Basic Physical and Chemical Information Needed for Development of Monte Carlo Codes

Mitio Inokuti[a]

Abstract

It is important to view track structure analysis as an application of a branch of theoretical physics (i.e., statistical physics and physical kinetics in the language of the Landau school). Monte Carlo methods and transport equation methods represent two major approaches.

In either approach, it is of paramount importance to use as input the cross section data that best represent the elementary microscopic processes. Transport analysis based on unrealistic input data must be viewed with caution, because results can be misleading. Work toward establishing the cross section data, which demands a wide scope of knowledge and expertise, is being carried out through extensive international collaborations. In track structure analysis for radiation biology, the need for cross sections for the interactions of electrons with DNA and neighboring protein molecules seems to be especially urgent.

Finally, it is important to interpret results of Monte Carlo calculations fully and adequately. To this end, workers should document input data as thoroughly as possible and report their results in detail in many ways. Workers in analytic transport theory are then likely to contribute to the interpretation of the results.

Introduction

In matter subjected to any ionizing radiation, energetic particles occur invariably. These include primary particles, charged or uncharged, and secondary particles such as electrons ejected in ionization processes. In track structure analysis, spatial distributions of collision processes of all the energetic particles and their consequences for molecules in matter are studied. The analysis amounts to an application of a branch of theoretical physics called statistical physics[1] (or physical kinetics[2] by the Landau school). Many of the known principles and results of the general theory of stochastic processes[3] are also valuable in

(a) Argonne National Laboratory, Argonne, Illinois 60439

particle transport analysis. The neutron transport theory[4] developed for the design of nuclear reactors also gives us important ideas and methods for treating electron tracks.

Monte Carlo methods[5-7] and methods of analytic transport equations[8] represent two major approaches to electron track analysis. These methods complement each other, rather than competing. To solve a complicated problem, e.g., a problem involving complex geometry, the Monte Carlo methods are more practical. To understand principles of physics, methods of transport equations are often transparent and effective.

In either approach, it is essential to use cross section and other data that correctly represent at least the most frequently occurring elementary processes. The primary purpose of the present discussion concerns the basic data necessary for track structure analysis.

Kinds of Basic Data Needed

The basic data needed can be classified into three kinds: (1) data on atomic structure of materials, (2) Spectroscopic data of materials, and (3) cross section data.

The data on atomic structure of materials must not be taken for granted. All too often data given in handbooks are incorrect or inappropriate for the problem at hand. An illustrative case was encountered during a comprehensive survey of stopping-power data of materials of radiological and dosimetric interest for the International Commission on Radiation Units and Measurements.[9,10] Data in the literature on carbon and silicon solids showed gross inconsistencies depending on the group of authors. The inconsistencies turned out to be largely attributable to measuring samples of different crystalline structures, and hence to different number densities of atoms.

With spectroscopic data, even advanced practitioners tend to take them for granted. Spectroscopic data, which concern energy levels of excited or ionized states of molecules, are highly nontrivial. The energy levels determine possible values of energy transfer from an energetic particle to the molecule. The character of each excited and ionized state, designated by a set of quantum numbers, governs the pertinent cross section, and also modes of decay such as dissociation into fragments, internal conversion, and fluorescnece. Knowledge of electronically excited states of all but the simplest small molecules is far from complete. Spectroscopic information about electronic excitation of DNA, RNA, proteins, and other biomolecules is extremely limited. Extensive research is needed as a prerequisite for mechanistic studies. Moreover, the energy levels depend on the environment of the molecule in question (the gas phase, the liquid phase, or the solid phase and pressure and temperature in the gas phase).[11,12]

Cross Section Data

At the Woods Hole Conference I gave a summary[13] of current knowledge about cross section data. (See in particular pp. 32-38 of Ref. 13.) To supplement that summary, I report here on three activities in which I am participating.

Work with the International Commission on Radiation Units and Measurements

One of the goals of the International Commission on Radiation Units and Measurements (ICRU) is to survey data on physical quantities basic to radiology and dosimetry, to reach an international consensus on the best recommendable values, and to disseminate results and conclusions in the form of ICRU reports. A subject of continuing study over two decades is the stopping power of materials for various charged particles. The first report[9] (on electrons and positrons) was published in 1984, and the second report[10] (on protons and alpha particles as well as muons and pions) is about to be published. Both reports were prepared by a committee chaired by M. J. Berger. A new committee chaired by P. Sigmund has begun to prepare a report on heavier ions.

The stopping power is the best studied of quantities describing the penetration of particles in matter, and knowledge of it is indispensable for quantification of radiation fields in dosimetry and is fundamental for modeling of radiation effects. Stopping power alone does not suffice to characterize track structure, but a result of a track structure analysis must be consistent with stopping-power data; otherwise it is untrustworthy.

The two ICRU reports[9,10] present the best possible values available and thus are standard references. They indicate a range of uncertainties in the recommended values and discuss important topics such as theoretical evaluation, methods of measurement, and the influence of chemical binding in molecules and of atomic aggregation in condensed matter.

A new ICRU program concerning the energy distribution of secondary electrons resulting from ionizing collisions of charged particles is now well underway under the chairmanship of M. E. Rudd. In view of its extensive work over nearly three decades, as reviewed by Toburen[14] and by Rudd et al.,[15] it is now appropriate for the ICRU to prepare a comprehensive report, which I expect to see in print in 1994.

Work with the International Atomic Energy Agency

The International Atomic Energy Agency (IAEA) has long been engaged surveys and data dissemination in selected areas such as dosimetric data for radiation protection, nuclear data for reactor research, and atomic data for fusion research. In 1985, the IAEA began to extend its activities to atomic and molecular cross section data for radiotherapy and related radiobiology and organized a meeting on this topic at Rijswijk.[16] In 1988, a second meeting was held in Vienna.[17]

Following recommendations of the Vienna meeting,[17] the IAEA initiated the Coordinated Research Program on Atomic and Molecular Data for Radiotherapy. The purpose of the program is to conduct an extensive survey of atomic and molecular data pertinent to radiology and dosimetry and eventually to prepare a comprehensive report. The participants are the following:

M. J. Berger, Bethesda, Maryland
Hans Bichsel, Seattle Washington
Ines Krajcar Bronić, Ruder Bošković Institute
D. T. Goodhead, Medical Research Council, Radiobiology Unit, Chilton
Yoshihiko Hatano, Tokyo Institute of Technology

Zdenek Herman, J. Heyrovsky Institute of Physical Chemistry and Electrochemistry
Mitio Inokuti, Argonne National Laboratory (Chairman of the Program)
I. G. Kaplan, Universidad Nacional Autonoma de Mexico
N. P. Kocherov, International Atomic Energy Agency (Scientific Secretary of the Program)
Tilmann Märk, Universität Innsbruck
H. G. Paretzke, GSF-Forschungszentrum für Strahlenschutz und Umweltforschung
Helmut Paul, Johannes-Kepler Universität, Linz
Pascal Pihet, Institut de Protection et Sûreté Nucleaires, Fontenay-aux-Roses
Leon Sanche, Université de Sherbrooke
Dušan Srdoč, Brookhaven National Laboratory
Michel Terrissol, Université Paul Sabatier, Toulouse
L. H. Toburen, National Academy of Sciences
Ernst Waibel, Physikalisch-Technische Bundesanstalt, Braunschweig
André Wambersie, Université Catholique de Louvain, Brussels

The work is close to completion; a final meeting to discuss a draft for the report will be held in June 1993 at Vienna. The contents of the report will be as follows:

Chapter 1. Development of charged-particle therapy and requirements for atomic and molecular data. (Wambersie, Goodhead, and Pihet)

Chapter 2. Ionization cross sections for charged particles. (Toburen)

Chapter 3. Electron collision cross sections. (Märk and Hatano)

Chapter 4. Interactions of low-energy electrons with condensed matter. (Sanche, Hatano, and Märk)

Chapter 5. Photoabsorption, photoionization, and photodissociation. (Hatano and Inokuti)

Chapter 6. Rapid conversion of initial ions and excited species in collision with other molecules. (Herman, Hatano, Sanche, and Märk)

Chapter 7. Stopping powers, ranges, and straggling. (Paul, Berger, Paretzke, and Bichsel)

Chapter 8. Yields of ionization and excitation in irradiated matter. (Srdoč, Inokuti, Bronić, Waibel, Hatano, and Kaplan)

Chapter 9. Track-structure quantities. (Paretzke, Goodhead, Terrissol, and Kaplan)

Chapter 10. Concluding remarks.

A Monograph on Cross Section Data

Another activity concerning cross-section data from a broader and more basic point of view is underway. I am editing a monograph entitled *Cross-Section Data*, to be published within a year as Volume 32 of Advances in Atomic, Molecular, and Optical Physics by Academic Press. The volume will contain 11 articles on various efforts to determine cross section data through experimental and theoretical studies, on needs for the data in selected applications, and on efforts toward compilation and dissemination of the data. However, the volume does not include extensive tables, graphs, or other presentations of the data themselves. Because of the great bulk of current data, they can be presented better in the form of computer databases than in a printed book. Similarly, no attempt has been made to cover all areas of atomic collision research in this volume; instead, the selected topics for the articles are illustrative rather than comprehensive.

Nevertheless, the volume should provide a guide to those who need to use cross section data with the best judgment and discretion, as well as to those who wish to produce better data through experimental or theoretical work. The volume will also convey a sense of the charms and challenges of what I would call *data physics*, a field that often fails to receive the appreciation and recognition that its importance warrants. I hope that the volume will serve educational purposes. The contents of the volume will be as follows:

Benchmark measurements of cross sections for electron collisions: Optical methods. (Lin, McConkey, Anderson, and Filippelli)

Benchmark measurements of cross sections for electron collisions: Analysis of scattered electrons. (Trajmar and McConkey)

Benchmark measurements of cross sections for electron collisions: Electron swarm methods. (Crompton)

Benchmark measurements of cross sections for simple heavy-particle collisions. (Gilbody)

Benchmark calculations of cross sections. (Schneider)

Analytic representation of cross-section data. (Inokuti, Dillon, Kimura, and Shimamura)

Electron collisions with N_2, O_2, and O: What we do and do not know. (Itikawa)

Needs for cross sections in fusion plasma research. (Summers)

Needs for cross sections in plasma chemistry. (Capitelli)

Guide for users of data resources. (Gallagher)

Guide to bibliographies, books, reviews, and compendia of data on atomic collisions. (McDaniel and Mansky)

Recommendations and Outlook

During the work on cross section data via the three above avenues, I found many issues that require study in the future. The following are three major issues.

1. Cross sections for charged-particle collisions with molecules in the gas phase remain far from well established. Even for simpler molecules such as H_2O, data are uncertain. For many of the polyatomic molecules of interest to radiation biology, cross section data are scarce. Cross sections of basic building blocks[18] of the DNA and proteins seem to be appropriate targets for immediate study.

Cross section data for individual molecules are important for interpreting and validating cross section data for condensed phases, which are generally harder to obtain directly from experiments.

2. Methods for determining cross sections, in both theory and experiment, are advancing greatly. Expanded support for cross section determinations will be highly productive for several years in the future, in view of the current status of techniques. One new area of study will be the influence of temperature, (i.e., rotational and vibrational excitations in the initial states of molecules due to thermal agitations) on cross sections for electron interactions with molecules. This topic is crucial to a full understanding of electron

thermalization and recombination with ions. Pioneering work[19,20] shows the feasibility of experiments on this topic, and is also stimulating to basic theoreticians.

3. Cross sections of molecules in condensed matter differ appreciably from those of isolated molecules, under circumstances that are generally understood. One circumstance concerns collisions involving slow electrons in either initial or final states, or both. Slow electrons in this context have sufficiently low kinetic energies (tens of eV and below) to be affected by forces due to condensed-matter structure. Knowledge about this topic is only beginning to be developed, as summarized in the forthcoming IAEA report. However, much remains to be done in both experiment and theory before fully convincing and comprehensive cross section data are established for any material.

Finally, I present the following recommendations to the workers in Monte Carlo studies of track structure and related topics:

1. Documentation of cross section data used in the published literature is generally too sketchy to allow reported studies to be reproduced. I certainly recognize that full documentation of all the input data will demand considerable effort. However, this effort is necessary to establish the credibility of the work and to improve its quality.

2. Results of Monte Carlo studies must be documented in the greatest detail possible. All too often we see such results presented only in figures and small tables. We can seldom identify the fraction of the uncertainty in the results that is due to limited sampling or the truly stochastic nature of the track structure. Documentation is desirable not only of final results but also of intermediate results. Analysis of intermediate results from a new point of view will be helpful for credibility and for deeper understanding of the physics involved.

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