

Dating of sea-floor hydrothermal barite collected at the Okinawa Trough by ESR and radioactive disequilibrium

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The temporal change of submarine hydrothermal activities has been an important issue in the aspect of the evolution of hydrothermal systems which is related with ore formation (Urabe, 1995) and biological systems sustained by the chemical species arising from hydrothermal activities (Macdonald et al., 1980). Dating methods using disequilibrium between radioisotopes such as U-Th method (e.g. You and Bickle, 1998), ²²⁶Ra-²¹⁰Pb and ²²⁸Ra-²²⁸Th method (e.g. Noguchi et al., 2011) have been employed for such studies.

Okumura et al., (2010) made the first practical application of ESR (electron spin resonance) dating technique to a sample of submarine hydrothermal barite to obtain preliminary ages, while Kasuya et al. (1991) first pointed out that barite can be used for ESR dating. Toyoda et al. (2011) determined the optimum ESR condition while Sato et al. (2011) confirmed that the signal is thermally stable enough for an age range of several thousand years. Takamasa et al. (2013) obtained U-Th and ESR ages which are roughly consistent with each other.

The samples were taken by research cruises operated by JAMSTEC. Barite (BaSO₄) was extracted from hydrothermal sulfide chimney samples taken from two sites at the Okinawa Trough. Blocks of sulfide deposits were cut into pieces, and about 2.0g was crushed. The samples were soaked in 12M hydrochloric acid, left for approximately 24 hours. Then, 13M nitric acid was added. Finally, after rinsing in distilled water, the sample was filtered and dried. Impurities were removed by handpicking. An X-ray diffraction study was made to confirm that the grains are pure barite. After gamma-ray irradiation at Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency, they were measured at room temperature with an ESR spectrometer (JES-PX2300) with a microwave power of 1mW, and the magnetic field modulation amplitude of 0.1mT. The bulk Ra concentration was measured by the low background pure Ge gamma ray spectrometer. Assuming that Ra is populated only in barite, the dose rate was calculated with the alpha effectiveness of 0.043 (Toyoda et al., 2012), where the decay of Ra (a half life of 1600 years) was also taken into account.

The obtained ages range from 4.1 to 16000 years, being consistent with detection of ²²⁸Ra in younger samples and radioactive equilibrium/disequilibrium between radium and daughter nuclei. The variation of the ages within each sample is mostly within the statistical error range. The relative order of the ages is consistent with the result of ²²⁶Ra-²¹⁰Pb method, where the difference in absolute ages would be explained by several hydrothermal events that form the chimney. It was found that Yoron Hole field is the youngest, then, Daiyon-Yonaguni Knoll field, Hatoma Knoll field, being nearly equal to Iheya North Knoll field, then Izena Hole field, which is consistent with the direct observation from the submersible.

Keywords: barite, hydrothermal activities, electron spin resonance, dating