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Photocatalytic H_2 production in natural fluid inclusions during Raman spectroscopic analyses

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Hydrogen (H_2) is hailed as the key energy source of the 21st century, while natural H_2, characterized by its abundant reserves, low cost, cleanliness, and renewability, is referred to as “golden hydrogen”[1]. However, the migration and accumulation mechanisms of natural H_2 are still poorly understood, and the exploration and development of natural H_2 reservoirs remain in their infancy[2]. Mineral inclusions serve as natural time capsules, preserving evidence of fluid activities. Raman spectroscopy is the most reliable method to determine H_2 in single inclusion at present. H_2's high reactivity and diffusivity preclude its long-term preservation in minerals, yet some reported H_2-bearing inclusions exhibit formation ages conflicting with theoretical preservation periods, which may lead to misinterpretations of H_2-bearing fluid activities. In this study, we conducted petrography and Raman spectroscopy on natural fluorite- and dolomite-hosted inclusions from the Ediacaran Dengying Formation in the Sichuan Basin, South China, containing CH_4+pyrobitumen±CO_2±H_2S±C_2H_6. We demonstrate the formation of H_2 within natural CH_4- and pyrobitumen-bearing fluid inclusions during Raman spectroscopic analyses, and exclude the potential for H_2 generation from pyrobitumen (Fig. 1). Raman spectroscopy temperature calibration and direct measurement by focusing the laser on a thermocouple show that the reaction temperature is approximately 32 °C. Our results indicate that, under 532 nm laser irradiation, pyrobitumen catalyzes CH_4 cracking, producing H radicals that rapidly recombine into H_2 within seconds to tens of seconds. If CO_2 exists in the inclusions, the H_2 yields is higher (11.5-43.0 mol%), while CO_2 is consumed and CO is produced (Fig. 1). This highlights the necessity of excluding artificially generated H_2 during spectral collection when studying the generation, migration, and accumulation of natural H_2 via fluid inclusion observations, to ensure the primary origin of H_2. Furthermore, considering that current H_2 production industries often face issues such as high reaction temperatures, catalyst deactivation, and significant greenhouse gas emissions, this low-temperature (32 °C) and rapid photocatalytic H_2 production may offer a novel perspective for industrial H_2 generation.

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2. Zgonnik, The occurrence and geoscience of natural hydrogen: A comprehensive review. *Earth-Sci. Rev.* **203**, (2020).