Radiolysis of Water as a Source of Bioavailable Chemical Energy

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Few geochemical studies have addressed decay of long-lived, naturally occurring radionuclides as a source of bioavailable chemical energy for microbes on Earth or other planetary bodies. Groundwaters in the vicinity of uraninite-pyrite mineralization are frequently associated with plumes of partially to fully oxidized sulfoxyanions inferred to result from radiolysis of water coupled to oxidative decomposition of sulfide minerals. When methane or other metabolizable organic constituents are present, radiolytic sulfate plumes are attractive habitats for microbial colonization. A series of experiments was conducted to explore this unusual biogeochemical pathway. Pyrite was reacted experimentally with millimolar solutions of hydrogen peroxide which is the main molecular oxidizing species produced during radiolysis. Stable sulfur isotopic compositions were determined for aqueous sulfate, mineral sulfate, and elemental sulfur recovered from sealed silica-tube experiments over a temperature range of 4 to 150°C. Using a cobalt-ring reactor at the Notre Dame Radiation Laboratory, pyrite was decomposed using only de novo generated oxidants. Ferrous ions, sulfide species, elemental sulfur, and sulfoxyanions appear to be active sources of transferable electrons in both the hydrogen peroxide and the radiolysis experiments. Similar seal-tubes experiments are in progress utilizing ammonia as reactant and recovering nitrate as the principal product of oxidation. Gaseous, aqueous, and solid products from radiolysis experiments provide critical insights into water-rock-microbe interactions in terrestrial and, potentially, extra-terrestrial natural environments where radiolysis is occurring as a result of radionuclide decay or ionizing irradiation.