Applied Radiation and Isotopes 94 (2014) 23–29

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Radioisotope studies of the farmville meteorite using $\gamma\gamma$-coincidence spectrometry

Chris Howard [a,b], Megan Ferm [c], John Cesaratto [a,b], Stephen Daigle [a,b], Christian Iliadis [a,b,*]

[a] Department of Physics and Astronomy, The University of North Carolina at Chapel Hill, Chapel Hill, NC 27599, USA
[b] Triangle Universities Nuclear Laboratory, Durham, NC 27708, USA
[c] Keene State College, Keene, NH 03435, USA

HIGHLIGHTS

- We measure the radioisotope ^{26}Al in the Farmville meteorite.
- Coincidence γ-ray spectroscopy has distinct advantages over other techniques.
- Many different modes of detection allow for a more reliable determination of absolute activity.

ARTICLE INFO

Article history:
Received 24 July 2013
Received in revised form
23 May 2014
Accepted 2 July 2014
Available online 10 July 2014

Keywords:
Farmville meteorite
^{26}Al
γ-ray spectrometry
Monte Carlo simulations

ABSTRACT

Radionuclides are cosmogenically produced in meteorites before they fall to the surface of the Earth. Measurement of the radioactive decay of such nuclides provides a wealth of information on the irradiation conditions of the meteorite fragment, the intensity of cosmic rays in the inner solar system, and the magnetic activity of the Sun. We report here on the detection of ^{26}Al using a sophisticated spectrometer consisting of a HPGe detector and a NaI(Tl) annulus. It is shown that modern γ-ray spectrometers represent an interesting alternative to other detection techniques. Data are obtained for a fragment of the Farmville meteorite and compared to results from Geant4 simulations. In particular, we report on optimizing the detection sensitivity by using suitable coincidence gates for deposited energy and event multiplicity. We measured an ^{26}Al activity of 48.5 ± 3.5 dpm/kg for the Farmville meteorite, in agreement with previously reported values for other H chondrites.

1. Introduction

Most meteorites are fragments of ancient parent bodies that formed from planetesimals between the orbits of Mars and Jupiter at about the same time as the planets of the solar system (Herzog, 2007). As soon as a meteoroid is excavated from an asteroid after a collision, it is exposed to cosmic radiation, consisting mainly of protons and α-particles of very high energies. The cosmic radiation has two components: the first originates from the Sun (solar cosmic rays) and the second arises from outside the solar system (galactic cosmic rays). Both components are sufficiently energetic to initiate nuclear reactions in meteoroids. These spallation reactions (and associated secondary reactions, such as neutron-capture) on meteoroid target elements give rise to the production of radioactive and stable isotopes, referred to as cosmogenic nuclides. An important example of a radioactive cosmogenic nuclide is ^{26}Al (Cameron and Top, 1974; Hampel and Schaeffer, 1979). Since the exposure of most meteoroids to cosmic radiation is longer than several million years, radionuclides formed by cosmic radiation with half-lives less than a million years are in equilibrium with the cosmic ray intensity. After a meteorite falls to the surface of the Earth, it is shielded by the atmosphere from further cosmogenic production. Consequently, its abundance decays exponentially over the residence time on Earth (i.e., its terrestrial age).

The number and identity of cosmogenic nuclides in a given meteoroid depends on the time of travel from the asteroid belt to Earth (exposure time), the initial size of the meteoroid, the depth beneath the pre-atmospheric surface (shielding depth), the elemental composition of the meteoroid, and the flux of the cosmic radiation. All of these factors have left a record in the form of cosmogenic nuclides that are embedded inside the meteorites. As a result, the detection of radioactive cosmogenic nuclides has

* Corresponding author at: Department of Physics and Astronomy, The University of North Carolina at Chapel Hill, Chapel Hill, NC 27599, USA.
E-mail address: iliadis@unc.edu (C. Iliadis).

http://dx.doi.org/10.1016/j.apradiso.2014.07.001
0969-8043/

far-reaching implications for a broad range of key questions, including the dating of cosmic ray exposure ages (Herzog, 2007), the constancy of the galactic cosmic ray intensity over the past 10 million years (Wieler et al., 2011), and variations of the solar magnetic field strength over the past few hundred years (Taricco et al., 2006).

Accelerator mass spectrometry (AMS) is widely considered to be the most sensitive technique for radionuclide detection in meteorite fragments. However, the decay of a number of important radionuclides, including ^{26}Al, gives rise to the simultaneous emission of three γ-rays (a characteristic γ-ray and two 511-keV photons). Therefore, the detection of these photons using $\gamma\gamma$-coincidence spectrometry will be highly selective and may offer an interesting alternative to AMS for certain applications. Other advantages of γ-ray spectrometry compared to AMS are the non-destructive nature of the detection, a significantly simpler experimental setup, and the measurement of absolute concentrations rather than abundance ratios. Spectrometers based on $\gamma\gamma$-coincidence techniques have been described in detail before (see, for example, Cooper and Perkins, 1972; Povinec et al., 2009; Taricco et al., 2010). However, these works employed spectrometers with few segments. In the present work, we employ a 1 8-segment spectrometer and focus on optimizing the coincidence conditions (i.e., the gating on energy deposition and event multiplicity) for detecting small radionuclide concentrations in meteorites.

The sample we investigated consists of a single fragment of the Farmville meteorite, described in more detail in Section 2.1. Information regarding our coincidence spectrometer is provided in Section 2.2. Coincidence detection schemes and Monte Carlo simulations are described in Section 3. Results for ^{26}Al are discussed in Section 4. A summary and conclusions are given in Section 5.

2. Experimental procedures

2.1. Farmville meteorite

Our sample consists of a single fragment from the Farmville meteorite, which fell on December 4, 1934. In total, 56 kg of this meteorite were collected in Pitt County, North Carolina. It is classified as an Ordinary Chondrite (H4), a type of stony meteorite that contains small spherical pellets (chondrules). Most of the fragments reside in the North Carolina Museum of Natural Sciences in Raleigh, North Carolina, from where we obtained the fragment on loan. The meteorite sample had a mass of 0.569 kg and a density of 3.72 g/cm^3. Its shape was irregular, with maximum dimensions of 7.90 cm $\times$ 8.87 cm $\times$ 4.62 cm. We imaged the sample using computed tomography (CT) at UNC Hospitals and the precise shape was used in our Monte Carlo simulations (see below). The fragment is shown in Fig. 1.

Several studies have been performed regarding the elemental and isotopic composition of the Farmville meteorite. The main elemental constituents are SiO_2 (37.05%), MgO (23.29%), Fe (16.53%), FeO (10.11%) and FeS (4.99%), with the abundances provided in units of atomic % (Kallemeyn et al., 1989; Jarosewich, 1990; Kothari and Goel, 1974). Cosmogenic noble gas concentrations are reported for ^{3}He, ^{21}Ne, and ^{38}Ar, resulting in an average cosmic-ray exposure age (i.e., the time between excavation of the meteoroid from its parent body until the time of fall) of 7.3 ± 0.9 My (Eugster et al., 1993). The gas retention age (i.e., the time since the parent body cooled to sufficiently low temperatures to retain radiogenically produced noble gases) has been estimated using the ^{40}K0–^{40}Ar method, yielding a value of 4.07 ± 0.10 Gy (Eugster et al., 1993). Properties of the Farmville meteorite are summarized in Table 1. To our knowledge, no radioisotope studies have yet been performed on this meteorite.

Fig. 1. Fragment of the Farmville meteorite, which fell on December 4, 1934. It is classified as an Ordinary Chondrite (H4), a type of stony meteorite that contains small spherical pellets (chondrules). The size of the fragment amounts to about 8 cm $\times$ 9 cm $\times$ 5 cm. It was obtained on loan from the North Carolina Museum of Natural Sciences in Raleigh, North Carolina.

Table 1
Properties of the Farmville meteorite and of the fragment measured in the present work.

Property	Information/value
Classification	Ordinary stone chondrite (H4)
Date/place of fall	December 4, 1934/Pitt County, NC
Total recovered mass	56 kg
Chemical composition[a]	SiO_2, MgO, Fe, FeO, FeS, …
Cosmic-ray exposure age[b]	7.3 ± 0.9 My
Gas retention age[c]	4.07 ± 0.10 Gy
Fragment mass[d]	0.569 kg
Fragment volume[d]	152.5 cm^3
Fragment density[d]	3.73 g/cm^3

[a] Kallemeyn et al. (1989), Jarosewich (1990), and Kothari and Goel (1974).
[b] Average of ^{3}He, ^{21}Ne and ^{38}Ar exposure ages (Eugster et al., 1993).
[c] Estimate using ^{40}K–^{40}Ar (Eugster et al., 1993).
[d] Measured in present work; and used for present Monte Carlo detector simulations (see later).

2.2. Spectrometer

The coincidence spectrometer employed in the present work is the main detection device used for nuclear reaction measurements at the Laboratory for Experimental Nuclear Astrophysics (LENA), which is part of the Triangle Universities Nuclear Laboratory (Cesaratto et al., 2010). In most nuclear astrophysics studies, several γ-rays are emitted simultaneously, with a total energy near or in excess of 10 MeV. By measuring these γ-rays in coincidence, and accepting only coincidence events with a total energy *above* about 4 MeV, the environmental background is reduced by several orders of magnitude. The performance of this spectrometer for total energies above 4 MeV has been described in detail in Longland et al. (2006). Note that there is no need for any passive shielding at these higher energies, because the coincidence requirement is very selective. In the present work, we were interested in exploring how useful this device can be for radioisotope studies in meteorites. While we previously rejected total γ-ray energies *below* 4 MeV, this energy region now becomes the