Production of the samarium-146 isotope and determination on its half-life

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論文要約

Samarium-146 with the half-life of 1.03×10^8 y is known as an extinct nuclide. If the nuclide is detected on the present Earth, it might be due to inflow of the nuclide that originates from supernova taking place in the vicinity of the Earth. Half-life with order of 10^8 is crucial value to exist on the present Solar system. Re-measurement of the half-life of ^{146}Sm is worthwhile to perform in order to search the nuclide on the present Earth. Atomic ratio and activity ratio of ^{146}Sm/^{147}Sm were measured by accelerator mass spectrometry and by alpha spectrometry, respectively, to determine the half-life of ^{146}Sm. The half-life of ^{146}Sm was calculated referring to the half-life of ^{147}Sm which is a naturally occurring long-lived radionuclide. Thus, the half-life of ^{147}Sm was also re-measured as a part of project for re-measurement of the half-life of ^{146}Sm. Moreover, it is considered that the nuclides produced in supernova explosions including the extinct nuclides accumulate onto deep-sea sediment or manganese encrustation together with extraterrestrial material. Manganese crust is a strong candidate to detect the extinct nuclides. However, accumulations rate of extraterrestrial material onto manganese encrustation have not been reported numerously. Isotopes of ^{10}Be, ^{230}Th, ^{231}Pa, and ^{239,240}Pu in the manganese crust were measured as a proxy for extraterrestrial material to estimate the deposition rate of extraterrestrial material.

学位論文要旨

Radionuclides with half-life of $< 10^8$ y are known as extinct nuclides. If the extinct nuclides are detected on the present Earth, it might be due to inflow of the nuclides that originate from supernova taking place in the vicinity of the Earth. The nuclide ^{146}Sm with the half-life of 1.03×10^8 y is known as one of the extinct nuclides. However, half-life of more than order of 10^8 y is crucial in existence on the present Solar system. Re-measurement of the half-life of ^{146}Sm is worthwhile to perform in order to search the nuclide on the present Earth. The author planned to derive the half-life of ^{146}Sm from atomic ratio and alpha activity ratio of ^{146}Sm/^{147}Sm, and the half-life of ^{147}Sm. Thus, the half-life of ^{147}Sm was re-measured prior to the measurement for that of ^{146}Sm. Moreover, it is considered that the nuclides produced in supernova explosions accumulate onto deep-sea sediment or manganese encrustation together with extraterrestrial material. Manganese crust is a strong candidate to detect the extinct nuclides. However, accumulation rates of the extraterrestrial material onto manganese encrustation have not been reported numerously. Isotopes of ^{10}Be, ^{230}Th, ^{231}Pa, and ^{239,240}Pu in the manganese crust were measured as a proxy for extraterrestrial

material.

Half-life of Samarium-147 In re-evaluation of the half-life of ^{147}Sm, known amounts of natural Sm and emitting standard (^{210}Po, ^{238}U, or ^{241}Am) were mixed well to prepare thin sources for the simultaneous counting of ^{147}Sm and the alpha emitting standard using a silicon surface barrier detector. The alpha activity of known amounts of ^{147}Sm was determined referring to the alpha activity of the standard. The sample preparation and counting were repeated to establish the reproducibility of the present half-life determination, and supplementary alpha-spectrometry was carried out by a liquid scintillation spectrometer. The counting efficiency of the liquid scintillation spectrometer is high (~100%), although, the spectrometer has disadvantages of high background and poor energy resolution. Hence, in spite of the agreement between the mean value measured with the liquid scintillation spectrometer, $(1.15 \pm 0.02) \times 10^{11}$ y, and the mean value with the surface barrier detector, $(1.17 \pm 0.02) \times 10^{11}$ y, within the error, the former value was regarded as a supporting data for the latter because of the disadvantages described above. The arithmetic mean of the half-life values obtained in this work for the nineteen counting sources is $(1.17 \pm 0.02) \times 10^{11}$ y. The error associated with the average value is on standard deviation. The average value is about 10% longer than the currently adopted value, $(1.06 \pm 0.02) \times 10^{11}$ y. In the earlier works, the half-life of ^{147}Sm was measured with a 4π gas flow counter, liquid scintillation spectrometers, and ionization chambers. According to the conventional method for the determination of the long half-lives, the half-life value for ^{147}Sm (T_{147}) is deduced by substituting measured value of A_{147} and N_{147} in the following equation:

$$T_{147} = \frac{\ln 2}{A_{147}} \times N_{147}$$

Hence, experimental errors in measuring A_{147} and N_{147} give inaccurate results for T_{147}. Impurities in Sm reagent bring error into the value of N_{147}, and uncertainty of counting efficiency, self-absorption of counting sample and radioactive impurities in Sm reagent bring error into the value of A_{147}. It is first suspected that the purity of Sm reagent was not sufficient in the earlier works. It is also suspected that the corrections for counting efficiency and self-absorption were not appropriate in the earlier works. High background and poor energy resolution of liquid scintillation spectrometers used in the earlier works might also result in inaccurate ^{147}Sm half-life. In contrast to the earlier works, in the present cautious work, all the error sources mentioned above were excluded. Hence, we conclude that the obtained in this work is reliable.

Half-life of Samarium-146 The samarium-146 atoms were produced in the reactions of ^{147}Sm(γ,n)^{146}Sm, ^{147}Sm(n,2n)^{146}Sm, and ^{147}Sm(p,2n)^{146}Eu(→^{146}Sm) to re-measure the half-life of ^{146}Sm. The Sm isotopes in target material were purified using the Ln resin and a cation exchange chromatography technique. The ^{146}Sm/^{147}Sm alpha activity ratios of the prepared samples were measured using a Si surface barrier detector. Measurements of ^{146}Sm with Accelerator Mass Spectrometry (AMS) using an Electron Cyclotron Resonance Ion Source (ECR-IS) were performed at Argonne National Laboratory (ANL). The Argonne Tandem-Linac Accelerator System (ATLAS) system was entirely tuned with ^{80}Kr^{12+} ($\delta(m/q)/(m/q) = 4.6 \times 10^{-3}$) pilot beam which has a mass-to-charge ratio close to that of ^{146}Sm^{22+}, prior to the AMS measurement of ^{146}Sm^{22+}. The ions produced in ECR-IS were extracted from the ion source, and analyzed through a bending trajectory with three analyzing magnets to select ions of interest, and then accelerated to 840 MeV. The ions were analyzed by a Gas-Filled Magnet (GFM) spectrometer. Finally,

data of position along the focal plane of the ions by Position-Sensitive Parallel-Grid Avalanche Counter (PGAC), multiple energy-losses along the ion path, ΔE_1–ΔE_5 and ΔE_{total}, and Time of Flight (TOF) were measured. The $^{152}Sm^{23+}$ ions which have a mass-to-charge ratio close to that of the $^{146}Sm^{22+}$ ion ($\delta(m/q)/(m/q) = 4.2 \times 10^{-3}$) were counted by a Faraday cup set up in the GFM beamline. The measurements with the cup were performed in between the measurements of the ^{146}Sm ions. Ions of $^{147}Sm^{22+}$ were also counted with the Faraday cup to derive the $^{146}Sm/^{147}Sm$ atomic ratio through the ratios of $^{146}Sm^{22+}/^{152}Sm^{23+}$ and $^{147}Sm^{22+}/^{152}Sm^{23+}$. Average half-life value was calculated to be $(4.42 \pm 0.35) \times 10^7$ y. 1σ of standard deviation was considered as an error included in the average value. The half-life obtained in the present work was shorter by a factor of ~0.4 than the currently adopted half-life value of $(1.03 \pm 0.05) \times 10^8$ y. However, we cannot exclude systematic uncertainties in the AMS experiments. Beam contaminants in the measurements of the $^{147}Sm^{22+}$ and $^{152}Sm^{23+}$ ions and transmission efficiency difference between the $^{146}Sm^{22+}$ and $^{152}Sm^{23+}$ ions from the ion source to detector are considered possible. However, in the present work, three kinds of irradiation were carried out to produce the ^{146}Sm samples that contain different activities and atomic ratios of $^{146}Sm/^{147}Sm$ in order to confirm influence from the activities and atomic ratios. No influences due to the activities and atomic ratios were observed in the work. It is considered that technological development for half-life measurement of ^{146}Sm nuclide was achieved except for the systematic uncertainties in the AMS experiment.

Accumulation Rate of Extraterrestrial Material onto Deep-Sea Manganese Crust The depth profile of contents of ^{10}Be, ^{230}Th, ^{231}Pa, and $^{239,240}Pu$ in a sample of manganese crust collected from the North Pacific Ocean were analyzed in order to estimate deposition rate of extraterrestrial material onto manganese crust in a project to search for supernova debris. Growth rate of the manganese crust was determined to be 2.3 mm My^{-1} from the depth profile of ^{10}Be. The deposition rate onto the manganese crust was determined to be 9.2×10^{-7} Bq cm^{-2} y^{-1} for ^{230}Th, 2.5×10^{-7} Bq cm^{-2} y^{-1} for ^{231}Pa, and 4.4×10^3 atoms cm^{-2} y^{-1} for ^{10}Be from the depth profiles of these nuclides. In discussion of the deposition process to manganese crust, we need to estimate the incorporation rate of radionuclide onto the manganese crust from the ambient water once the radionuclides arrive at the sea floor. The sediment incorporation rate of these radionuclides in the sampling site may be assumed equivalent to the fluxes of those radionuclides arriving at the seafloor, considering their high particle affinity in seawater. Consequently, the uptake rates into the manganese crust comparing with deposition rate onto deep-sea sediment are estimated 0.11–0.73% for ^{230}Th, 1.4–4.5% for ^{231}Pa, and 0.22–0.44% for ^{10}Be. On the other hand, same method as the cases for ^{10}Be, ^{230}Th, and ^{231}Pa cannot be applied to the uptake rate of $^{239,240}Pu$ because $^{239,240}Pu$ in seawater is not in a steady state in contrast to the natural radionuclides. It is considered that the Pu atoms in seawater below the sampling depth of the crust, and those in sediment might have interacted with the manganese crust. The uptake rate of $^{239,240}Pu$ onto the manganese crust compared with its total inventory is estimated to be 0.14%. The uptake rate of sinking particle onto the manganese crust is deduced to be 0.11–4.5% from the results of ^{10}Be, ^{230}Th, ^{231}Pa and $^{239,240}Pu$. These uptake rates correspond to uptake rate of ^3He which is known as a sensitive tracer for extraterrestrial material. The nuclides produced in supernova explosions, which incorporated into a particle of extraterrestrial material, deposit to the manganese crust. Deposition rate of the extraterrestrial material could be estimated using the uptake rate of particle. Influx of extraterrestrial material onto all over the earth was estimated to be 10^4–10^5 tons y^{-1}. The deposition rate of extraterrestrial

material into the manganese crust is estimated to be approximately 20–800 μg cm^{-2} My^{-1} using the uptake rate. On the other hand, the deposition rate of extraterrestrial material was estimated to be 9–90 μg cm^{-2} My^{-1} from comparison between the deposition rate of ^{10}Be onto the manganese crust and production rate of ^{10}Be on the whole earth. The deposition rates of extraterrestrial material estimated by two methods agree fairly well with each other.

学位論文審査結果の要旨

本学位論文について審査委員全員による予備審査を経て、平成19年2月6日に口頭発表及び質疑応答による最終試験を行い、同日開催の審査委員会において以下の判定を行った。

消滅核種の探索は宇宙からの物質の流入や地球化学的な物質動態に関係し、太陽系近辺で起こった超新星爆発との関連も示唆されており重要なテーマとなっている。サマリウム－146は最も半減期が長い消滅核種であるが、測定手法が確立されておらず、現在のところ天然試料からの測定例はない。本論文は探索の見積りにおいて重要な物理量となる当該核種の半減期再測定について検討した結果を述べている。まず、基準データとなるサマリウム－147 ついて半減期の追試測定を行い、現在通用している値より15%長いことを明らかにした。さらにサマリウム－146の半減期決定においては、ネオジム同重体が原子数定量の妨害であったが、米国アルゴンヌ国立研究所ATLAS施設において加速器質量スペクトルの取得を行い、世界で初めて同重体ネオジム－146と明確に分離してサマリウム－146を測定することに成功した。また、非常に遅い成長速度で海底に生成するマンガンクラストを消滅核種探索のための測定試料候補として選び、この試料中の地球外起源物質の堆積速度を放射性核種測定によって定量した。本論文はサマリウム－146の原子数精密測定に道を拓いたと同時に、消滅核種探索に有望な試料についても精密な分析を行っており学術的な価値は高い。

以上の成果を審査した結果、論文提出者は博士(理学)の学位に値すると判定したので報告する。