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Advances in the application of *in situ* Raman spectroscopy technology in deep-sea extreme environments

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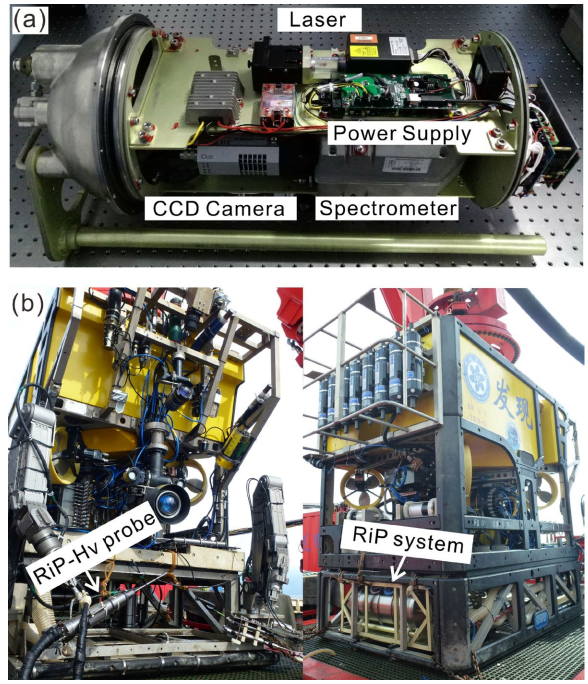

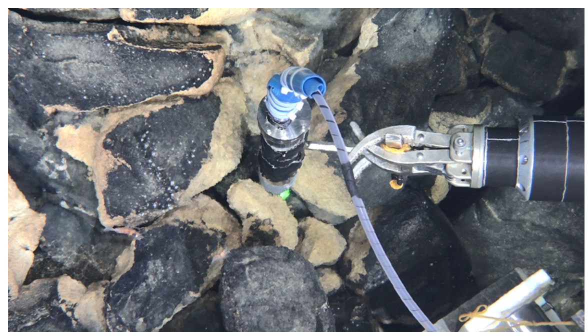

Deep-sea exploration technology is a critical factor in advancing fundamental deep-sea science. In recent years, in situ Raman spectroscopy has been widely used in marine science research, thanks to the pioneering efforts of two prominent scholars in the field of Earth sciences. Dr. I-Ming Chou, who developed a series of high-temperature, high-pressure experimental devices that brought Raman spectroscopy into the realm of quantitative research, and Dr. Peter George Brewer, who developed the world's first deep-ocean Raman in situ spectrometer systems (DORISS), have made substantial contributions to this field. Building on their exceptional work, we have recently developed a high-temperature resistant (up to 450°C) Raman insertion probe (RiP) for use in deep-sea extreme environments. Utilizing the high-temperature, high-pressure experimental methods established by Dr. Chou, we have created a series of Raman spectroscopic in situ quantification methods for dissolved gases, anions, and other components in deep-sea hydrothermal and cold seep environments.

Through the use of the RiP system, we discovered that methane concentrations in deep-sea reductive sediments are significantly underestimated. We also identified the presence of bare-leakage gas hydrates for the first time in the South China Sea. In hydrothermal fields, we observed the emission of natural supercritical carbon dioxide fluid and proposed a new model for the formation of early organic matter on Earth. We measured the fluid composition of ultra-high-temperature, low-density hydrothermal emissions and evaluated their influence on hydrothermal mineralization. We quantified the hydrothermal gas fluxes and found that diffuse flow environments exhibited volatile gas fluxes 1-2 orders of magnitude higher than those from focused flow environments. We also measured in situ pH levels of high-temperature hydrothermal vents and found that the pH of hydrothermal systems influenced by sediments is alkaline. In recent studies, using in situ Raman spectroscopy, we observed abundant in situ formate generation during CO_2 hydrogenation under hydrothermal conditions (≤230°C), which is several orders of magnitude higher than previously reported. The technology has also been applied to underwater hydrogen energy research, where hydrogen-rich fluid release was detected in the Okinawa Trough and Mussau Trench (Kunlun hydrothermal field), and the flux was quantified.