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^{182}Hf – FROM GEOPHYSICS TO ASTROPHYSICS

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^{182}Hf is a so-called "extinct" radionuclide interesting for both geophysical and astrophysical studies. The discovery of live ^{182}Hf in the early solar system through isotopic anomalies of stable ^{182}W in meteorites opened up an important application as a chronometer for the formation of the Earth and Moon. In addition, ^{182}Hf plays an important role for the understanding of nucleosynthesis of heavy elements in stellar environments, since both *r*- and *s*-processes can be responsible for the high abundance in the early solar system. In contrast to most other extinct radionuclides there is no significant natural or anthropogenic production of ^{182}Hf on Earth. Thus finding live ^{182}Hf on Earth today would be a strong indication for introduction of material from recent nucleosynthesis, e.g. from nearby supernovae. This paper gives a short overview of applications and summarizes recent and ongoing experimental nuclear physics measurements of ^{182}Hf.

1. INTRODUCTION

"Extinct" radionuclides act as a unique tool for the understanding of the birth of our solar system and thus provide some constraints on the origin of the elements. The evidence that these radionuclides were live (=present) in the early solar system comes from their stable decay products, now measurable through isotopic anomalies. Different geochemical evolution of radionuclide and stable daughter product can provide insight in formation and chronology of objects of the inner solar system.

^{182}Hf is one example of an "extinct" radionuclide with a high abundance in the early solar system, which has led to a powerful application as a geochronometer for dating the formation of objects in the inner solar system. The explanation of this high abundance challenges astrophysical models. However, some of the nuclear properties essential for the production, destruction and decay are not well known experimentally. A recent half-life measurement of ^{182}Hf resulted in a precise value, essential for the chronometer, but also necessary for the astrophysical interpretation of the data from the early solar system.

Additionally a direct detection of live ^{182}Hf in appropriate reservoirs on Earth using accelerator mass spectrometry (AMS) would provide evidence of ongoing nucleosynthesis of heavy elements near earth.

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2. GEOCHRONOMETER

^{182}Hf decays with a half-life of about 9 million years through ^{182}Ta ($t_{1/2} = 114$ d) into stable ^{182}W, which is the basis of a particulary useful chronometer for the early solar system, proposed by Norman and Schramm [1]. In the last few years this isotopic system evolved into one of the most used chronometers for time constraints on formation of objects of the inner solar system because (1) the half-life of ^{182}Hf is in the same order of timescale as for planetary accretion, (2) the initial ^{182}Hf isotopic abundance was high (^{182}Hf/^{180}Hf $\sim 1.0 \times 10^{-4}$ [2–4], or 1.6×10^{-4} [5]), (3) Hf and W were fractionated during planet core formation and (4) Hf and W are both refractory and thus not affected by high temperature processes in the early solar system. All these effects lead to well resolvable W isotopic anomalies in differentiated materials of objects which were formed during the lifetime of ^{182}Hf. These isotopic anomalies are in the order of 10^{-4} and are now measurable with high precision using MC-ICPMS (multi collector inductively coupled plasma mass spectrometry).

Successful applications are dating the fast core formation of the Earth (within a few 10 million years) and supporting models for the formation of the Moon by a giant impact of a Mars-sized body during a late stage of the Earth's accretion as well as the geochemical evolution of several meteorites (for a review see e.g. [6]). However, due to the high precision of this chronometers the results are in some conflict with other isotopic chronometers (e.g. U/Pb) [7]. One possible reason could have been the large uncertainty of the half-life of ^{182}Hf from the previous measurement with $(9 \pm 2) \times 10^6$ y [8]. With the new and much more precise value for the half-life (see sec. 4) this possible explanation is ruled out.

3. ASTROPHYSICS

This high initial solar system abundance of ^{182}Hf is a challenge for astrophysical models. ^{182}Hf is primarily an r-process nuclide. However, compared to other r-process nuclei (e.g. ^{129}I, $t_{1/2} = 1.7\times10^7$ y) the abundance of ^{182}Hf is higher than expected from nucleosynthesis models based on uniform production and a time interval of about 10^8 y between the last r-process production and the birth of the solar system. Thus Qian and Wasserburg [9] proposed the existence of two distinct r-process sites at different rates with only one responsible for ^{182}Hf. In contrast Meyer and Clayton [10] developed a model where the high abundance of ^{182}Hf is due to production by a 'fast' s-process in helium and carbon burning shells of massive stars. However, cross sections for nuclear capture reactions on ^{181}Hf and ^{182}Hf are experimentally not measured yet and models rely on theoretical considerations [11]. Additionally an accurate knowledge of the half-life of the nuclei involved in the models is crucial for an interpretation of the high solar system abundance.

4. NEW HALF-LIFE MEASUREMENT

The half-life of ^{182}Hf was remeasured based on the decay law, $t_{1/2} = \ln(2)N_{182}/A_{182}$, which requires absolute measurements of the activity, A_{182}, and the amount of the ^{182}Hf atoms, N_{182}, in the sample [12]. Our samples stem from Hf which was irradiated with an intense neutron flux by Helmer and Reich more than 30 years ago. We used two independent methods for determination of N_{182}:

In the first method N_{182} was measured by the activity of ^{181}Hf induced by a moderate neutron irradiation and the isotopic ratio of ^{182}Hf/^{180}Hf. Solid samples from the original ^{182}Hf material were irradiated together with standard samples at the TRIGA Mark II reactor of the Atominstitute in Vienna. In the second method N_{182} was determined using isotope dilution (with ^{178}Hf spike) and MC-ICPMS measurements at the Department of Earth Sciences, ETH Zurich. The original material was dissolved and four gravimetrical controlled samples were prepared in solution to avoid any differences in geometry to the γ-efficiency calibration samples. In both methods A_{182} was measured with HPGe detectors evaluating the most abundant γ-ray line (270.4 keV, absolute intensity of 0.790 ± 0.006, recently measured by [13]) following the β-decay to ^{182}Ta ($t_{1/2} = 114$ d). For γ-efficiency calibration mixed radionuclide solutions QCY44 and QCY46 from Amersham were used.

The results of individual measurement series were combined considering correlated uncertainties. Our final half-life value $t_{1/2}(^{182}\text{Hf}) = (8.90 \pm 0.09) \times 10^6$ y [12] is calculated as the weighted mean of the results of both independent methods.

5. SEARCH FOR LIVE ^{182}Hf TODAY USING AMS

"Extinct" radionuclides are also expected to be present today in the interstellar medium (ISM) as a result of recent nucleosynthesis, e.g. nearby supernovae [14]. Detection through γ-rays using γ-ray telescopes is only feasible for radionuclides with high overall activity (e.g. ^{26}Al, $t_{1/2} = 7.1 \times 10^5$ y). However, deposition of ISM grains by accretion onto Earth could make direct detection of some of these radioactivities possible [15]. This may be accomplished by finding minute traces of live atoms out of gram size samples from suitable terrestrial archives using Accelerator Mass Spectrometry (AMS). For the shorter lived radionuclides ^{60}Fe ($t_{1/2} = 1.5 \times 10^6$ y) a beautiful isolated event 2.8 million years ago was found in Fe-Mn crusts [16]. In addition, for longer-lived candidates with a relatively high steady state abundance (like ^{244}Pu, $t_{1/2} = 8.0 \times 10^7$ y) a significant continuous deposition should be expected [17]. Weak evidence for extraterrestrial ^{244}Pu was already found by [18].

^{182}Hf is another candidate since it has no significant natural or artificial production on Earth, mainly because of (1) the lack of heavy target nuclei in the atmosphere for production through spallation, (2) neutron-induced reactions start from unstable nuclides, and (3) the fact that ^{182}Hf is well above the fission peaks.

A sensitive AMS technique for ^{182}Hf has been developed at the Vienna Environmental Research Accelerator (VERA) in Austria, which requires high mass resolution (because of the stable Hf isotopes) and isobar separation (because of the stable isobar ^{182}W). The complete separation of stable isotopes at energies of 7–14 MeV and a reduction of ^{182}W by almost 10^4 using HfF_5^- as injection molecule into the 3 MV tandem accelerator, results in a detection limit of ^{182}Hf/^{180}Hf $\sim 10^{-11}$ with a detection efficiency of $\sim 10^{-4}$ [19].

Under these conditions a first search for ^{182}Hf from continuous deposition of ISM was performed in deep-sea sediment samples which were prepared at the Racah Institute in Jerusalem. So far only an upper limit for the abundance of live ^{182}Hf, expressed as flux from the ISM onto Earth of $< 2 \times 10^5$ atoms cm^{-2} y^{-1}, was found [20]. This is much higher than the expected value (~ 0.05 ^{182}Hf atoms cm^{-2} y^{-1} [20]) from simple considerations. The main limitation in this experiment was the low overall abundance of Hf in the sample due to the dilution with Zr which is chemically very similar. In addition this experiment revealed that an improved isobar separation at the detection stage is necessary for a clear identification of ^{182}Hf which is only possible at higher ion energies. At the Munich 13 MV MP tandem we are investigating the separation capabilities of isobars at about 1 MeV/u using a new detector approach. Other methods using a gas-filled detector at very high ion energies (> 1 GeV) provided by ATLAS at Argonne Nat. Lab. and inverse PIXE at the 14UD pelletron in Canberra are under development as well. An isobar suppression of at least 10^3 at the detector together with the W suppression at the ion source using HfF_5^- and chemical separation of Hf from W during sample preparation down to the ppm level should push the detection limit to the point where it may become feasible to find traces of supernova-produced ^{182}Hf on Earth.

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REFERENCES

1. E. B. Norman and D. N. Schramm, Nature 304 (1983) 515.
2. R. Schoenberg, et al., Geochim. Cosmochim. Acta 66 (2002) 3151.
3. T. Kleine, C. Muenker, K. Mezger, and H. Palme, Nature 418 (2002) 952.
4. Q. Yin, et al., Nature 418 (2002) 494.
5. G. Quitté and J. L. Birck, Earth Planet. Sci. Lett. 219 (2004) 2001.
6. A. N. Halliday and D. C. Lee, Geochim. Cosmochim. Acta 63 (1999) 4157.
7. A. N. Halliday, In: Treatise on Geochemistry, Vol. 1 (2003).
8. J. Wing, B. A. Swartz, and J. R. Huizenga, Phys. Rev. 1231 (1961) 1354.
9. Y.-Z. Qian, Progress in Particle and Nuclear Physics 50 (2003) 153.
10. B. S. Meyer and D. D. Clayton, Space Sci. Rev. 92 (2000) 133.
11. F. Kaeppeler, A. Mengoni and R. Gallino, Nucl. Phys. A718 (2003) 173c.
12. C. Vockenhuber et al., Phys. Rev. Lett. 93 (2004) 172501.
13. I. Ahmad, et al., Phys. Rev. C 70 (2004) 047301.
14. B. D. Fields, New Astr. Rev. 48 (2004) 119.
15. J. Ellis, B. D. Fields, and D. N. Schramm, Astrophys. J. 470 (1996) 1227.
16. K. Knie, et al., Phys. Rev. Lett. 93 (2004) 171103.
17. M. Paul et al., Nucl. Phys. A719 (2003) C29.
18. C. Wallner et al., New Astr. Rev. 48 (2004) 145.
19. C. Vockenhuber, et al., Nucl. Inst. and Meth. B 223-224 (2004) 823.
20. C. Vockenhuber et al., New Astr. Rev. 48 (2004) 161.