

Half-life determination of long-lived hard-to-measure isotopes: examples ^{53}Mn and ^{93}Mo

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Knowing the half-life of a radioactive isotope as precisely as possible is crucial for many research areas, for example nuclear astrophysics, nuclear energy technology or environmental research. Radionuclides with comparable long half-lives of hundreds to millions of years are in particular focus, not only due to their high impact in the corresponding scientific field, but also because of their possible effects on humans and the environment. Their half-lives cannot be determined by following the decay any more, but have to be measured by the direct method, requiring the determination of the specific activity and the corresponding number of atoms. Surprisingly, the half-lives of a considerable number of these long-lived nuclides are not well known, with data based on only a small number of – in some cases additionally very old – measurements. Some of them have large uncertainties or the results are even contradictory. Reasons for this situation, among others, are the low quantity and poor quality of the available samples. Especially for radionuclides that decay via electron capture only (so-called “hard-to-measure-isotopes”), the exact determination of the activity is a big challenge, because a radiochemically pure sample is required. This can be fulfilled either by a dedicated production route or isolation of the wanted isotope using preparative mass separation.

In our present contribution, we focus on two such isotopes, ^{53}Mn and ^{93}Mo . ^{53}Mn is a cosmogenic radionuclide, which is mainly produced in freeze-out phases of nuclear statistical equilibria of explosive burning in both thermonuclear and core-collapse supernovae explosions. Although the primordial ^{53}Mn is now extinct, its former presence can be observed by enhancements of its daughter ^{53}Cr in meteoritic inclusions originating from the Early Solar System. The $^{53}\text{Mn}/^{53}\text{Cr}$ chronometer could be used for precise dating of events in the Early Solar System on the timescale of 20 Ma [1], given that the half-life is known with the requested high accuracy and low uncertainty.

^{93}Mo is produced by neutron activation of stable ^{92}Mo , and, since molybdenum is a component of the construction materials in nuclear power plants (e.g. steel, Inconel alloys or cables), it becomes the dominant dose contributor in low and intermediate level radioactive waste when

the shorter-lived radionuclides have decayed. It is one of the few long-lived isotopes where no direct experimental determination existed so far.

The half-lives of both isotopes have recently been re-determined (^{53}Mn), respectively directly measured for the first time (^{93}Mo), using a combination of multi collector inductively coupled plasma mass spectrometry – at high mass resolution in the case of ^{53}Mn – for the determination of the numbers of atoms and liquid scintillation counting for determining the activity. While ^{93}Mo could be directly produced by proton irradiation of niobium and following chemical separation of molybdenum with a decontamination factor larger than 1.6×10^{14} to obtain a radiochemically pure sample, the ^{53}Mn sample, chemically separated from irradiated steel samples, underwent a mass separation using the RISIKO laser mass separator facility at Uni Mainz to get rid of the disturbing ^{54}Mn .

The determination of the activity is particularly challenging in the case of ^{93}Mo since the decay partly leads to the isomer $^{93\text{m}}\text{Nb}$. With the help of long-term liquid scintillation counting measurements and a very complex evaluation method, the decay probability of ^{93}Mo to $^{93\text{m}}\text{Nb}$ could be determined with unprecedented accuracy in addition to the activity and the half-life value [2].

We will present details on the sample preparation as well as the corresponding activity and mass spectrometry measurements and report on the results [2,3].

[1] Birck, J.L., Allègre, C. Manganese—chromium isotope systematics and the development of the early Solar System. *Nature* 331, 579–584 (1988). <https://doi.org/10.1038/331579a0>

[3] I. Kajan et al., First direct determination of the ^{93}Mo half-life, *Nature Scientific Reports* 2021, accepted

[3] J. Ulrich, PhD thesis 2020, High precision nuclear data of ^{53}Mn for astrophysics and geosciences, University of Berne, Switzerland