Graphical Abstract

Low-background gamma spectrometry and neutron diffraction in the study of stony meteorites

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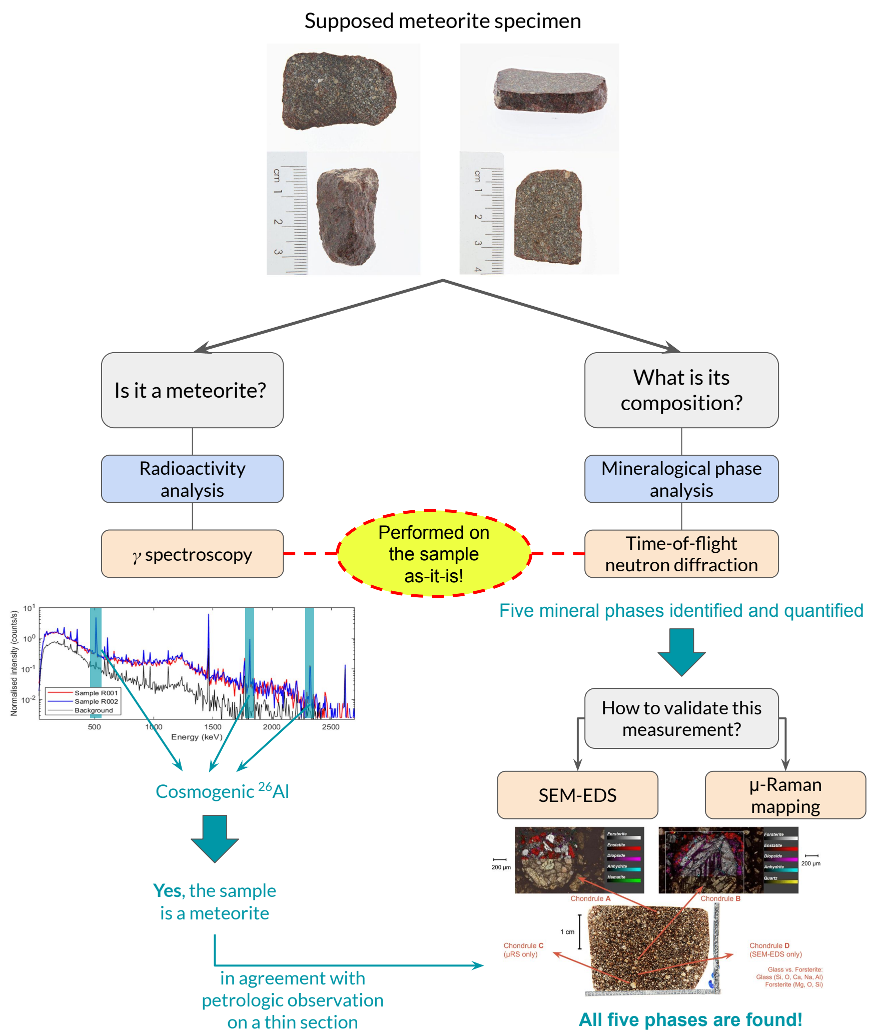

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Highlights

Low-background gamma spectrometry and neutron diffraction in the study of stony meteorites

Riccardo Rossini[2], Massimiliano Clemenza, Daniela Di Martino, Matthias Laubenstein, Antonella Scherillo, Maya Musa, Maria Pia Riccardi, Giuseppe Gorini

- Non-destructive characterisation of meteorites;
- Gamma spectrometry to identify meteorites by analysing their gamma ray emission;
- Time-of-Flight Neutron Diffraction to quantify mineral phases in meteorites.

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Low-background gamma spectrometry and neutron diffraction in the study of stony meteorites

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Abstract

Non-destructive characterisation of meteorites is here performed on a stony meteorite. The identification of the sample is performed by low-background γ-ray spectrometry in order to determine the presence of certain cosmogenic radionuclides, whereas a mineralogical phase quantitative analysis is carried out by Time-of-Flight Neutron Diffraction (ToF-ND) on the sample as-it-is. The protocol is then validated by applying micro-Raman Spectroscopy (µRS) and Energy Dispersive X-ray Spectroscopy (EDS). This paper is focused on γ-ray spectrometry, proving the meteoric origin of the sample, and it also presents some preliminary results of ToF-ND.

Keywords: meteorite, low-background gamma spectrometry, time-of-flight neutron diffraction, non-destructive
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1. Introduction

An identification and characterisation study for stony meteorites is here proposed in order to extract information from such samples with a completely

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non-destructive combination of techniques.

In particular, two massive samples (called R001 and R002) and a thin section (R003) of a supposed meteorite were received by a private collection they belonged for more than 30 years. The samples can be seen in Figure 1, the masses of R001 and R002 are 12.61(36) g and 14.65(14) g, respectively.

Figure 1: The two massive samples (R001 and R002) and the thin section (R003) of meteorite studied in this work.

The study of the sample γ-ray emission was applied in order to analyse the presence of fossil and cosmogenic radionuclides, and in particular to prove that the sample is a meteorite. This is a non-destructive method to perform meteorite identification, alternative to the traditional petrological observations which require the extraction of a thin section. The main radioactive radionuclides of cosmogenic origin which can be found in meteorites are the following[1]: ^{26}Al ($t_{1/2} = 7.6 \cdot 10^5$ y), ^{60}Co ($t_{1/2} = 5.27$ y), ^{22}Na ($t_{1/2} = 2.6$ y), ^{54}Mn ($t_{1/2} = 312$ d), ^{46}Sc ($t_{1/2} = 84$ d) and ^{48}V ($t_{1/2} = 16$ d). As our sample fall happened at least 30 years ago, we expected that only ^{26}Al could be still present, with an expected low activity. As a consequence, low-background gamma spectrometry was performed at the STELLA facility in the underground LNGS laboratories[2] in Italy.

After this measurement, the whole samples R001 and R002 were studied with Time-of-Flight Neutron Diffraction (ToF-ND) at the INES beamline[3] within the ISIS Neutron and Muon Source (UK) in order to understand their mineral composition in a non-destructive way. In fact, the depth of penetration of neutrons (few centimeters) in samples having rock-like composition allows to perform this measurement on whole samples. Preliminary results of the application of this technique are here presented.

In parallel, two surface techniques were applied on the thin section R003: microRaman Spectroscopy (μRS) and Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy (SEM-EDS) in order to verify the conclusions made with ToF-ND.

2. Low-background gamma spectrometry

The *GeMi* p-type coaxial High-Purity Germanium (HPGe) detector at the STELLA facility was used on this purpose. The p-type and n-type doping of germanium in the p-i-n junction of this detector are obtained by diffusing lithium and boron, respectively. Monte Carlo (MC) simulation on this detector were made using the Arby interface to Geant4[4]. The first panel in Figure 2 shows the simulation geometry for sample R001 on the detector. The nominal thickness of the Li-doped germanium dead layer is 0.6 mm, but MC simulation optimisation enabled to calculate the actual equivalent dead layer as 1.5 mm, which takes into account both the diffusion of lithium in germanium over the years and other uncertainties in the simulation process. The second panel of Figure 2 shows the comparison by MC simulation of this effect on the 1808 keV ^{26}Al peak with the nominal and estimated values of the dead layer.

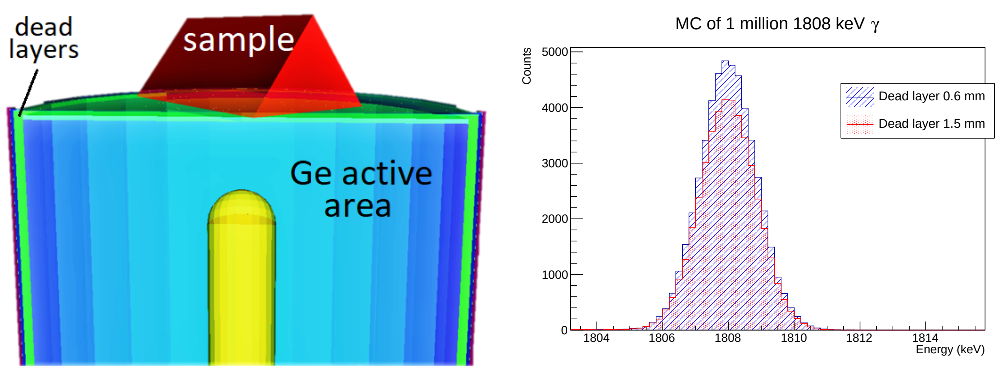

Figure 2: Left: MC simulation geometry for sample on the detector (red: sample, green: Li-doped germanium and other external dead layers, yellow: B-doped germanium, light blue: pure germanium). Right: simulated effect of the dead layer enhancement, due to the diffusion of lithium in the pure germanium, on the 1808 keV peak from ^{26}Al (left).

The two samples were measured for 16 days each, whereas the background measurement took 30 days. The spectra have been analysed by Gauss-fitting all peaks exceeding three standard deviations from the baseline and efficiencies were estimated by MC simulation (assuming the shape of a triangular prism for R001 and a parallelepiped for R002). The spectra are reported with radionuclide attributions in Figure 3.

The uncertainty budget on the final estimations of the activity has been characterised by executing MC simulations changing the sample size and composition according to the corresponding uncertainties. In particular, the leading contribution is given by the irregular shape of the sample, which introduces an uncertainty on the efficiencies around 10% in R001 and 15% in R002. Minor contributions ($< 1\%$ in total) come from the uncertainty on the sample composition and the statistics in the experimental data and MC simulations. Finally, the specific activity is calculated as:

$$A = \frac{s - b}{m \ \mathtt{BR}\varepsilon} \tag{1}$$

whereas its uncertainty:

$$\frac{\sigma_A}{A} = \sqrt{\left(\frac{\sqrt{\sigma_s^2 + \sigma_b^2}}{s - b}\right)^2 + \left(\frac{\sigma_{\mathtt{BR}\varepsilon}}{\mathtt{BR}\varepsilon}\right)^2 + \left(\frac{\sigma_m}{m}\right)^2} \tag{2}$$

where s is the sample count rate under the selected peak and b the background one, $\mathtt{BR}$ the channel branching ratio, m the sample mass, ε the efficiency and σ the uncertainty on the subscripted variable. All MC simulations used for quantification were carried out simulating whole radionuclide decays. As a consequence, the efficiencies coming from the MC simulation already contain $\mathtt{BR}$'s, and so do their uncertainties. This is the reason why the uncertainty on the product $\mathtt{BR}\varepsilon$ is reported as a single contribution in Equation 2. All activity values are obtained by a weighted average (with standard uncertainty) among all peaks corresponding to a certain radionuclide and between the two samples.

The estimated specific activity of fossil ^{40}K is (20 ± 2) Bq/kg. The results of the quantification of fossil radionuclides from ^{232}Th and ^{238}U radioactive chains are summarised in Figure 4. The ^{232}Th chain appears at secular equilibrium, even though a systematic discrepancy is visible between the two samples, due to the simplification of the sample shapes in MC simulation. Conversely, the ^{238}U chain is not at secular equilibrium, and in particular it appears to be broken at the level of ^{226}Rn (which is a noble gas having 3.8 d half-live, which strongly enables its migration away from the sample).

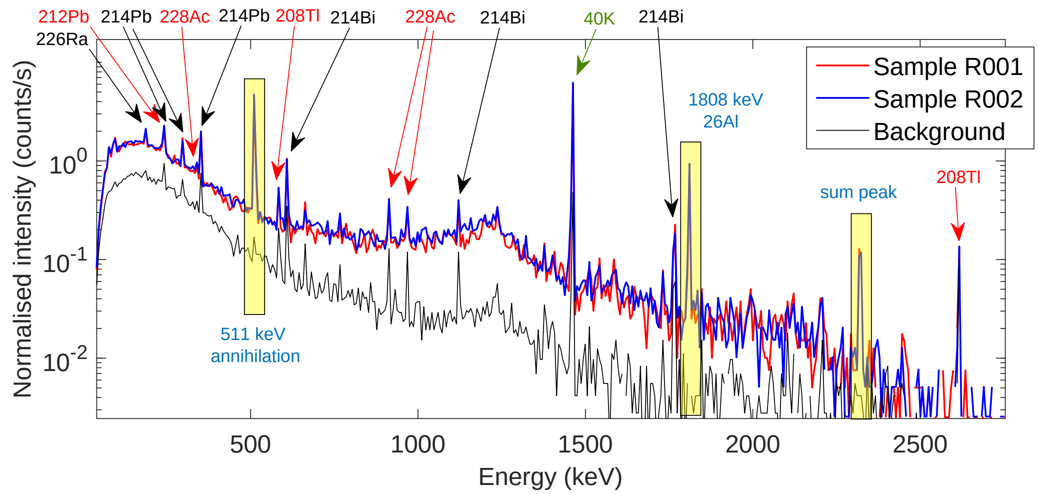

Figure 3: Gamma spectra from samples R001, R002 and background. The peaks due to fossil radioactivity from ^{40}K (green arrow), ^{232}Th (red arrows) and ^{238}U (black arrows) are indicated. Yellow boxes mark the three peaks due to the decay of cosmogenic ^{26}Al.

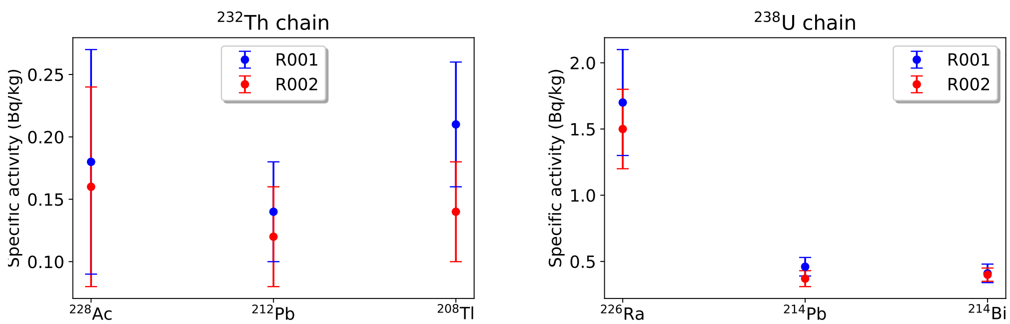

Figure 4: Weighted-average activities from the γ-emitting radionuclides in the ^{232}Th & ^{238}U chains, where the radionuclides on the x-axis are ordered as in the radioactive chains. The ^{232}Th appears to be at secular equilibrium, whereas the ^{238}U chain is broken at ^{226}Rn.

The presence of ^{26}Al is a clear marker of the fact that the sample is a meteorite. ^{26}Al is a β^+ emitter, which decays on a 1808 keV excited state of ^{26}Mg. As a consequence, the observables of its presence are the 511 keV annihilation peak, the 1808 keV de-excitation peak of ^{26}Mg and their 2319 keV sum peak. Its quantification was made on the latter two peaks: 1808 keV and 2319 keV. The obtained activity is (9.7 ± 1.3) mBq for R001 and (9.9 ± 1.2) mBq for R002, corresponding to an average specific activity of (0.69 ± 0.07) Bq/kg, which is only consistent with a meteoric origin of the studied samples.

3. ToF-ND

Time-of-Flight Neutron Diffraction measurements were performed on samples R001 and R002 at the INES diffractometer of the ISIS Neutron and Muon Source, using thermal neutrons. As a preliminary result, the presence of five different mineral phases was observed: Forsterite (silicate, Mg_2SiO_4), Enstatite (silicate, $MgSiO_3$) and three altered phases of iron: Magnetite (Fe_3O_4), Troilite (FeS) and traces of Kamacite (Fe-Ni alloy up to 95:5 ratio). These results were obtained by preliminary qualitative analysis, but detailed quantitative results obtained by means of Rietveld Refinement are currently being obtained and will be published by the end of the year. This analysis is being performed with the General Structure Analysis System (GSAS)[5].

4. Validation measurements

A full thin section study on sample R003 based on μRS and SEM-EDS has been performed[6]. First of all, petrological observations at sub-millimetric level reveal that the structure of the sample is made of irregular silicate-based fragments called *chondrules*, immersed in a glass containing also iron-based formations. This structure is consistent with the classification of the samples as meteorite fragments, particularly from a chondrite[7]. The analysis included four μRS and SEM-EDS maps, covering four chondrules and several intra-glass structures.

There is clear evidence of the presence in the chondrules of the thin section R003 of silicates such as Forsterite and Enstatite, which appear to be by far the main constituents of chondrules. An extensive EDS study on the iron-based formations returned compositions compatible with the altered phases

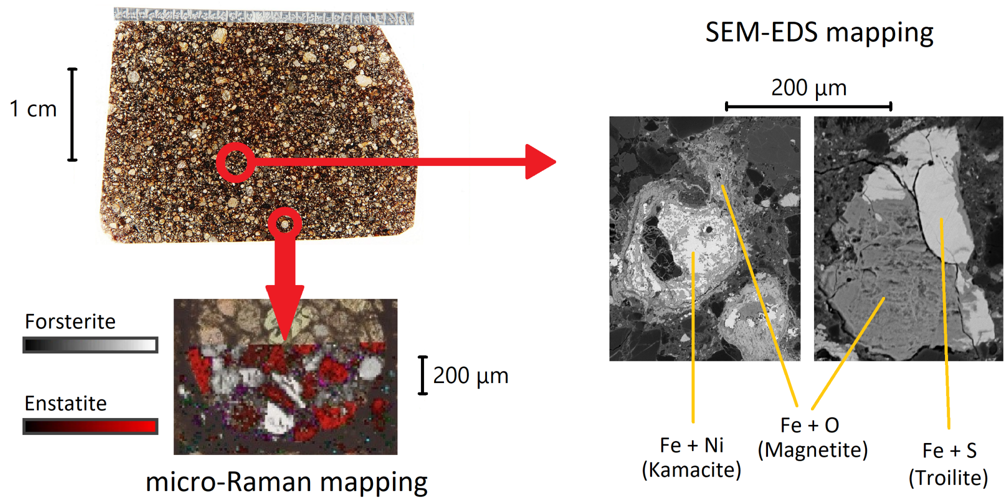

Figure 5: Validation measurements with µRS and SEM-EDS are consistent with the presence of the five minerals identified with ToF-ND. Furthermore, the petrological observation tells that the sample is effectively a meteorite, in accordance with the results of gamma spectrometry.

of iron identified with ToF-ND, despite being Raman-inactive and therefore not visible with µRS. This validation study is resumed in Figure 5.

As a consequence, all five mineral phases identified by means of non-destructive ToF-ND on the sample as-it-is are also visible with consolidated techniques on a thin section.

5. Conclusion

Gamma spectrometry allowed the identification of the sample as a meteorite by verifying the presence of ^{26}Al, but it also enabled the study of the fossil radioactivity due to ^{40}K, ^{232}Th and ^{238}U.

Time-of-Flight Neutron Diffraction made it possible to identify, in a totally non-destructive way, the mineral phases present in the samples, whose presence was verified on a thin section by means of consolidated surface techniques µRS and SEM-EDS.

This work goes in the direction of developing a fully non-destructive analysis protocol for meteorites, which should be considered in order to start studying such samples without provoking damage.

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