

Hydrogen Atomic Radicals Stabilized in Hydrogen Clathrate Hydrate

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At sufficiently low temperatures, a single hydrogen atom (H atom) that becomes an atomic free radical by irradiation is known to be a relatively stable paramagnetic species in a wide variety of substances. Solid hydrogen in particular is a well-known quantum medium that has attracted much attention for its potential incorporation in novel materials having high-temperature superconductivity, because of its structural, magnetic, and electronic properties. These molecular properties of H atoms at ultra low temperature have been explored through the identification of unpaired electrons in H atoms observed by electron spin resonance (ESR) spectroscopy. Pure solid *para*-hydrogen has been used only as a valuable matrix material and enables high-resolution measurements via ESR spectroscopy due to its paired nuclear spins. Unlike pure liquid or solid hydrogen matrix, the successful imprisonment of a single hydrogen molecule in a cage could readily provide key clues for attaining a comprehensive understanding of the unique spectroscopic and dynamic patterns of the γ -irradiated hydrogen. Through combined ESR and NMR approaches, we attempted to identify the existence of ionized hydrogen derivatives entrapped in the cages of icy hydrate materials. Such stable existence of single hydrogen molecule/radical in icy crystalline matrices may offer significant advantages in exploring hydrogen as a quantum medium because icy hydrogen hydrates can be formed at milder conditions when compared with pure solid hydrogen, which requires an ultra low temperature of 4.2 K. The novel design and synthesis of ionic and radicalized hydrates are expected to contribute to inclusion chemistry. Furthermore, the stable existence of guest-derived radicals in the hydrate cages might lead to suggesting the preliminary concept of the hydrate-based nano-reactor because of their high reactivity.