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On the half-life of ^{44}Ti by results in meteorites (*)

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Summary. — Measurements of the ^{44}Ti half-life display a large spread ranging from 46.4 to 66.6 years. The activity of this radioisotope measured in eight meteorites (chondrites) fell in the time interval 1883-1992 and calculated at the time of fall by the different values of $T_{1/2}$ shows that the shorter values are not reliable, if it is assumed that the average cosmic-ray intensity has remained the same during the past two centuries. The low activity of the cosmogenic isotope ^{44}Ti has been determined by means of a selective Ge-NaI (Tl) γ-spectrometer with a very low background (about 1 count per day in the γ-peak at 1157 keV of ^{44}Sc in equilibrium with its parent ^{44}Ti). The high stability of this system allows long-lasting runs ($\sim 10^7$ s), in order to achieve the standard deviation of counting up to $\sim 10\%$.

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1. – Introduction

Recently, interest in ^{44}Ti has significantly increased due to the potentiality of this radioisotope for the solution of many cosmophysical problems. In fact the ^{44}Ti isotope, produced by spallation reactions of cosmic rays on meteorites, can be used for investigating the century scale variation of solar activity [1-4] and for determining the terrestrial ages of meteorite finds (not seen to fall) in the hundred-year range. ^{44}Ti is also predicted to be present in young supernova remnants and in particular in the SN1987a. In all these problems, the knowledge of its half-life is crucial. Measurements of the ^{44}Ti half-life, determined by different methods, display a large spread ranging from 46.4 to 66.6 years [5-9]. We show here that the largest value, determined recently

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by Alburger and Harbottle [9], is the most reliable in order to explain the ^{44}Ti measurements performed in a series of meteorites which fell in the time interval 1883-1992.

2. – Measurement of ^{44}Ti in meteorites

The measurements of ^{44}Ti activities in different meteorites were performed with the purpose of searching for the century scale solar variability imprinted in extraterrestrial materials. In fact the solar variability affects the intensity of the galactic cosmic-ray (GCR) protons responsible for the isotope production in meteorites. The cosmogenic radioisotope ^{44}Ti has a very low activity in meteorites. The most convenient procedure is to measure ^{44}Sc ($T_{1/2} = 3.93$ h) activity which is in secular equilibrium with its parent ^{44}Ti. ^{44}Sc is a positron emitter (95%) and its emission is accompanied by a 1157 keV gamma-ray. For this purpose we have designed and set up a very selective γ-spectrometer. This system has been described in detail elsewhere [10, 11]. Briefly, a large-volume (~ 370 cm^3) high-purity germanium diode is used as the main gamma-ray detector. It is operated with a cylindrical umbrella consisting of 10 cm thick NaI(Tl) crystal assembly, in anticoincidence, to reduce the environmental background as well as in coincidence to obtain high selectivity for radioisotopes which decay by positron emission resulting in annihilation gamma-rays. The detector assembly is located in a 20 cm thick high-purity passive lead shield with 1 mm cadmium and 3 cm inner OFHC (Oxygen Free High Conductivity) copper casing. The system is housed in the underground laboratory of Monte dei Cappuccini in Torino, offering 70 m.w.e. (meters water equivalent) of shielding from cosmic rays. The γ-spectra are recorded by a PC-supported multichannel analyzer which stores data from Ge, Ge-NaI in anticoincidence and Ge-NaI in coincidence. N_2 (radon free) gas is flushed in the cavity between Ge and NaI detectors to reduce the radon level. Long-term background measurements demonstrated that the system has a very high stability both in energy and counting rate. Therefore, it can operate steadily for the long-lasting (more than 1 month) measurements.

Several stony meteorites which fell in the past 110 years, borrowed from various museums, have been measured. The counting efficiency for different gamma-rays was determined by multi standard gamma emitters doped sediment, having the same geometry as the Torino meteorite [1, 10]. ^{40}K, inherent in the meteorites, is used as an internal radioactive standard for the purpose of calculating the counting efficiency of each stone.

3. – Results

We measured the γ-activity of ^{44}Ti(^{44}Sc) is eight chondrites that fell in the time interval 1883 (Alfianello)–1992 (Mbale). In each measurement with Ge-NaI(Tl) in coincidence, the γ-peak at 1157 keV of ^{44}Sc is evident and the interference of the γ-peak of ^{214}Bi at 1155.2 keV present in the background and in the meteorites is negligible. As a consequence of the very low activity of ^{44}Ti, we need long-lasting runs of measurement ($\sim 10^7$ s) in order to attain results with a standard deviation up to 10%. A list of the analyzed meteorites with the time of fall, the period of the measurements and the activity in counts per day are reported in table I. Following the procedure reported at the end of the previous paragraph, we have evaluated the counting efficiency on the

TABLE I. – *Date of fall and counting data of the meteorites.*

Meteorite	Fall date	Counting period	^{44}Ti (c.p.d.)
Alfianello (L6)	16/2/1883	11/9/1992-2/2/1993	1.1 ± 0.2
Lancon (H6)	20/6/1897	28/3/1994-8/7/1994	1.9 ± 0.28
Olivenza (LL5)	19/6/1924	18/2/1993-22/6/1993	1.0 ± 0.2
Rio Negro (L3/4)	21/9/1934	29/5/1992-8/9/1992	1.9 ± 0.28
Monze (L6)	5/10/1950	29/11/1993-2/3/1994	1.2 ± 0.21
Dhajala (H3)	28/1/1976	31/7/1990-3/9/1990	3.0 ± 0.5
Torino (H6)	18/5/1988	January-March 1991	2.8 ± 0.3
Mbale (L5/6)	14/8/1992	12/7/1994-22/11/1994	2.9 ± 0.37

TABLE II. – *Calculated activity of* ^{44}Ti *at the time of meteorites fall for different values of half-life.*

Meteorite	K (p.p.m.)	(Fe + Ni) (%)	^{44}Ti (d.p.m./kg(Fe + Ni))			
			$T_{1/2} = 46.4$ y	$T_{1/2} = 48.2$ y	$T_{1/2} = 54.2$ y	$T_{1/2} = 66.6$ y
Alfianello	859	22.95	7.7	7.3	6.1	4.7 ± 0.9
Lancon	782	28.52	6.7	6.3	5.4	4.3 ± 0.6
Olivenza	917	20.26	7.9	7.6	6.8	5.8 ± 1.2
Rio Negro	830	22.57	7.3	7.0	6.4	5.6 ± 0.8
Monze	855	22.0	6.3	6.1	5.7	5.2 ± 0.9
Dhajala	830	28.74	4.6	4.6	4.5	4.4 ± 0.8
Torino	680	27.26	4.3	4.3	4.3	4.2 ± 0.5
Mbale	855	22.9	4.0	4.0	4.0	4.0 ± 0.4

basis of ^{40}K content in the meteorites, deduced by K concentration in each stone and used as an internal radioactive standard. In table II we report the calculated ^{44}Ti activity at the time of fall for different values of the half-life. Since ^{44}Ti is a spallogenic product, formed mainly in Fe and Ni in meteorites, we report the ^{44}Ti activities normalized to the target abundance present in the different chondrites. The value of $T_{1/2} = 46.4 \pm 1.7$ years was measured via Low-Energy Mass Spectrometry (LEMS) by Wing *et al.* [5]. With the same procedure (LEMS), Moreland and Heymann [6] measured $T_{1/2} = 48.2 \pm 0.9$ years. These are the values reported in the commonly used table of isotopes [7]. Freekers *et al.* [8] by means of Accelerator Mass Spectrometry (AMS) evaluated $T_{1/2} = 54.2 \pm 2.1$ years. In 1990, Alburger and Harbottle published [9] the measured value $T_{1/2} = 66.6 \pm 1.6$ years on the basis of real-time decay measurement over ~ 3 years.

In fig. 1 we show the ^{44}Ti activity (d.p.m./kg (Fe + Ni)) at the time of fall for $T_{1/2} = 66.6$ y and for the value reported on the table of radioisotopes [7] $T_{1/2} = 48.2$ y. For $T_{1/2} = 66.6$ y, the ^{44}Ti activity of Alfianello and Lancon meteorites which fell at the end of the past century is about equal to the activities of Dhajala, Torino and Mbale meteorites which fell recently. The activity of Olivenza, Rio Negro and Monze meteorites which fell in the time interval 1924-1950 is about 20% higher. This century scale modulation of the ^{44}Ti activity is consistent with the century scale solar activity

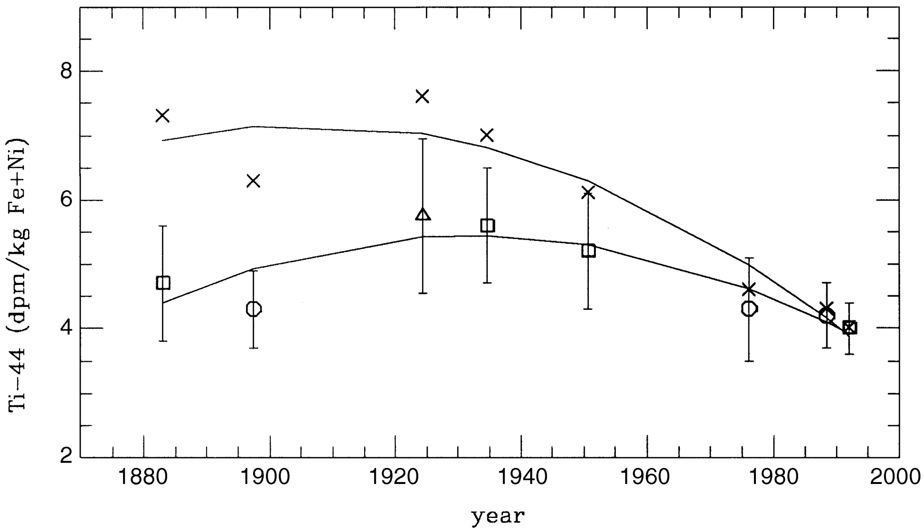

Fig. 1. – Variation of ^{44}Ti (d.p.m./kg (Fe + Ni)) in meteorites as a function of time of fall. Open simbols with standard deviations are for $T_{1/2} = 66.6$ y. Crosses correspond to the calculated activity adopting $T_{1/2} = 48.2$ y. Solid curves are the parabolic best fit of data.

variation, called Gleissberg cycle [3, 4]. It can be noticed that the higher activity of ^{44}Ti, related to the Gleissberg minimum at the turn of this century, is expected between 1925-1955. The phase difference between the sunspot minimum and ^{44}Ti maximum arises because of the integrating nature of the radioisotope production. The fact that we find comparable and low activities in meteorites which fell at the end of the past century and at present is due to the high solar activity periods during the second halves of the XIX and the XX century, as shown by the sunspot number series. On the contrary, from the same fig. 1, the ^{44}Ti activity for $T_{1/2} = 48.2$ y shows an increase back in time. In particular, the older meteorite, Alfianello, shows twice the activity with respect to the most recent Mbale meteorite. There is evidence that the solar activity has not changed significantly over the past two centuries, based on ^{14}C, ^{10}Be measurements in terrestrial archives like tree rings, ice cores and sea sediments, further supported by direct neutron monitor measurements since 1953. Therefore, the result of higher ^{44}Ti, if a half-life of 48.2 years is used, is not plausible and the meteorite measurements indicate that $T_{1/2} = 48.2$ y is too short a half-life for ^{44}Ti. Qualitatively similar results can be deduced by the values reported in table II for $T_{1/2} = 46.4$ y and 54.2 y.

4. – Conclusions

We measured the ^{44}Ti activity in eight chondrites that fell in the time interval 1883-1992. The very low activity of this radioisotope has been evaluated by means of a high selective Ge-NaI(Tl) γ-spectrometer with very low background in the underground station of Monte dei Cappuccini. The results obtained on the ^{44}Ti activity in the meteorites at the time of fall and for the different values reported in the literature, of the half-life of this radioisotope demonstrate that the most reliable value is $T_{1/2} = 66.6$ y obtained by Alburger and Harbottle on the basis of real-time decay measurement over ~ 3 years. The shorter values of half-life give an increase of the ^{44}Ti activity in meteorites going back with the time of fall which cannot be justified since it would imply decreasing GCR flux with time, contrary to ^{10}Be, ^{14}C and neutron monitor observations.

On the basis of the measurements presented here, we cannot deduce the half-life of ^{44}Ti with a precision better than the direct estimation by following its decay. Since ^{44}Ti has applications in several cosmophysical problems, and its half-life value is crucial, new direct measurements are in progress in different laboratories as well as in our laboratory, for obtaining a more precise estimate of its half life.

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