Hydrogen Peroxide Production in the Radiolysis of Water with Heavy Ions

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The radiation chemical yields of hydrogen peroxide in water at neutral pH have been determined with protons, helium ions, and carbon ions at energies of a few to 30 MeV. The long-time yields of hydrogen peroxide increase with increasing linear energy transfer, LET, for protons and helium ions, but it decreases for carbon ions due to higher order reactions within the particle track. However, the maximum increase in hydrogen peroxide yields is only about 50% from γ rays (LET = 0.2 eV/nm) to helium ions (LET = 156 eV/nm). Methanol was used as an OH radical scavenger in order to probe the temporal dependence of hydrogen peroxide formation. The differences in the time dependence of the formation of hydrogen peroxide with various ions are discussed and compared to that observed with γ rays.

Introduction

Hydrogen peroxide is the main oxidizing molecular product formed during the radiolysis of water. It is formed primarily by combination reactions of OH radicals produced in the radiolytic decomposition of water. Therefore, its yield and chemistry are central to understanding the fundamental processes of the OH radical and other oxidizing species. On a more practical aspect, hydrogen peroxide produced in water radiolysis is involved in oxidation damage in almost every domain of nuclear technology where high linear energy transfer (LET = stopping power) radiation is involved. For instance, hydrogen peroxide scavenging of H atoms has a major role in the water cooling circuit of nuclear reactors.[1] Despite its importance, the few published reports on the production of hydrogen peroxide are somewhat inconsistent and there is virtually no information on the temporal variation of hydrogen peroxide with high LET heavy particles.

The effect of track structure on radiolytic yields with high LET particles has been investigated in aqueous solutions.[2−13] Most of the results concern the yields of molecular hydrogen and of the radicals formed by water radiolysis (H, OH, HO_2). No experimental data are available on the yields of hydrogen peroxide obtained with heavy ions as a function of the concentration of added scavengers, and there are few studies at neutral pH. It is known that the increased energy deposition density by high LET particles leads to an increase in hydrogen peroxide production.[14] However, the exact correlation between the effects of track structure on OH radicals, hydrogen peroxide, and other possible oxidizing species has not been established, in part because of lack of information on the hydrogen peroxide yield. It is especially important to know the temporal variation in the hydrogen peroxide yield in the particle track. The addition of a scavenger for the OH radical enables one to gain knowledge on the time scale of hydrogen peroxide formation and on the relationship between OH radicals and hydrogen peroxide. This information will help elucidate the role of oxidizing species in water radiolysis and on the general decomposition of water with high LET radiation.

TABLE 1: Characteristics of the Particles Used in This Work[a]

particle	energy (MeV)	track average LET in water (eV/nm)	range in water (mg/cm^2)
^{1}H	2	34.8	7.31
	5	20.7	35
	10	13.8	119
	15	10.3	246
^{4}He	5	156	3.56
	15	92.1	21.1
	20	78	34.7
^{12}C	10	787	1.35
	20	703	2.99
	30	629	5.06

[a] The track average LET is the stopping power averaged over the entire range of the particle.[24]

In this work, hydrogen peroxide yields were determined in the radiolysis of water at neutral pH with protons, helium ions, and carbon ions of different energies. The characteristics of the particles used are listed in Table 1. Some of these particles are particularly important for nuclear technology. For instance, 5 MeV helium ions are equivalent in energy to most natural α particles. The temporal variation of hydrogen peroxide formation was examined by the addition of various concentrations of methanol, an efficient OH radical scavenger. The experimental techniques are presented next, followed by the results and their discussion.

Experimental Section

The heavy ion experiments were performed using the facilities of the Nuclear Structure Laboratory of the University of Notre Dame Physics Department. The particles were produced and accelerated using a FN Tandem Van de Graaff. After acceleration, the ions were energy and charge-state selected magnetically with a resulting energy resolution of less than 0.1%. The window assembly and the irradiation procedure were the same as reported earlier.[10,11] The energy of the particles after passing through all windows was determined from the TRIM stopping power compilation.[15] The solutions were irradiated with completely stripped particles at beam currents of about 5 charge

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nA. Absolute dosimetry was obtained from the product of the integrated beam current and the particle energy. The doses given were about 70 krad (700 Gy) in 22 mL samples.

γ ray radiolysis was performed using a Gammacell 220 ^{60}Co γ ray source at the Radiation Laboratory of the University of Notre Dame. The dose rate was 1.6 krad/min (16 Gy/min). Samples were irradiated to total a dose of 70 krad (700 Gy). The Fricke dosimeter was used to determine dose rate.[14]

All solutions were prepared using water from a Millipore Milli-Q UV system. The samples consisted of N_2-saturated aqueous solutions containing 25 mM of sodium nitrate (reagent grade, Aldrich) and various concentrations of methanol (reagent grade, Fischer Scientific). The pH of the solutions was near neutral (pH $\approx$ 6.5) and did not change during the course of the experiments. The doses at the highest methanol concentrations (1 and 10 M) have been corrected according to the fractional electron density of the water. The hydrogen peroxide concentrations were measured using the Ghormley method, in which I^- is oxidized to I_3^- by the hydrogen peroxide.[16,17] The absorbance of the solution was measured at 350 nm using a diode array spectrophotometer (Hewlett-Packard HP8453). The molar extinction coefficient for the I_3^- was measured at 350 nm and calibrated with a standard solution of H_2O_2 (Fischer Scientific). It was found that $\epsilon_{350} = 25850\ M^{-1}\ cm^{-1}$, in good agreement with previous data.[17] The molar extinction coefficient of I_3^- changes slightly in mixtures of water and methanol, and for the 10 M methanol solution it was found to be 26000 $M^{-1}\ cm^{-1}$. The results using 10 M methanol give a minimum estimation of the yield because of the secondary reaction between the radical of the methyl alcohol and the hydrogen peroxide.[18] However, the agreement of the present results with those obtained in γ radiolysis with other chemical systems (see below) suggests that the contribution of this secondary reaction is small.

Results and Discussion

The energy deposited by all ionizing radiation is localized in nonhomogeneous regions along their path as they are stopped in the bulk of the water medium. Hydrogen peroxide, H_2O_2, is formed within these tracks mainly by the combination of OH radicals formed by the decomposition of water molecules, reaction 1. At very high LET, reactions of the hydroperoxyl radical, HO_2, may also lead to hydrogen peroxide formation, reactions 2 and 3.

$$\mathrm{OH} + \mathrm{OH} \rightarrow \mathrm{H_2O_2}; \quad 2k_1 = 1.1 \times 10^{10}\ \mathrm{M^{-1}\ s^{-1}} \qquad (1)$$

$$\mathrm{HO_2} + \mathrm{HO_2} \rightarrow \mathrm{H_2O_2} + \mathrm{O_2}; \quad k_2 = 9.8 \times 10^{5}\ \mathrm{M^{-1}\ s^{-1}} \qquad (2)$$

$$\mathrm{HO_2} + \mathrm{O_2}^- \rightarrow \mathrm{HO_2}^- + \mathrm{O_2}; \quad k_3 = 9.7 \times 10^{7}\ \mathrm{M^{-1}\ s^{-1}} \qquad (3)$$

All rate constants are from the compilation of Buxton et al.[19] unless otherwise stated specifically in the text. Hydroperoxyl radical yields are usually considered to be negligible, but they increase with increasing LET.[14] The maximum yield of hydroperoxyl radicals with low-energy carbon ions has been measured by LaVerne and Schuler to be approximately 0.25 molecules/100 eV.[11] This relatively low yield and the small rate coefficients suggest that for the present purposes the contributions of reactions 2 and 3 to the formation of hydrogen peroxide can be neglected. However, these reactions must be included in any refined track structure calculation.

In this work, methanol has been used as an OH radical scavenger, (4), to compete with the production of hydrogen peroxide in reaction 1. Sodium nitrate was also added to protect the hydrogen peroxide from further reaction with the hydrated electron, (5) and (6), and with H atom, (7) and (8).

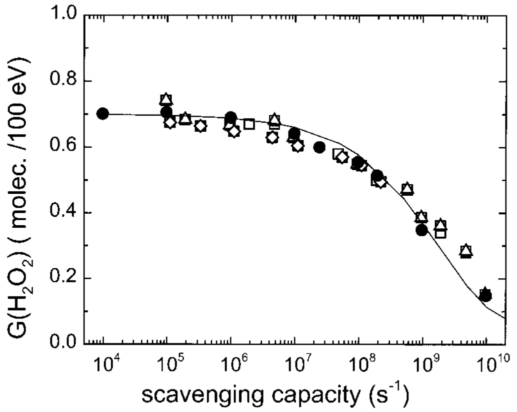

Figure 1. Hydrogen peroxide yields in γ radiolysis as a function of the scavenging capacity for OH radicals: (●) methanol and 25 mM nitrate, this work; (△) ethanol and sodium nitrate, ref 21; (□) ethanol and acetone, ref 22; (◇) aerated bromide, ref 20.

$$\mathrm{OH} + \mathrm{CH_3OH} \rightarrow \mathrm{CH_2OH} + \mathrm{H_2O}; \quad k_4 = 9.7 \times 10^{8}\ \mathrm{M^{-1}\ s^{-1}} \qquad (4)$$

$$\mathrm{e_{aq}}^- + \mathrm{H_2O_2} \rightarrow \mathrm{OH} + \mathrm{OH}^-; \quad k_5 = 1.1 \times 10^{10}\ \mathrm{M^{-1}\ s^{-1}} \qquad (5)$$

$$\mathrm{e_{aq}}^- + \mathrm{NO_3}^- \rightarrow \mathrm{NO_3}^{2-}; \quad k_6 = 9.7 \times 10^{9}\ \mathrm{M^{-1}\ s^{-1}} \qquad (6)$$

$$\mathrm{H} + \mathrm{H_2O_2} \rightarrow \mathrm{OH} + \mathrm{H_2O}; \quad k_7 = 9.0 \times 10^{7}\ \mathrm{M^{-1}\ s^{-1}} \qquad (7)$$

$$\mathrm{H} + \mathrm{NO_3}^- \rightarrow \mathrm{HNO_3}^-; \quad k_8 = 1.4 \times 10^{6}\ \mathrm{M^{-1}\ s^{-1}} \qquad (8)$$

By varying the concentration of methanol, one can probe the chemistry in the track using the competition between reactions 1 and 4. The resulting dependence of hydrogen peroxide yields on methanol concentration can be used to obtain information on its time scale of formation.

The chemical system ($NaNO_3$, CH_3OH, and the Ghormley analysis method) was first verified by comparing the $G(H_2O_2)$ obtained with γ rays to those values published in the literature for water at neutral pH.[20−22] (Radiation chemical yields are expressed as G values in units of molecules/100 eV of total energy deposition.) Figure 1 shows the hydrogen peroxide yields as a function of the scavenging capacity for OH radicals. The scavenging capacity is the product of the rate coefficient and the scavenger concentration and it is equivalent to the pseudo-first-order rate constant for the scavenging reaction. The inverse of the scavenging capacity gives the lifetime of the radical with respect to that reaction. It is seen in Figure 1 that there is good agreement between all the sets of data over the entire scavenging capacity range. Low scavenger concentrations correspond to long times in the track, and the limiting yield represents the amount of product that escapes the track into the bulk medium, the escape yield. The escape yield of hydrogen peroxide with γ rays is found to be 0.7 molecules/ 100 eV. Experiments show this value is linear with dose up to about 150 krad (1500 Gy). This linearity suggests that possible side effects due to the reactions of nitrate or its products are negligible.

Nitrate was added to the system to protect the hydrogen peroxide from attack by the hydrated electron and H atom, reactions 5−7. However, the addition of nitrate can also present problems. A high concentration of nitrate can scavenge the hydrated electron before the OH radical is scavenged. Since

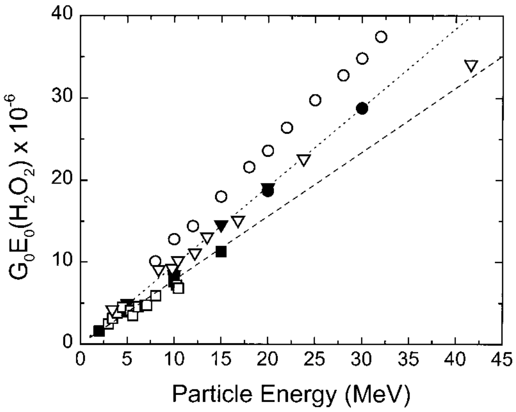

Figure 2. Production of hydrogen peroxide [$G_0E_0(H_2O_2)$] as a function of the initial particle energy, E_0: (■)^{1}H. (▼)^{4}He, (●)^{12}C, this work (10^{-5} M methanol, 25 mM nitrate); (○) ^{12}C, ref 12; (□) ^{2}H and (▽) ^{4}He, ref 7; The dashed line (---) gives the yield with fast electrons (0.7), ref 23, and the dotted line (···) with fission fragments (0.96), ref 5.

one of the primary radical reactions is between the OH radical and the hydrated electron, removal of one species will raise the apparent yield of the other. At the lowest concentrations of methanol used here, the concentration of the nitrate should be about 2.5 mM and not the 25 mM used in these experiments in order not to interfere in OH radical chemistry. However, it has been shown that the increase in OH yields with γ radiolysis is only about 5% over these nitrate concentrations.[22] This increase is within experimental error, and the higher nitrate concentration is better for minimizing effects due to H atoms, which may be important at the higher LET. The products of reactions 6 and 8 involve radical species that may scavenge primary radicals from the water decomposition or that interfere with the analysis of hydrogen peroxide. However, the data shown in Figure 1 involve several different radical scavenging systems and they give similar results. Any interference in the radiation chemistry or the analysis by the addition of nitrate is expected to be negligible.

The production of hydrogen peroxide, $G_0E_0(H_2O_2)$ (molecules/100 particles), by the proton, helium ion, and carbon ion irradiation of deaerated 10^{-5} M methanol with 25 mM sodium nitrate aqueous solutions is shown in Figure 2 as a function of the initial particle energy, E_0. Here, G_0 is the observed radiation chemical yield, which is the integral, or track averaged, yield since the particles are stopped in the solution. Also shown are the yields found with ^{12}C, ref 10; ^{2}H (plotted at one-half energy) and ^{4}He, ref 7; fast electrons, ref 23; and fission fragments, ref 5. The later experiments were performed at low scavenging capacity of OH radicals comparable to the present work. The studies with the light ions are the few for which hydrogen peroxide yields were examined over a substantial energy range. The near linearity of the lines for a given set of experiments with a particular particle shows that the integral radiation chemical yield is nearly independent of the particle energy and that track segment, or differential, yields are virtually equivalent to the integral yields. Therefore, to a first approximation, one can probe the effects of track structure by examining the integral or track averaged yields as a function of the track average LET.[24] As with γ rays, experiments show the production of hydrogen peroxide is linear with dose up to about 150 krad (1500 Gy). This linearity suggests that possible side effects due to the reactions of nitrate or its products are negligible.

There are noticeable differences between the yields obtained with the same type of particles that can be explained by variations in the experimental conditions. Many of the previous experiments were performed in acidic conditions (0.4 M sulfuric acid). With the same particle, the *G* values measured in this work at lower LET are 10−16% lower than those measured in acidic conditions. For example, the data of Burns and Sims[6] are about 15% higher than expected for the 20 MeV He ions examined here. At high LET, the pH effects are more striking. The data of LaVerne[12] for 10 MeV carbon ions are 50% higher than those found here. Apparently, the effect due to pH also depends on the LET. The lifetime of the hydrated electron is much shorter in acidic solutions than in neutral solutions because of its reaction with the hydrated proton. Hydrated electrons normally react with part of the OH radicals that would otherwise lead to the formation of H_2O_2, reaction 1. Removal of the hydrated electrons results in an increase in OH radical yields and in the subsequent hydrogen peroxide yields. The yields of hydrogen peroxide in a neutral solution are expected to be lower than in 0.4 M acid solution, which is observed. Note that in the present work a hydrated electron and H atom scavenger were used to eliminate their reactions with hydrogen peroxide, reactions 6 and 8. If the hydrated electron scavenger is not added, then one observes a decrease in hydrogen peroxide yields with decreasing pH.[25]

The results obtained by Schwarz et al. in neutral solutions are 11% higher than the present values.[2] They measured the hydrogen peroxide yield using an air-saturated potassium bromide solution. The *G* value was determined by correcting the measured hydrogen peroxide yield by the estimated contribution of hydroperoxyl radicals. This correction may be the source of the small differences in the data. The yields of hydrogen peroxide obtained with helium and carbon ions are very similar to those obtained by Bibler with fission fragments ($G = 0.96$ molecules/100 eV), although those measurement were made in 0.4 M sulfuric acid.[5] The striking feature of Figure 2 is that the *G* value for the formation of hydrogen peroxide is nearly independent of the nature of the particle or its energy. This result is also confirmed by deterministic track calculations that predict an increase of about 50% from γ rays to 1000 eV/nm particles, which is similar to the LET of carbon ions.[12] The calculations suggest that competing reactions between several oxidizing species are responsible for determining the hydrogen peroxide yield. These reactions become more significant with increasing LET due to the buildup of H_2O_2 and HO_2 to high concentrations in the track at short times. These products are then capable of scavenging radicals that would otherwise have reacted with each other. The near constancy of the hydrogen peroxide yields suggests that it would make a practical chemical dosimeter. An average *G* value of 0.88 is reasonably (within about ±20%) independent of particle type or LET.

The track of a low LET fast electron in water mainly consists of isolated spurs containing two to three ion pairs.[26] The OH radicals produced from the decomposition of water molecules can combine with sibling radicals or diffuse into the bulk medium. Reaction of OH radicals with other OH radicals obviously leads to the production of hydrogen peroxide. With increasing LET, the ionization events in water become, on average, more closely spaced from one another. At sufficiently high LET, the individual spurs coalesce into a track that is often depicted like a cylinder surrounded by spurs formed by the secondary ionization processes (δ rays). Increasing the density of radicals leads to an increase in molecular products. In Figure 3 are shown the radiation chemical yields of hydrogen peroxide obtained in the present work with protons, helium ions, and carbon ions as a function of their integral or track average LET.

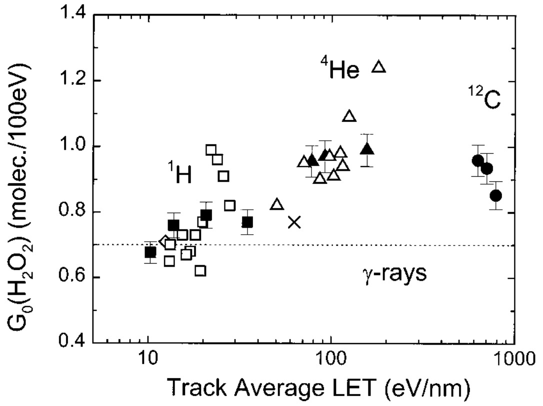

Figure 3. Hydrogen peroxide yields as a function of the LET for: (■) ^{1}H, (▲)^{4}He, and (●) ^{12}C (10^{-5} M methanol, 25 mM nitrate), this work; (□) ^{2}H and (△) ^{4}He, ref 7; (◇) ^{1}H and (×) ^{7}Li, ref 8. The error bars represent ±5%. The dotted line represents the limiting yield with γ rays.

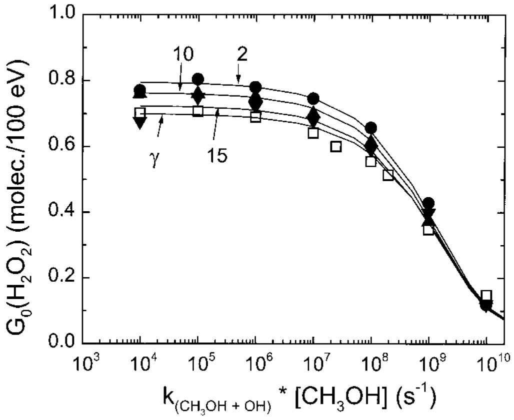

Figure 4. Hydrogen peroxide yields as a function of the scavenging capacity of methanol (25 mM nitrate) with protons of different energies: (●) 2 MeV; (▲) 10 MeV; (▼) 15 MeV. The results with γ rays are (□).

The data are determined with 10^{-5} M methanol concentration and represent the yields at about 100 μs when most of the track structure has collapsed. Also shown in Figure 3 are the data of Anderson and Hart[7] and Elliot et al.[8] at nearly the same OH radical scavenging capacity. It can be seen that for near neutral solutions the results agree. The data of Anderson and Hart at the lowest particle energies appear to be slightly high, but that could easily be due to straggling effects. As the LET of protons increases, only a small increase of $G(H_2O_2)$ is observed and the yields are very close to that obtained with γ rays. This result is expected since high-energy protons produce tracks of (on average) well separated spurs. Only at the lowest proton energies would one expect a cylindrical track with a resultant increase in hydrogen peroxide yields.

Irradiation with helium ions leads to an increase in hydrogen peroxide yields over that observed with γ rays. The LET with helium ions is more than 2 orders of magnitude greater than that for γ rays (0.2 eV/nm), but the yield of hydrogen peroxide is only greater by about 50%. The yield of hydrogen peroxide with carbon ions is similar to that obtained with helium ions. However, the yield of hydrogen peroxide increases with increasing helium ion LET and it decreases with increasing carbon ion LET. It is possible that some unknown experimental artifact is giving too low of a yield for the highest LET carbon ion and that the hydrogen peroxide yields are constant with carbon ions in this energy range. Nevertheless, it is readily apparent from the data that at least the yields of hydrogen peroxide are not increasing steadily with increasing LET. Such a scenario, as possibly suggested by the early helium ion data, is expected if the hydrogen peroxide chemistry is almost exclusively due to the combination of two OH radicals. Any leveling or drop in the hydrogen peroxide yield with increasing LET to the *G* values found here indicates that other, as yet unknown, reactions are depleting it. These reactions can become significant with relatively small increases in LET.

Burns and Sims[6] suggest that the concentration of hydrogen peroxide formed within the track can be considerable at very high LET and it may react with the other primary species, reactions 5, 7, and 9.

$$H_2O_2 + OH \rightarrow HO_2 + H_2O; \quad k_9 = 2.7 \times 10^7\ M^{-1}\ s^{-1} \quad (9)$$

Another possible explanation for the decreased yield of hydrogen peroxide may be due to the increased yield of the hydroperoxyl radical, HO_2, at high LET. This radical can also deplete OH radicals in the track.

$$HO_2 + OH \rightarrow H_2O + O_2; \quad k_{10} = 1 \times 10^{10}\ M^{-1}\ s^{-1}\ \text{(ref 27)} \quad (10)$$

reactions 9 and 10 could be competing with reaction 1 causing a decrease in the observed hydrogen peroxide yield. The exact nature and extent of the reactions leading to a decrease in hydrogen peroxide yields at high LET will probably only be fully explained by extensive track model calculations.

Figure 4 shows the influence of the OH radical scavenger concentration on the hydrogen peroxide yields for protons of different energy. It can be seen that the results are comparable to those found with γ rays, suggesting that the spatial distribution of the primary species is not very different for protons and for γ rays. This result is expected considering the coalescing of the spurs considered above. Cobalt γ rays consist of two photons of average energy 1.25 MeV.[28] These photons undergo Compton scattering to give fast electrons with an average LET of about 0.36 eV/nm.[28] The average energy loss event by electrons and, to a first approximation, protons involves about 60 eV.[29] Energy loss events are stochastic in nature, but on the average the spurs formed by these events are separated by about 170 nm. The typical spur has a Gaussian distribution of OH radicals with σ of only about 1 nm, and therefore spurs are well separated in γ radiolysis.[30] Only at an LET greater than about 15 eV/nm (60 eV/4σ) can one expect sufficient spur overlap along the main particle track that OH radicals in one spur can react to a significant extent with those from a different spur to increase the hydrogen peroxide yield. From the data in Table 1, low-energy protons are expected to have a track structure, and subsequent chemistry, much different than that of γ rays.

The hydrogen peroxide yield decreases with increasing methanol concentration for the same reasons given above for γ rays. Significant (>10%) scavenging of the OH radicals occurs at scavenging capacity greater than about 10^8 s^{-1}. This scavenging capacity suggests that the tracks (spurs) produced by γ rays and protons are very nearly dissipated by about 10 ns. Although the time scale may be similar for both types of particles, the difference in the yields in the limiting plateau region suggests that there are small variations in the distributions of radicals in the tracks.

The results of Figure 3 indicate that at higher LET the yields in the plateau region are dependent on particle type as is observed in Figure 5. This figure shows the dependence of

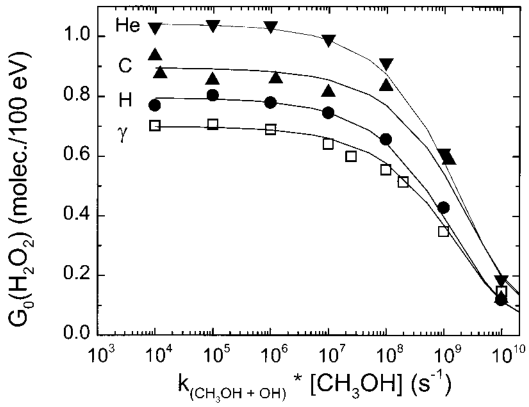

Figure 5. Hydrogen peroxide yields as a function of the scavenging capacity of methanol (25 mM nitrate) for: (□) γ rays; (●) 2 MeV ^{1}H; (▼) 5 MeV ^{4}He; (▲) 10 MeV ^{12}C.

$G(H_2O_2)$ on the OH radical scavenging capacity for γ rays, 2 MeV protons, 5 MeV helium ions, and 10 MeV carbon ions. The 5 MeV helium ions give the highest hydrogen peroxide yields. At high concentrations of OH scavenger, the curves for helium ions and carbon ions coalesce, which superficially could be interpreted as the probing of track regions with similar characteristics. This assumption is clearly wrong since the long-time yields are different. At high scavenging capacity, OH radicals are being scavenged at short times where transient concentrations can be high. The reaction of hydrogen peroxide with these transients may lead to an apparent lower yield. Further studies giving a direct comparison of the temporal variation of hydrogen peroxide with that for OH radicals will be very illuminating on the track processes at high LET.

The relative invariance of the hydrogen peroxide yield at scavenging capacities below about 10^8 s^{-1} appears to be in disagreement with the temporal evolution of the particle track as predicted with previous studies on OH radicals[31] and hydrated electrons.[32] Scavenger studies on the radicals suggest that considerable track chemistry can occur on the time scales of 10^{-9} to 10^{-6} s depending on the LET. However, a constant yield of hydrogen peroxide at low scavenger capacities indicates that the track has completely relaxed spatially and one is measuring the escape or long-time yield. Calculations have shown that the molecular yields, H_2O_2 and H_2, are almost always less sensitive to track conditions at long times relative to the radical yields, e_{aq}^- and OH.[33] Molecular yields are generally smaller than radical yields, so it is difficult to observe small relative changes. Furthermore, two radicals are involved in the production of one molecular product, so the net change of radicals is greater. The observed temporal variation in radical yields strongly suggests that the tracks of particles in the LET range of 10−100 eV/nm are still evolving on the microsecond time scale. It is important to have reliable data for both radicals and molecular products to properly assess the effects of track structure on the radiation chemistry of water.

Conclusion

The yields of hydrogen peroxide in water at neutral pH have been measured in solutions of sodium nitrate and methanol. It has been observed that the yields are nearly constant with the energy of the particle, suggesting that track averaged yields are virtually the same as track segment yields. Furthermore, the hydrogen peroxide yields depend very little on the nature of the particle over a wide range of LET. This result suggests that a hydrogen peroxide yield of 0.88 can be used as a dosimeter for heavy ions with an accuracy of ±20%. This information is useful for reactor technology since roughly the same yield can be used for a wide variety of different particles, including fission fragments, fast neutrons, and α particles.

Hydrogen peroxide yields show very similar OH radical scavenging capacity dependencies for the different types of particles examined here. Although the results would suggest similar distributions of transient species in the tracks, extreme caution must be used. The limiting yields at low scavenger capacities clearly show increased competition in the track at high LET. The increased importance of short-time reactions which decrease hydrogen peroxide yields may coincidentally give similar yields as particles with lower LET even though they have very different track structure. Detailed track model calculations in conjunction with results on other water species produced at similar LET will help elucidate the correct track structures and their effects on radiation chemical processes.

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