SUPPRESSED RADIOLYSIS OF HYDROGEN PEROXIDE IN WATER ICE - HYDROGEN MIXTURES.
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Introduction: Water ice has been identified as a major constituent of icy surfaces in the outer solar systems / interstellar medium, where it is bombarded by solar radiation, solar wind ions, cosmic rays, energetic particles trapped in the planetary magnetospheres and micrometeorites.

Radiation chemistry leads to the formation of species such as H, O and OH within the ice films from dissociation of water molecules due to ion bombardment. Oxygen enrichment occurs when the H atoms are preferentially lost to vacuum, due to its small size which favors the diffusion and desorption of H out of the ice films. The OH radicals, on the other hand, are much less mobile compared to the H atoms due to the cage effect, which tend to confine the OH radicals within the vicinity of the position from where they are formed, or the ion track. Most of the OH will recombine with H to form H_2O and the excess OH radicals can combine with OH radicals formed in the vicinity by a following ion or those diffused from nearby ion tracks, to form Hydrogen Peroxide (H_2O_2). The H_2O_2 production is found to decrease with increasing temperature, and is attributed to the competing phenomina of OH diffusion away from the ion track and hydrogen escape from the ice [1], so that H_2O_2 production at higher temperatures is less efficient due to higher H escaping rate.

Experiments of photolysis of H_2O_2 in ice films with 193 nm photons at various ambient O_2 pressures[2] showed that the oxygen enrichment in the irradiation environment enhances the H_2O_2 production. Radiolysis of ice films with 50 keV protons at low ambient O_2 pressure showed no significant enhancement of H_2O_2 production for fluences that saturates O_2 trapping due to ice compaction [3], a fluence that is lower than required to saturate H_2O_2 production. However, higher ambient O_2 pressures and higher total fluences might result in enhancement of H_2O_2 production, probably by transferring trapped O_2 molecules into H_2O_2 through radiation chemical reactions.

Inspired by the above oxygen enrichment experiments, we tried a different method of enriching hydrogen in the ice films by growing an ice + H_2 mixture, and studying H_2O_2 production under bombardment of 100 keV H^+ with H_2 ambient pressure in the vacuum chamber.

The Experimental Setup: The experiments were conducted in a UHV chamber with a base pressure of 10^{-10} mbar. Water ice films were vapor deposited onto a Li-He cooled gold coated quartz crystal microbalance at 20 K, through a collimating micro-capillary array doser. The areal mass of the deposited films was measured by converting the change of the resonant frequency of the microbalance due to mass increase to column density. The ice films were irradiated at 9 degree incidence with a mass-analyzed collimated 100 keV H^+ beam from a 300 keV ion accelerator. The reflectance spectra of the ice films within the region of 650 - 9000 cm^{-1} were collected at an incident angle of 35 degree with a Thermo-Nicolet Nexus 670 Fourier Transform infrared spectrometer (FTIR) at 2 cm^{-1} resolution. The reflectance R was obtained by dividing the reflectance spectra by the spectrum of the gold mirror, and then converted to optical depth, $-\ln(R)$. Methods for quantification of the infrared spectra are published elsewhere [4].

Results: In the first series of experiments, we grew pure ice films at 20 K at 45 degree incidence angle to enhance the porosity of the film, in order to enhance the adsorption of H_2 with an ambient pressure. We maintained constant ambient H_2 pressures between 3×10^{-8} and 3×10^{-5} Torr. Then we introduced 100 keV H^+ ions into the ice film (flux: $f = 1 \times 10^{12}$ $cm^{-2}s^{-1}$). The production of H_2O_2 was monitored by the intensity of the H_2O_2 absorption band at ~ 2860 cm^{-1}. We found that the H_2O_2 band area is not affected consistent with blank experiments without ambient H_2.

In comparison, we repeated the experiments with H_2O + H_2 mixtures instead of pure ice films. During the growth of the mixture, the ambient H_2 pressure was first introduced and maintained at a constant value, then H_2O vapor was leaked into the chamber. During the proton irradiation (100 keV H^+, flux: $f = 1 \times 10^{12}$ $cm^{-2}s^{-1}$), the ambient H_2 pressure was kept constant. We varied the ambient pressure during the growth of the ice mixtures and repeated the above procedures. Surprisingly, we found that at an ambient pressure $P > 3 \times 10^{-6}$ torr, the amount of H_2O_2 produced is negligible and the hydrogen enrichment leads to suppression of H_2O_2 production. For pressures $P < 3 \times 10^{-7}$ torr, we observed a partial suppression of H_2O_2 production. We explored the flux dependence of H_2O_2 production under ambient H_2 pressure (3×10^{-7} torr) and did not find a further suppression of H_2O_2 production for the lowest ion fluxes we used ($f = 5 \times 10^{10}$ $cm^{-2}s^{-1}$).

We investigated the temperature dependence. At 22 K, we repeated the experiments at ambient H_2 pressures upto 3×10^{-6} torr and did not observe a suppression of H_2O_2 production, suggesting that lower temperature favors the hydrogen enrichment that lead to suppression of H_2O_2 production, consistent with the reported findings [1] of lower H_2 escape rate from ice films at lower temperatures leading to higher H_2O_2 production.

Discussion: The results of this research showed that the H_2O_2 production in ice + H_2 mixture is suppressed compared to pure ice films, and the suppression is stronger at lower temperatures and higher ambient H_2 pressures. In the interstellar medium, ~99% of the mass are attributed to gas phase matter, ~3 quarters of which are H/H_2, ice rich grains may trap H/H_2 in the lattice when they are formed, Environmental radiation of these hydrogen enriched grains may not be able to produce as much H_2O_2 as in pure ice grains.

References:

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