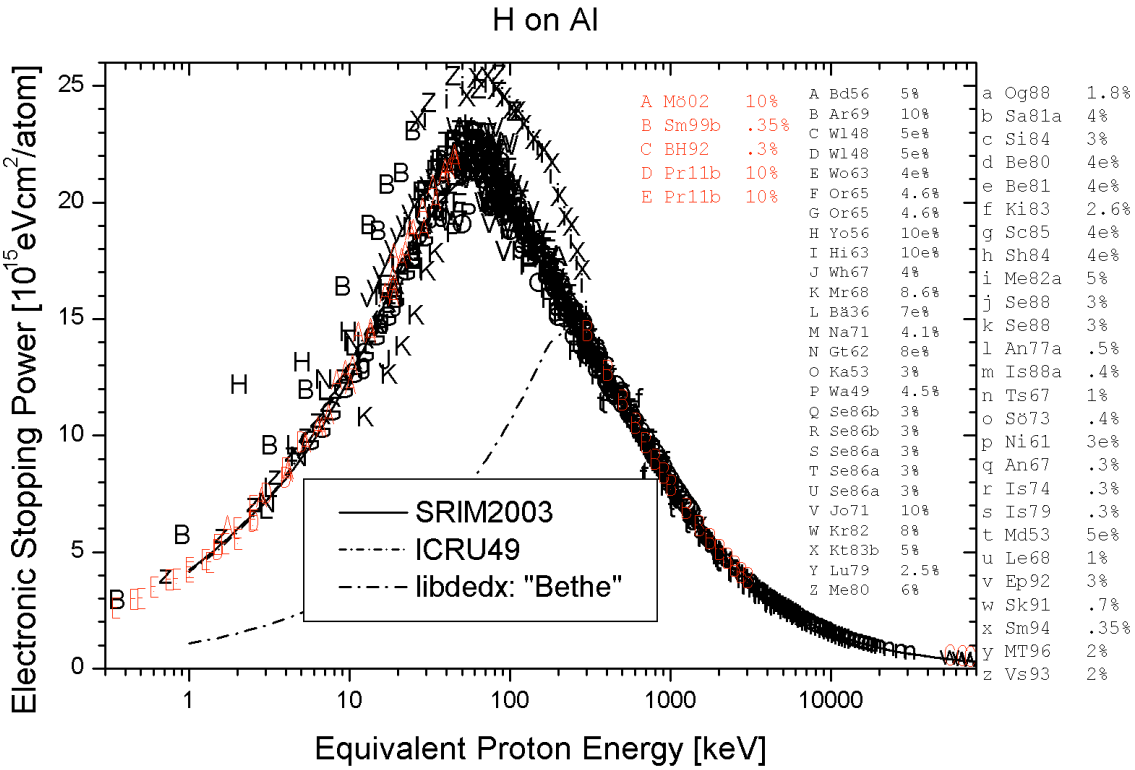

Fig. 8. Like Fig. 6, for H ions in aluminum.

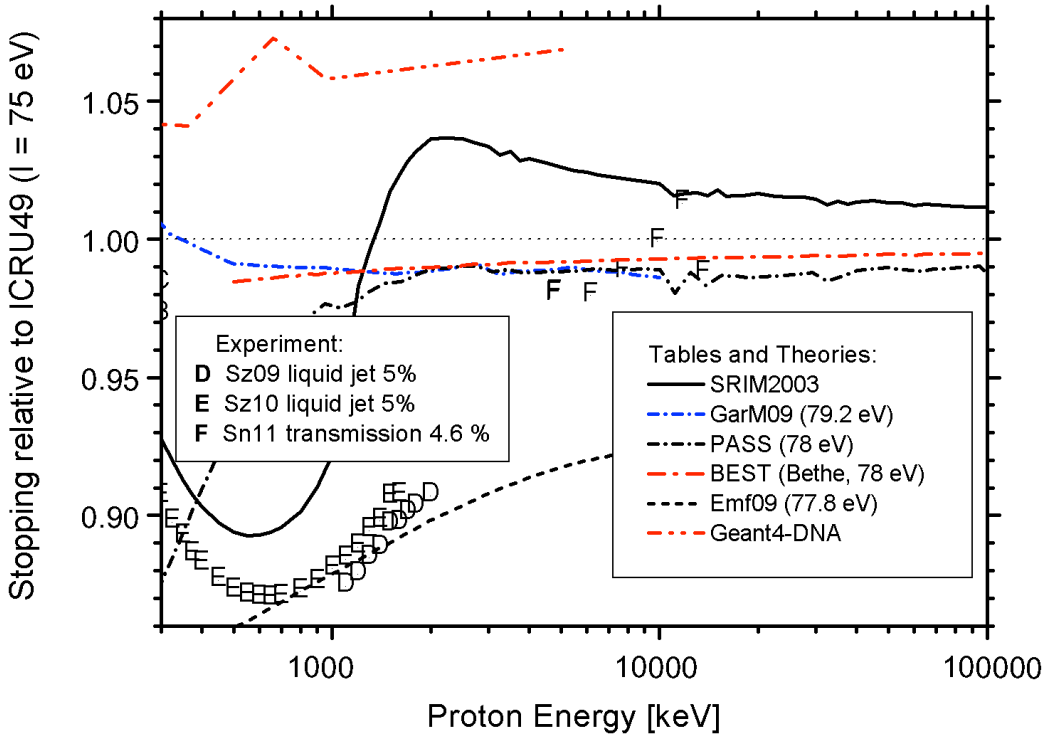

Fig. 9. Electronic stopping power of liquid water for protons, normalized by the data from the proton table of ICRU Report 49 [9]. The designations for curves and for experimental points are as follows: Sz09 [35], Sz10 [36], Sn11 [37], SRIM [7], GarM09 [41], PASS [40], Emf09 [42], Geant4-DNA [34]. *I* values are shown in parentheses where known.

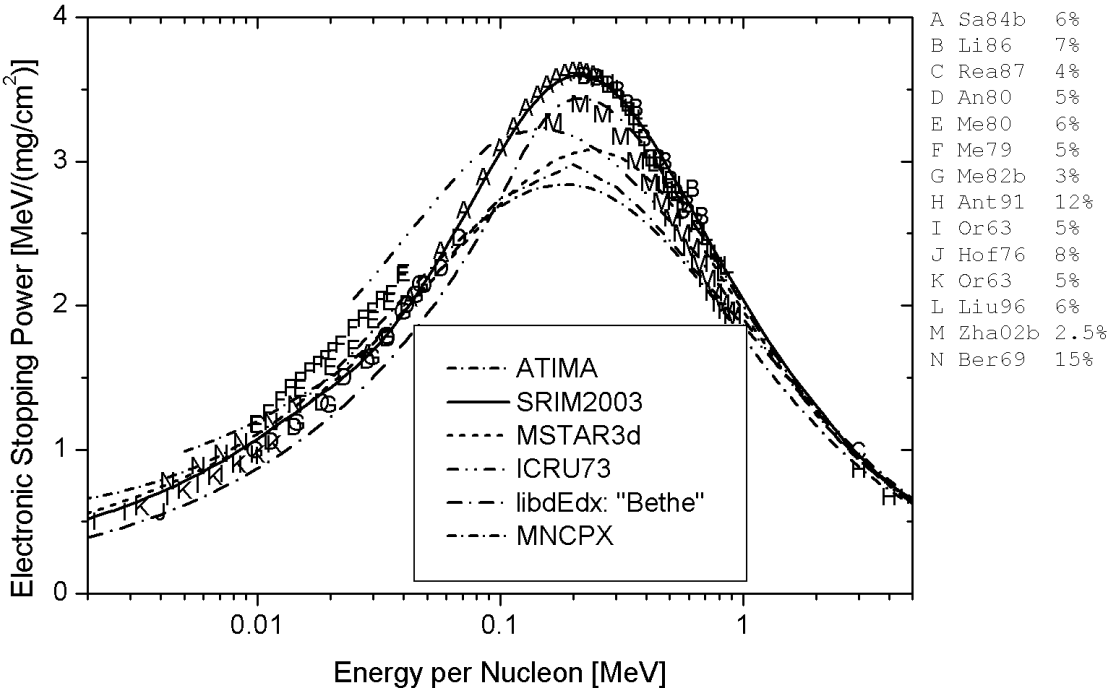

Fig. 10. Like Fig. 1, for Li ions in carbon. Curve designations are: ATIMA [52], SRIM [7], MSTAR [56], ICRU73 [10], libdEdx [26], MNCPX [51].

Table 1. Tables and computer programs for the stopping power of matter for positive ions that have been available in the literature for some time.

Name, reference	Z_1	Z_2	(Specific) energy range	Remarks
ATIMA [52]	1 - 92	1 – 92	≥10 MeV/u (as used here)	Based[7] on Lindhard-Sørensen [53] above 30 MeV/u
BEST [38]	1 - 92	1 – 92; 180 compounds[8]	≥ 0.5 MeV/u	Bethe theory with corrections. Used for high energy parts of ICRU Rep. 49
CasP v. 5.0 [54]	1 - 92	1 – 92, any compound[9]	0.0001 – 200 MeV/u	Target and projectile ionization must be calculated separately
Hubert et al. [21]	2 - 103	36 solid elements	2.5 – 500 MeV/u	
ICRU Report 49 [9]	1, 2	25 elements, 48 compounds or mixtures	0.001-10000 MeV (p); 0.001-1000 MeV (α)	Programs NIST PSTAR, NIST ASTAR
ICRU Report 73 [10, 32]	3 – 18, 26	25 elements, 31 compounds	0.025 – 1000 MeV/u	Based on PASS. For water, the revised table [54] must be used.
Janni [55]	1	1 – 92; 63 compounds	0.001 – 10000 MeV	Outdated list of mean ionization energies
MSTAR[56]	3 - 18	31 elements, 48 compounds or mixtures	0.00025 – 250 MeV/u	Based on alpha stopping powers of ASTAR
PASS [39]	many	many	Above 0.025 MeV/u	Binary Theory, Used for ICRU Rep. 73
SRIM[10] 2003 – 2012 [7].	1 – 92	1 – 92, many compounds	1.1 eV – 10 GeV/u	SRIM stopping was not changed since 2003
Ziegler et al. 1985 [57]	1 - 92	1 – 92; many other targets	0.1 – 100000 keV/u	First program to treat all ions, all targets

Table 2. Mean normalized deviations $\Delta \pm \sigma$ (in %) between experimental data and fits by Barbui [14] and SRIM [7] for ions with $Z_1 > 12$ in Ni and Cu

E/A_1 (MeV)	0.1 – 1	1 - 10	10 - 15	0.1 -15
No. of points	*478*	*481*	*7*	*966*
Barbui10	16 ± 20	-0.6 ± 5.4	-4.6 ± 1.3	7.4 ± 17
SRIM 2008	-2.4 ± 7.7	-1.2 ± 5.0	-6.3 ± 1.1	-1.9 ± 6.5

[7] Below 10 MeV/u, the values are based on an old version of SRIM (Ziegler et al., 1985). Between 10 and 30 MeV/u, the values are interpolated between SRIM and Lindhard-Sørensen.

[8] Additional compounds may be calculated by entering a chemical formula, assuming Bragg's additivity.

[9] Compounds are calculated according to chemical formula, assuming Bragg's additivity.

[10] SRIM was called TRIM in earlier times

Table 3. Mean normalized deviations $\Delta \pm \sigma$ (in %) for C, N, and O ions in all solid elements for which we have data, compared to various tables[11].

E/A_1 (MeV)	0.001 – 0.1	0.1 – 10	10 - 1000	0.001 - 1000
No. of points	*1006*	*2359*	*93*	*3458*
Jav12 [17]	7.9 ± 14.4	5.9 ± 9.8	-9.6 ± 6.6	6.1 ± 11.6
SRIM 2008 [7]	1.8 ± 10.3	-0.0 ± 5.6	0.0 ± 2.2	0.5 ± 7.3
No. of points	*953*	*2166*	*93*	*3212*
MSTAR [56]	-1.2 ± 10.5	0.6 ± 5.5	0.3 ± 1.6	0.1 ± 7.3
No. of points	*720*	*2059*	*85*	*2864*
ICRU73 [10]	-9.7 ± 13	-7.1 ± 9.2	0.1 ± 2.4	-7.5 ± 10.3
No. of points		*199*	*89*	*288*
Hubert [**Error! Bookmark not defined.**]		0.6 ± 5.9	0.6 ± 1.6	0.6 ± 5.0

Table 4. Mean normalized deviations $\Delta \pm \sigma$ (in %) for Li, C, N and O ions in C, Al, Ni, Ag, and Au, compared to the "Bethe" code of libdEdx [26], version 1.2.1

E/A_1 (MeV)	0.001 – 0.1	0.1 – 10	10 - 1000	0.001 - 1000
No. of points	*896*	*1934*	*47*	*2877*
"Bethe"	42 ± 15	21 ± 24	0.6 ± 1.7	27 ± 24

Table 5: Mean normalized deviations $\Delta \pm \sigma$ for Li, C, N, and O ions in many[12] solid targets, compared to various tables.

E/A_1 (MeV)	0.0123 – 0.1	0.1 - 10	10 - 1000	0.0123 - 1000
No of points	*854*	*2240*	*58*	*3152*
MNCPX	5.4 ± 11.8	6.2 ± 7.7	1.3 ± 1.9	5.8 ± 8.9
No. of points	*1406*	*3570*	*84*	*5068*
SRIM	2.2 ± 7.6	-0.5 ± 5.6	0.1 ± 2.3	0.3 ± 6.3
No. of points	*1142*	*3211*	*84*	*4437*
MSTAR	-0.4 ± 8.6	0.7 ± 5.5	0.4 ± 1.8	0.4 ± 6.4

[11] MSTAR has less points than SRIM/Jav12 since not all targets are treated. ICRU73 has even less points since it is not defined below 0.025 MeV/nucleon. Hubert is only defined above 2.5 MeV/nucleon.
[12] For SRIM, these means all targets for which we have data; for MNCPX, the calculation covers only C, Al, Si, Ni, Ag, and Au targets.

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