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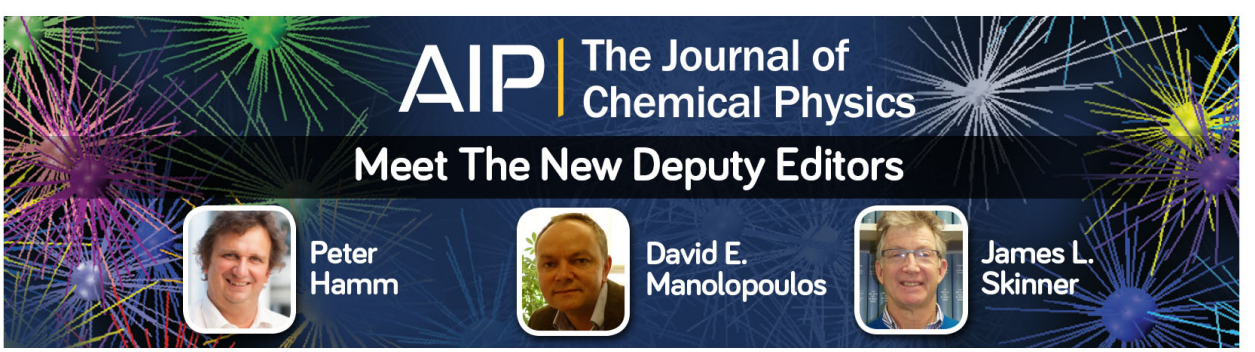
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Calculated yields and fluctuations for electron degradation in liquid water and water vapor[a)]

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Two sets of physical interaction cross sections for detailed electron track-structure calculations in liquid water and water vapor have been used to investigate the possible magnitude of phase-dependent differences in the primary yields of ionizations and their fluctuations produced during complete slowing down of electrons in the energy range from 10 eV to 10 keV. For fast electrons the calculated values of the mean energy absorbed per ion pair are 25.8 eV/ip for the liquid as compared to 30.0 eV/ip for the vapor; both results are consistent with experimental data. A similar phase effect is found in the ionization yields from each molecular subshell, since essentially the same partitioning of the total ionization cross section has been used in the calculations for the liquid and the vapor. The relative fluctuations of the ionization yields as described by the Fano factor are 0.15 for the liquid and 0.25 for the vapor; the Fano factors for each single molecular orbital are typically between 0.7 and 0.9 in both phases. The distributions in the total number of ionizations produced by the complete slowing down of electrons with initial energies above about 100 eV are Gaussian within about two standard deviations around the mean. The significance of the differences in the yields of ionizations and excitations calculated for both phases is discussed. In the absence of adequate experimental data for the basic processes, the differences are interpreted in terms of the respective assumptions made for single scattering processes.

INTRODUCTION

Quantitative knowledge of electron transport in water and the subsequent molecular changes produced is important in a number of areas of radiation physics, chemistry, and biology. In addition to its intrinsic physical interest, water is a solvent used in many chemical studies and is a principal constituent of many biological organisms. Understanding the effects of ionizing radiation on aqueous systems must ultimately be based on detailed knowledge of the type and location of all relevant molecular changes that radiation produces in water, i.e., on the track structure of individual charged particles.

In principle, such stochastic tracks can be simulated on a computer by Monte Carlo programs and then used for generating and testing hypotheses for radiation action on targets. Such detailed calculations require (1) identification of the relevant interaction and relaxation processes and (2) the quantification of the cross sections that describe the differential interaction probabilities and the subsequent reaction probabilities of newly formed physical and chemical species. In contrast to the rather extensive knowledge one has about changes in quantities that describe a radiation field interacting with matter (e.g., the fluence and energy spectrum of charged or uncharged particles), one knows much less about the types and amounts of molecular changes that are produced in the target material by radiation interaction. This lack of knowledge applies particularly to condensed matter. Only for some gases, including water vapor, can one at least identify and quantify those processes that contribute most to energy absorption from a radiation field and to the subsequent degradation of that absorbed energy.[1,2] For water vapor, cross sections for a large number of discrete and continuous electronic transitions have been measured by many authors for photons, electrons, and heavy charged particles over a wide range of energies.[3–12] These experimental data, together with the well established status of electron-scattering theories for dilute gases (e.g., Ref. 13) and the availability of integral electron-transport measurements (e.g., W-value measurements[14]) and energy spectra within the absorber,[15] permit the derivation of rather reliable cross sections for track-structure calculations for the gaseous form of water.[16,17]

Unfortunately, in spite of its greater relevance for many biological and chemical radiation effects, correspondingly detailed theories and experiments for the derivation of cross sections for liquid water are not available and do not appear feasible in the near future. For water, the only experimental data available specifically for the liquid phase are (i) optical reflection measurements for photons from several eV up to 26 eV,[18–20] (ii) stopping-power data for fast ions (summarized recently in Ref. 21), and (iii) yields of hydrated electrons for high-energy radiation.[22–25] From the optical reflection data, the real and imaginary parts of the complex refractive index and the complex dielectric constant ϵ can be obtained by a fit to Fresnel's equation or by a Kramers–Kronig analysis.[26] From the dielectric constant ϵ, the energy-loss function for collisions of fast charged particles with

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zero momentum transfer can be derived in the first Born approximation as $\mathrm{Im}(-1/\epsilon)$.[27] For collisions of slower particles, however, and for calculations with nonzero momentum transfer, other theoretical assumptions must be made.[27] Furthermore, in contrast to the vapor phase, no direct data exist for liquid water as a basis for assigning cross sections for forbidden transitions, dissociative electron-attachment collisions, and estimates of the fragmentation and deexcitation patterns of excited molecules. The latter is particularly important for understanding the subsequent chemical changes produced within a charged-particle track in liquid water.

The validity of the approximations in track-structure calculations for liquid water[28–30] cannot be tested directly against experimental data or generally accepted theories. However, the reported values for yields of hydrated electrons at early times, together with other radiochemical information, allow some consistency checks to be made.[31,32] The available data on the magnitude of stopping-power modification due to differences in the phase of the same material[21] are still too vague and contradicting to be of much help for the derivation of cross sections for track-structure calculations in liquid water.

Therefore, it appears useful to analyze empirically the magnitude and sensitivity of the effects on yields that phase-specific calculations might have. If these effects are small, then a resolution of discrepancies in physical assumptions may not be urgent for research on chemical or biological reactions following the primary radiation interaction. If, on the other hand, these effects are large and sensitive to the assumptions made, then the results of chemical-yield calculations and extrapolations from the gas to the liquid phase must be considered uncertain until the problems in the physical cross sections are resolved. In the latter case, as we shall see below, differences in calculated yields for liquid water and water vapor may not be due entirely to real phase differences, but partly to the still uncertain assumptions made for the liquid phase. Further research is needed to clarify these differences.

The yield of ionization is of paramount importance for understanding the action of radiation in many cases. Therefore, we focus on a comparison of the values for ionization yields and their fluctuations calculated for liquid water and water vapor. The possible origin and significance of the differences found are discussed in terms of the respective primary cross sections used to describe energy loss and subsequent target reactions.

METHOD OF CALCULATION

The calculations for water vapor were carried out with the Monte Carlo computer code MOCA-8 developed by one of us (HGP) at the GSF-Research Center. The cross sections used and results from MOCA-8 are in good agreement with those of Berger's code for water vapor and have been described elsewhere.[16,17,31] In short, evaluated experimental cross sections for water vapor for energy transfer, deexcitations, and fragmentation of the water molecule have been used whenever possible. The quantitative analysis of the important energy-loss channel leading to secondary electrons

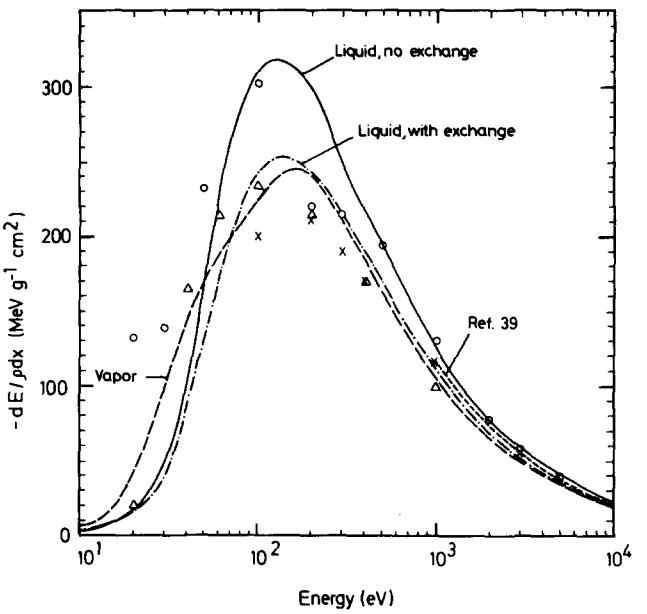


FIG. 1. Calculated mass stopping power of water as a function of electron energy. The newest calculations for the liquid include exchange and result in closer agreement with the mass stopping power of the vapor than before. Values for liquid water from other sources are shown for comparison: the dashed curve above 1 keV is from Ref. 39; O Ref. 40; × Ref. 29; △ Ref. 30.

was performed according to the recommendations of Platzman[33] and Kim.[34,35] The code has been tested with respect to yields and spatial transport quantities (depth doses, transmission, back scattering, etc.) and has been found to be consistent with available experimental data.[14,16]

The calculations for liquid water were carried out with the Monte Carlo code OREC, developed at Oak Ridge National Laboratory explicitly for the liquid state.[36] The ener-

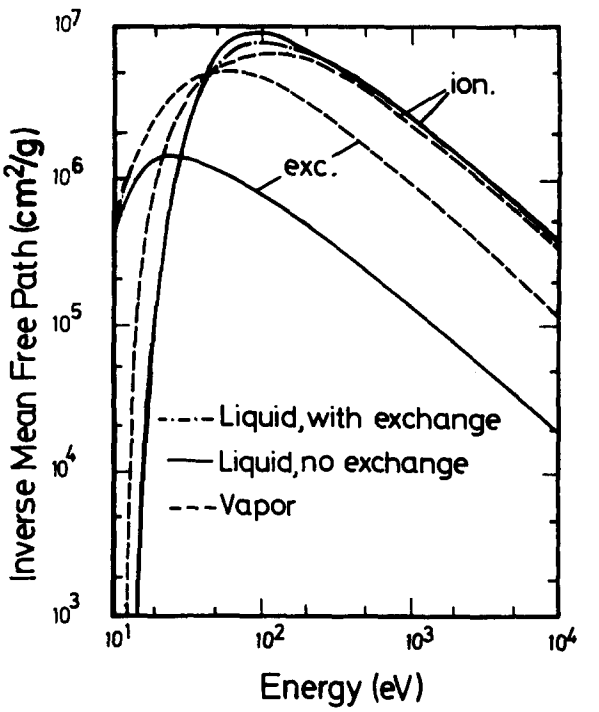
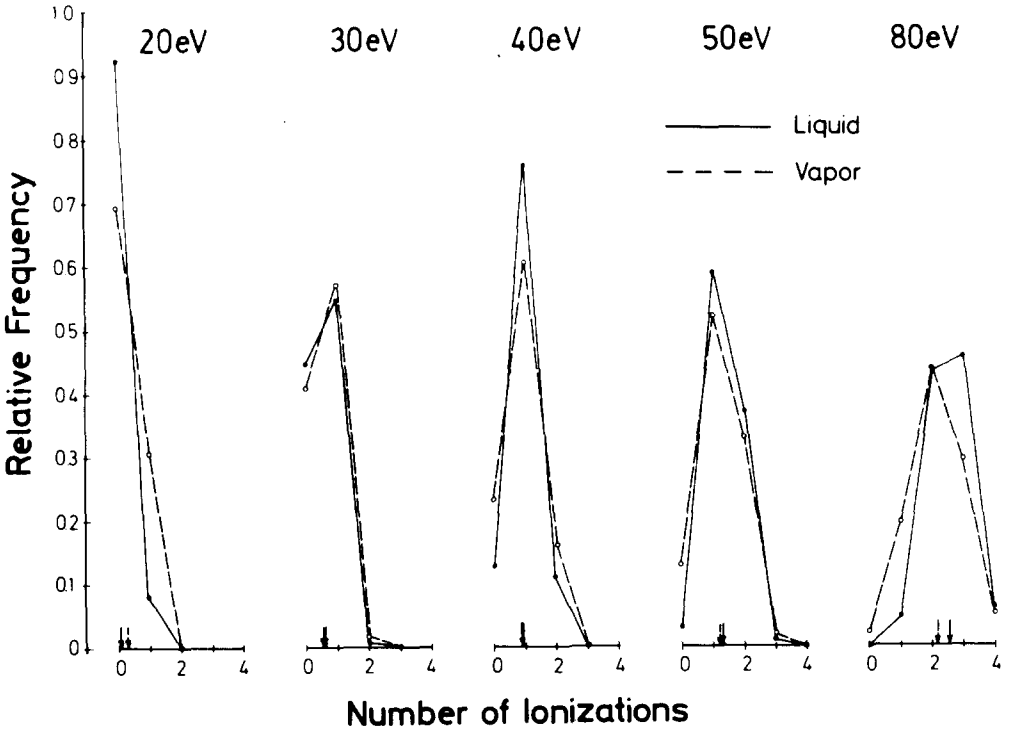


FIG. 2. Inelastic mean free paths as a function of energy for electrons in water. Values for excitation (exc.) and ionization (ion.) are shown separately.

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gy-loss cross sections are based on the dielectric response function derived from an analysis of UV reflection data for liquid water up to 26 eV.[18] In the absence of other specific data for the liquid state, the same experimental data for water vapor were considered in describing the liquid ionization efficiency, the relative participation of different molecular orbitals in producing ionizations, the fragmentation patterns, and (discrete) excitation transitions. The only explicit differences used for the liquid calculations as compared with those for the gas phase are:



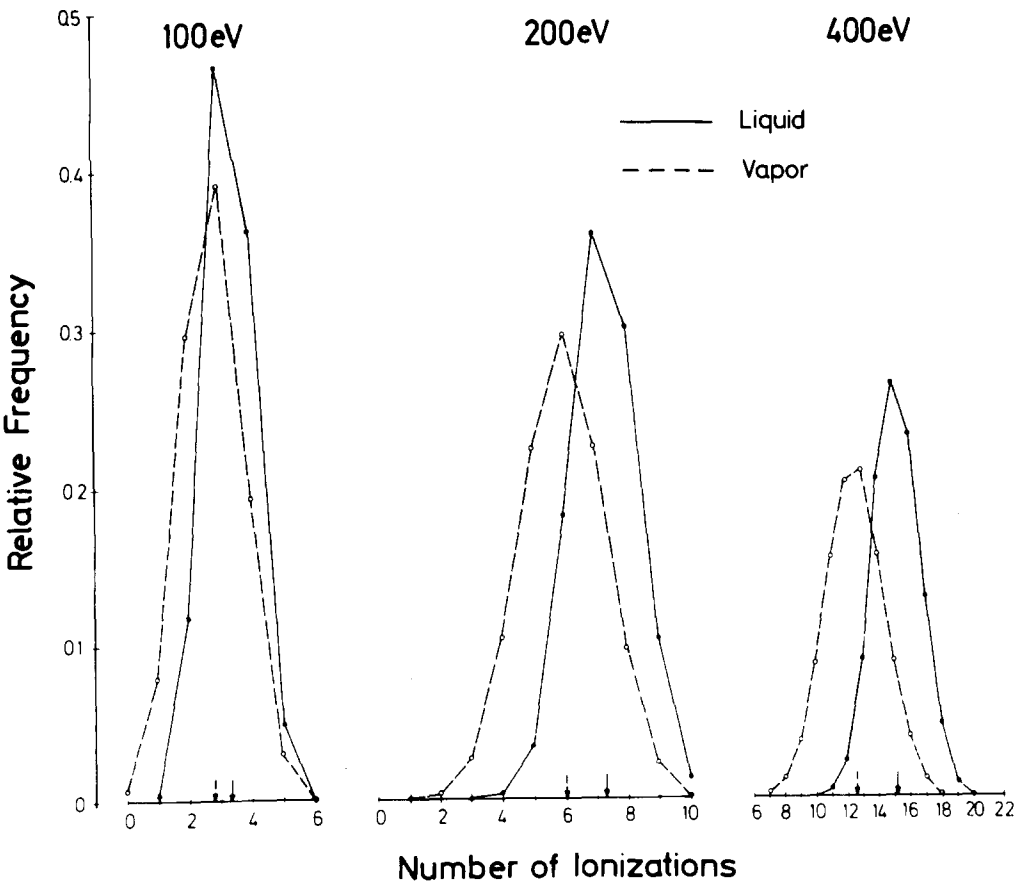


FIG. 3. Relative frequency of the numbers of ionizations produced by electrons (and all of their secondaries) of initial energies $E_0 = 20$, 30, 40, 50, 80, 100, 200, and 400 eV. All curves are normalized to unit area. Arrows on abscissa show the respective mean values.

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(i) A decrease in the binding energy of the four outer ionization levels by about 4 eV, as indicated by ESCA data of Siegbahn *et al.*[37]

(ii) A shift of the energy loss spectrum $\mathrm{Im}(-1/\epsilon)$ to higher energies because of the decrease of the real part ϵ_1 of the dielectric function above 10 eV.

(iii) A decrease and broadening of the oscillator-strength distribution for discrete transitions.

(iv) A radial delocalization of the sites of energy absorption for energy transfers less than 50 eV, to take account of the assumed interim collective sharing of energy by many molecules in the liquid phase. (This spatial aspect is of no relevance for primary-yield calculations, but is important for the subsequent reactions that determine final yields of various chemical species.)

Comparisons of several cross sections used in the GSF and ORNL codes and of some yield and transport results have been reported previously.[31,38] Electron exchange has been subsequently added to the ORNL code, and thus the present study modifies some details of the earlier results without invalidating the finding of general differences between the two phases. For example, as shown in Fig. 1, the inclusion of exchange brings the mass stopping power of the liquid in the energy range of its maximum closer to that of the vapor than calculated before.[31] It also agrees well with the independent calculation made for the liquid by Ashley,[41] his curve being about 5% higher in the peak region. The total yield of ionization remains essentially unchanged, and the total ionization cross section (Fig. 2) changes only in the region around 100 eV, i.e., in the vicinity of its maximum. Further descriptions of the cross sections and data used in these codes and their theoretical and experimental basis are given in Refs. 16, 17, 28, 31, 36, and 38.

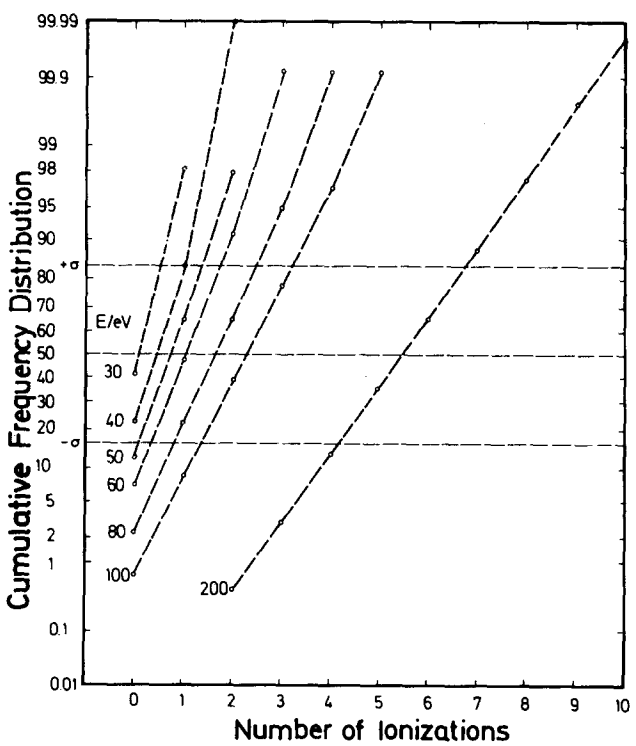


FIG. 4. Cumulative distributions for the number of ionizations produced in water vapor by electrons of initial energies E_0 from 30 to 200 eV. A Gaussian frequency distribution is represented by a straight line; the interval $\pm\sigma$, where σ is the standard deviation, is noted on the ordinate.

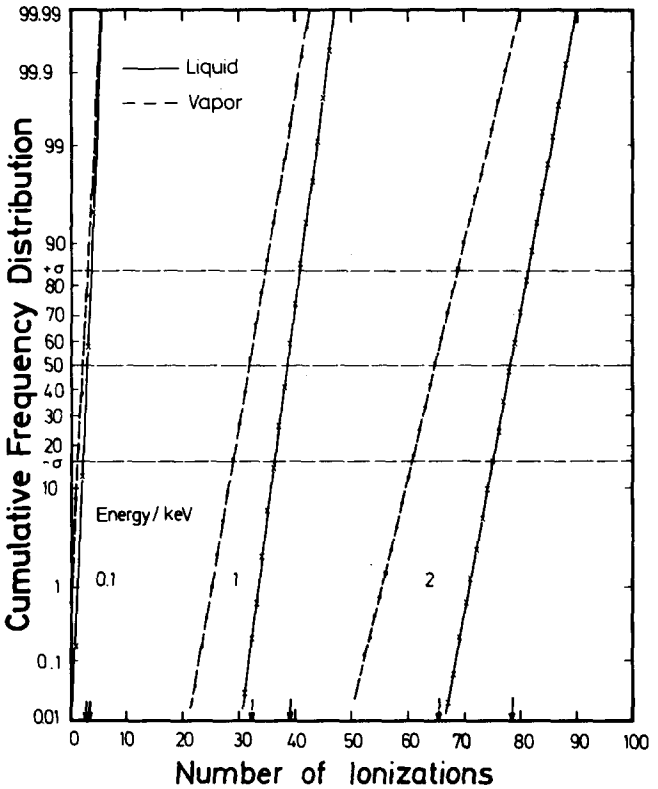


FIG. 5. Cumulative distributions for the number of ionizations produced in water vapor and liquid water by electrons of initial energies $E_0 = 0.1$, 1, and 2 keV. Arrows on abscissa show the mean values.

CALCULATED YIELDS AND FLUCTUATIONS

A large number of primary electrons (between 2000 and 20 000 at each energy) with initial energies E_0 between 10 eV and 10 keV and all of their secondaries were completely slowed down in water, as calculated by each computer code. The respective numbers of ionizations produced in the four outer shells and in the K shell were scored for each primary electron history. Because of the statistical character of electron energy-loss processes, the number of ionizations produced by an electron of initial energy E_0 is a stochastic quantity. Several examples of the calculated relative frequency $P(E_0,j)$ in the number of ionizations obtained for liquid water and water vapor are given in Fig. 3. It is instructive to compare these results in terms of the normalized cumulative distribution $P(E_0,n)$, defined by

$$P(E_0,n) = \frac{\sum_{j=0}^{n} p(E_0,j)}{\sum_{j=0}^{\infty} p(E_0,j)}.$$

Figure 4 shows a plot of the cumulative distribution for water vapor at a number of energies E_0; Fig. 5 gives a comparison of results for the vapor and liquid. For primary electrons with energies $E_0 \gtrsim 0.1$ keV, three distinct properties emerge:

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(1) The relative-frequency distributions are nearly Gaussian, at least in the range $\pm 2\sigma$ around the mean, independently of the physical state of water.
(2) The mean values of the distributions are larger for liquid water than for water vapor.
(3) The standard deviations σ of the distributions are smaller for the liquid than for the vapor.

The expectation value, or first moment, of the frequency distribution gives the mean number of ionizations $\bar{N}(E_0)$ produced by a primary electron of energy E_0 and all of its secondaries:

$$\bar{N}(E_0) = \sum_{j=0}^{\infty} jp(E_0, j).$$

This quantity is shown in Fig. 6 for the two phases.

The calculated yields per 100 eV absorbed energy, or G values, for ionizations from the various electronic shells of water in the two phases are shown in Fig. 7 together with the total ionization yields (top curves) and experimental data. The differences generally show expected trends. While the initial ionization yield has not yet been directly measured in liquid water, the measured initial yield of hydrated electrons may be considered as an equivalent quantity. Initial G values for the hydrated electron in the range 4.1 ± 0.2 have been reported from measurements carried out with fast pulse-radiolysis techniques[24]; however, data up to 20% higher can also be found in the literature. The calculated high-energy G value for vapor is 3.3 (corresponding to a W value of 30.0 eV per ion pair), which agrees well with experimental measurements.[14]

Fluctuations in the ionization yields are conveniently described by the Fano factor,[42,43] which is unity when fluctuations are governed by Poisson statistics. Because the number of ions that can be formed by the expenditure of a given amount of energy is limited (e.g., by the existence of an ionization energy threshold), Fano factors are generally less than unity. When the initial electron energy is low ($\lesssim 50$ eV) and the electron has a high probability of producing only a single ionization, however, the Fano factor begins to approach unity. Figure 8 shows the calculated results for total ionization and for ionizations from the various shells for liquid water and water vapor. Except at the lowest energies, the Fano factor for total ionization is smaller in the liquid. The Fano factors for the different shells are all generally greater than ~ 0.7.

Figure 9 shows the excitation yields per 100 eV absorbed energy for water vapor, for liquid water without exchange taken into account, and liquid water with exchange corrections. The perhaps surprising result seen here is that the excitation yield is larger in the liquid than in the vapor.

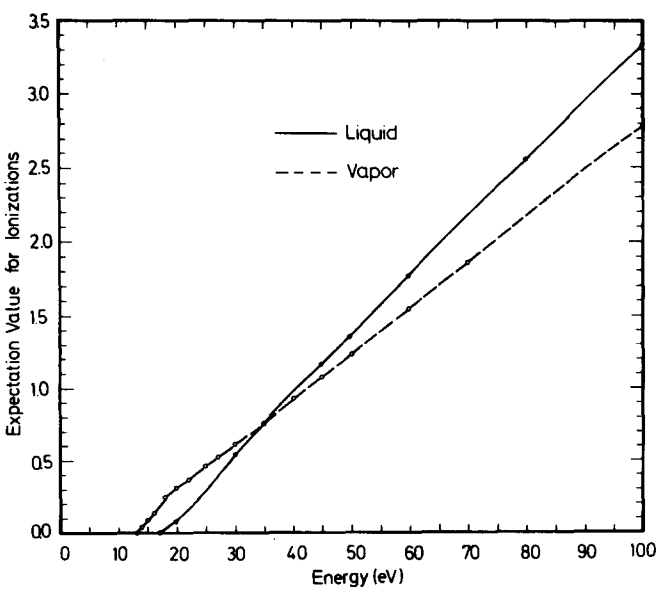


FIG. 6. Expectation value of the number of ionizations produced by electrons of different initial energies E_0 and all of their secondaries in water vapor and liquid water.

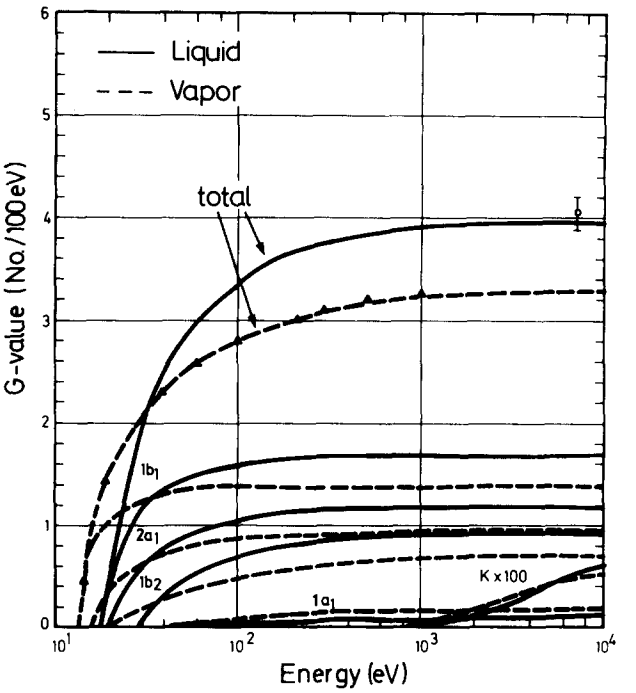


FIG. 7. Calculated total and partial yields for ionizations from the various shells in water vapor and liquid water as a function of initial electron energy. Solid triangles along the curve for total ionization in the vapor are the data of Combecher (Ref. 14). "Experimental" value shown on top curve is from Hunt (Ref. 24).

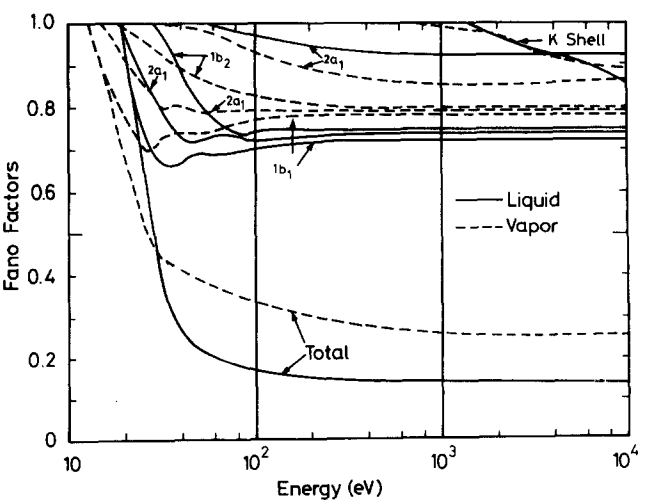


FIG. 8. Calculated Fano factors for total ionization and for ionizations from the various shells in liquid water and water vapor.

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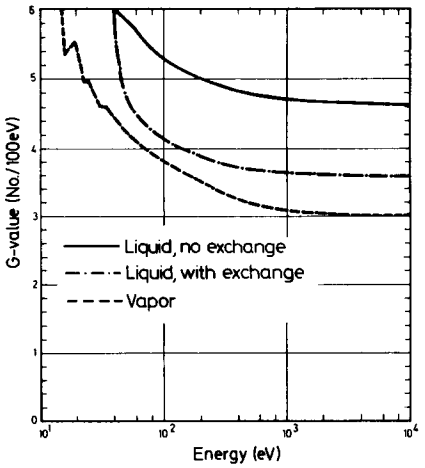


FIG. 9. Calculated excitation yields as a function of initial electron energy for water vapor, liquid water without exchange, and liquid water with exchange corrections.

DISCUSSION OF RESULTS

It is of interest to analyze the origin of the differences found here in terms of the basic physical information that is incorporated in the two computer codes, MOCA-8 and OREC. The codes were developed completely independently of one another, although, as pointed out above, much of the same experimental data was considered in both. The specific differences were enumerated above. Differences in the single collision energy-loss spectra are illustrated by the normalized curves for 5 keV electrons shown in Fig. 10 for the two phases. The energy-loss spectrum is "harder" for the liquid. This fact, coupled with the assumed smaller binding energies of the outer-shell electrons in the liquid, gives rise to the greater amount of ionization (lower W value) and smaller Fano factor for total ionization in the liquid.

Because of the harder collision spectrum and lower ionization potentials in the liquid, ionization accounts for about 95% of all inelastic collisions at higher energies, while the corresponding figure for the vapor is about 74%. This can be seen from the ratio of the ionization cross section σ_i and the total inelastic cross section σ_t which is shown in Fig. 11 for both phases. At ~ 30 eV the curves in Fig. 11 cross, and at the lower energies excitation accounts for a larger fraction of the cross section for the liquid than for the vapor. Therefore, at lower values of E_0 the excitation cross section in the liquid becomes relatively more important and the mean number of ionizations falls below that for the vapor. All secondary electrons get degraded in energy to well below 30 eV. These low-energy electrons lose a much larger share of their energy to excitations, rather than ionizations, in the liquid compared with the vapor. This greatly augments the excitation yield for the liquid at all initial electron energies.

The large value of the excitation yield can also be inferred from an inspection of the inverse mean free path for

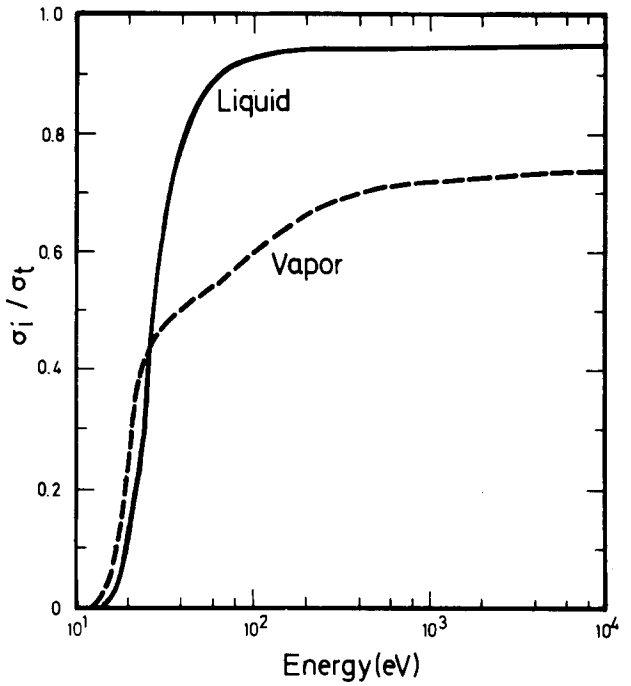


FIG. 11. Ratio of ionization (σ_i) and total inelastic (σ_t) cross sections for liquid water and water vapor.

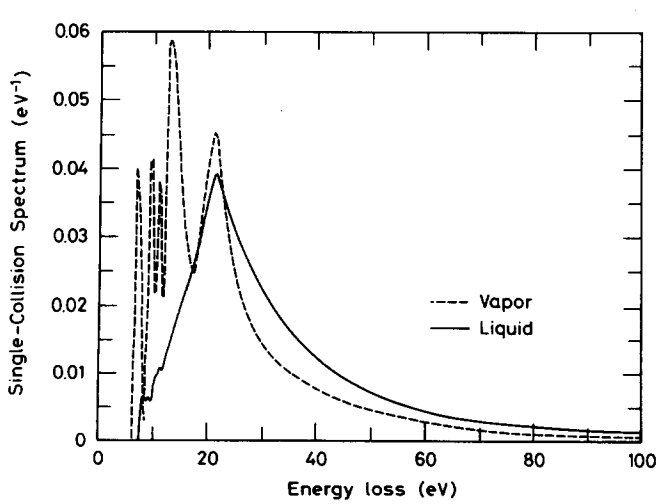


FIG. 10. Normalized single-collision spectra for 5 keV electrons in water vapor and in liquid water.

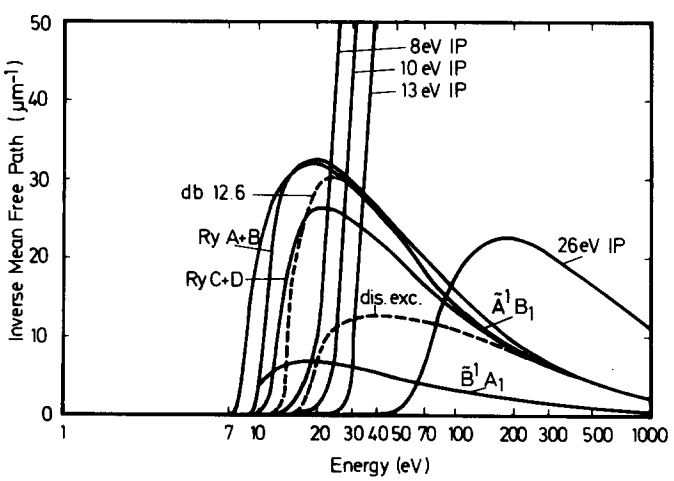


FIG. 12. Partial inverse mean free paths for various transitions as a function of electron energy in liquid water. Some curves are shown as dashes for clarity.

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different inelastic interactions in the liquid as a function of energy (Fig. 12). Below about 18 eV, excitation is practically the only mechanism of energy loss. In contrast, the partial inverse mean free paths for ionization in the vapor are still significantly larger compared with those for excitation in this energy range.

The differences in total and partial Fano factors are mainly due to the respective differences in the total numbers of ionizations in both phases.

SUMMARY

A systematic study has been made of calculated yields and fluctuations in connection with the complete energy degradation of electrons with initial energies up to 10 keV in liquid water and in water vapor. The differences and trends found can be explained in terms of the assumptions made for the basic cross sections for the two phases. Although the validity of some assumptions remains to be proven, in the absence of experimental data for liquid water, studies such as these might help in gaining a better understanding of the chemical and biological effects of ionizing radiation on aqueous systems.

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