Acta Physica Universitatis Comenianae
Volume LIV (2019) 109–114

Non-destructive Determination of Elemental Composition of Meteorite Samples Using PIXE Technique*

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Abstract: The non-destructive approach to the sample treatment during the analytical process is one of the crucial advantages of the PIXE (Particle Induced X-ray Emission) technique. Rare and precious environmental samples can be analysed non-destructively in order to evaluate the concentration of individual elements presented in the specimen. A non-destructive analysis of two meteorite samples was carried out using 3 MeV protons incident in a narrow ion beam (1.5 mm diam.). GUPIXWIN software package was used for spectra evaluation. Concentrations of several elements (Fe, Ni, Cu and Zn) were determined and surface distribution maps were constructed.

Keywords: PIXE, meteorite, proton beam, distribution maps, relative concentration.

1. Introduction

The PIXE technique is specialized on trace elements contents determination. It can be used for almost all elements contents evaluation depending on the sample structure. Important aspect is that the number of ions which induce the emission of detected radiation has to be treated carefully. The PIXE technique is considered as non-destructive material analytical technique. This statement is valid within standard conditions. Obviously, if the intensity and energy of the incident beam exceed certain level then the analysed material can be damaged even destroyed, especially in the case of thin samples. For low beam intensities (represented by measured current in Faraday cup) at nanoamperes levels and short measurement times, e.g. 10 minutes, the effects caused by incident particles are usually negligible. Details about PIXE technique can be found in [1].

The aim of the present work was to analyse two meteorite samples using the PIXE technique, namely the Rumanová chondrit (Slovakia) and the Canyon Diablo iron meteorite (Arizona, US).

*Dedicated to Professor Peter Prešnajder on the occasion of his 70th birthday

2. Materials and methods

1. Meteorite samples

The Rumanová meteorite finding is described in [2]. This finding comprised of one 4.3 kg piece of stone with a density of 3.53 g/cm^3, and dimensions of 18.5×14.0×12.5 cm^3. The meteorite showed an evident degree of weathering, as well as visible chondrules of different size and structure. The meteorite was classified as H5 chondrite with dominant minerals such as enstatite, olivine, kamacite, taenite and troilite [3]. It was previously analysed using Mössbauer spectroscopy [3, 4] and gamma-ray spectrometry [5]. The situation with the Canyon Diablo Arizona meteorite sample is more complex since the original material was enormous comparing to Rumanová meteorite. The Barringer Crater (named in the honour of Daniel Barringer, who was first to suggest that it was produced by a meteorite impact) is approximately 1200 m wide, 170 m deep and its age is 49 000 years [10]. Research in the last century described various aspects, e.g. [7–9], but relatively recent work [10] concludes that the meteor crater was formed by a high-velocity impact of a tight projectile swarm. The authors claimed that a similar crater could be formed as well as at lower impact velocity, but this would mean that much more solid projectile ejecta than has been observed. We analysed just a small fragment (mass of 14 g) using the PIXE technique. The non-destructive analysis should provide information about surface distribution of elements presented in the sample.

2. Sample preparation

The samples, their dimensions and detailed surfaces are shown in Figs. 1 and 2. The Rumanová meteorite sample is represented by a cut block from the original material, while the Arizone meteorite is represented by a 14 g fragment.

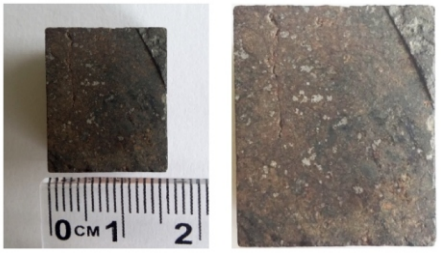

Fig. 1. The Rumanová meteorite sample.

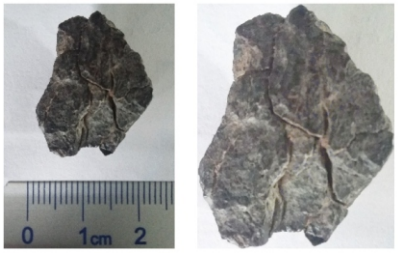

Fig. 2. The Arizona meteorite sample.

In order to avoid contamination during handling the samples a special approach to the sample attachment to the sample holder was involved. An easily mouldable pad made of aluminium foil was attached to the sample holder using conductive double coated adhesive carbon tape. Subsequently, the sample was slightly pressed into this pad. Such attached samples were installed into the PIXE chamber for analysis (Fig. 3).

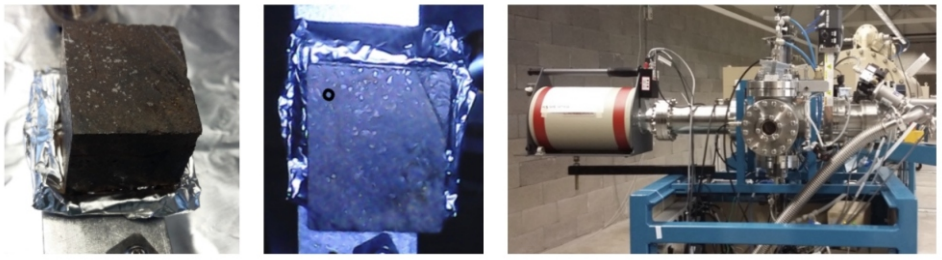

Fig. 3. The sample attachment to the sample holder, position of beam inside the PIXE chamber and the PIXE chamber with BEGe detector.

3. The PIXE analysis

The PIXE technique used at the CENTA laboratory has already been described in [11] therefore we shall present here only few comments. The 3 MeV proton beam was used for analysis. The beam size at the sample surface was approx. 1.5 mm. The beam was focused using a magnetic quadrupole and collimator slits to achieve this diameter. During the measurements an average beam current was approx. 100 pA. The total collected charge was 50 nC (same for all analysed spots) and the ORTEC digital current integrator was used for its measurement. Emission of the secondary electrons from the sample surface was suppressed using a special electrode kept at the negative potential of 400 V. The emitted X-rays were detected by the Canberra BEGe detector (model BE2825). This detector is covering the energy range from 3 keV to 3 MeV, with energy resolution of 390 eV for 5.9 keV (^{55}Fe) and 1.8 keV for 1332 keV (^{60}Co). Overall 30 positions (spots) in a mesh with 3 mm step were analysed across the Rumanová meteorite surface. The detail of the analysed surface is shown in Fig. 1. The brightness of the original photography (Fig. 1 left) was slightly adjusted for better visibility of surface inhomogeneities. A black circle in a photography taken from the chamber inside (Fig. 3 middle) represents beam size during the measurements. The spectra were processed using GUPIXWIN software package. Using current detection spectrometer relative concentrations of a few elements (Fe, Ni, Cu and Zn) could be evaluated.

The second sample – the Arizona meteorite – was measured in a similar way, but the analysis was more complicated as the surface had an irregular shape. The photography of the Arizona sample surface (Fig. 2 right) was adjusted in the same manner as for the Rumanová sample.

3. Results and discussion

Each of 30 measured spots on the Rumanová meteorite sample were analysed individually using the same parameters in GUPIXWIN. Spectra were fitted and information about individual peak areas were obtained. The maximum area for each element (out of the 30 analysed spectra) was pronounced as 100% concentration and all other spots with lower area values were normalized to this maximum. Such relative concentrations of iron, nickel, copper and zinc were calculated and distribution maps were created (Fig. 4).

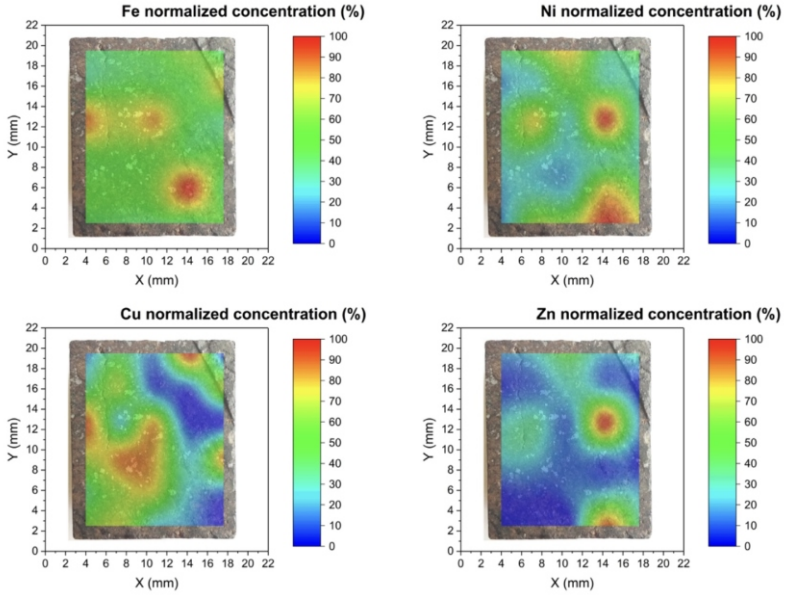

Fig. 4. Normalized concentration distribution maps of the Rumanová meteorite sample.

Measured spectra observed with maximum concentrations (together with the GUPIXWIN fit) are shown in Fig. 5.

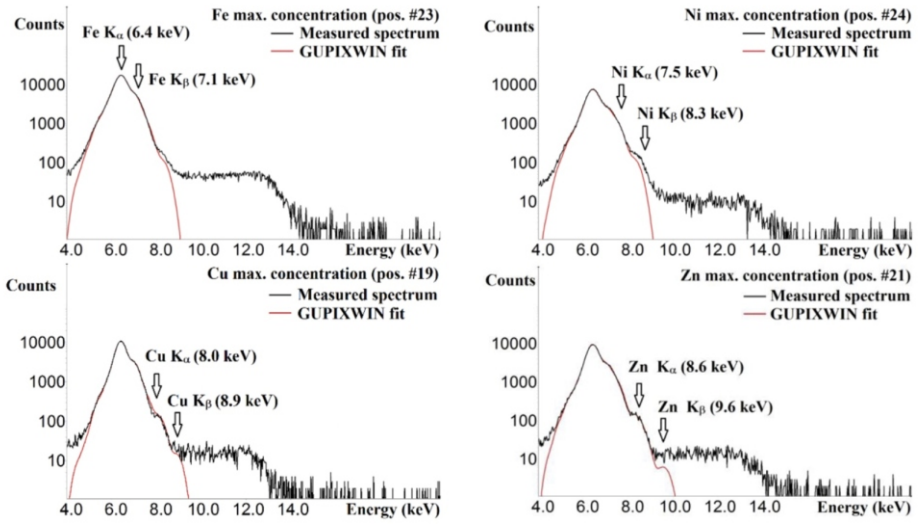

Fig. 5. Measured spectra and GUPIXWIN fits of positions with maximum concentrations of Fe, Ni, Cu and Zn in Rumanová meteorite sample. Position of X-ray lines are denoted by arrows; the areas for normalized concentration distribution maps were determined using the GUPIXWIN fit.

For all 30 positions such spectra were analysed, and areas were used for normalized concentration calculations. The presence of any other elements was plausible since the GUPIXWIN fitting procedure was affected by high uncertainties. Iron was measured in all analysed spots with the lowest value at approximately 40%. Nickel was measured in all analysed spots; the lowest value was approximately 16%. Copper exhibited lower concentrations and in 3 spots no copper was observed. The concentration of zinc was low (lower than copper). Again, in 3 spots (but different from Cu) no Zn was observed. The distribution maps show surprisingly high inhomogeneities in the distribution of elements in the Rumanová meteorite which should be taken into account when other non-destructive analytical methods are used for determination of elemental composition (e.g. microscopic methods).

The situation with the Canyon Diablo meteorite sample is more complex. Since the surface of the sample is very rough and the composition is different, i.e., the iron content is much higher comparing with other elements, the relative elemental composition could not be calculated. Also current detector resolution (approx. 400 eV at 6.4 keV iron K line) was not helping to improve this problem. Measured spectrum of the Canyon Diablo meteorite sample is shown in Fig. 6.

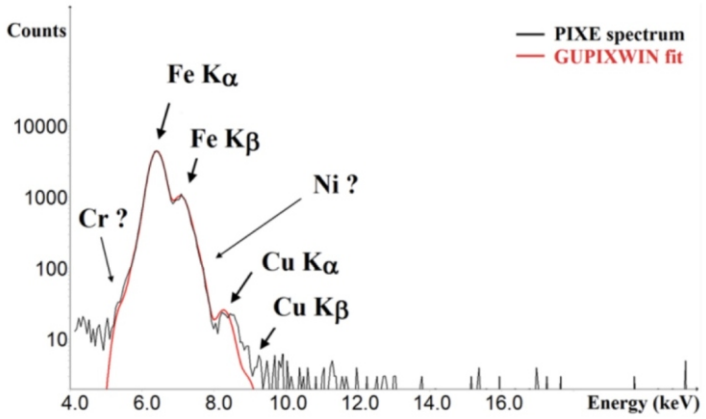

Fig. 6. Measured PIXE spectrum and GUPIXWIN fit for the Canyon Diablo meteorite sample. Iron and copper could be evaluated, but presence of other elements (e.g. chromium and nickel) was questionable because of the detector resolution and high iron content.

The GUPIXWIN fitting procedure managed to fit iron and copper with low uncertainties, but other elements were not found, or the fitted area exhibited high uncertainty thus the presence of individual elements is questionable. The chromium and nickel are the most probable candidates, but also manganese (K X-ray line of 5.9 keV lies between chromium and iron) could be present in this sample. Nickel (K X-ray line of 7.5 keV) and zinc (K X-ray line of 8.6 keV) could be also present in the sample. For such analysis a better spectrometer resolution is needed. Hopefully, a new SDD detector (resolution of 140 eV) will be installed into the PIXE chamber (planned during the summer 2019) and more precise analysis will be possibly performed.

4. Conclusions

The obtained results of the analysis of the meteorite Rumanová with PIXE technique are promising. The next step will be to determine the absolute concentrations of observed elements (in ppm or mg/cm^2) as calibration of the spectrometer is needed for thick samples. Measurements of geological standard materials are planned, and results should provide appropriate calibration parameters in order to determine the absolute abundance of elements in the sample. We have demonstrated that the described non-destructive PIXE approach can be used for precious samples analyses. The distribution maps showed, however, high inhomogeneities in the distribution of elements in the Rumanová meteorite which should be taken into account when other non-destructive analytical methods are used for determination of elemental composition (e.g. microscopic methods).

Acknowledgements

The authors are acknowledging support provided by the EU Research and Development Operational Program funded by the ERDF (projects # 26240120012, 26240120026 and 26240220004), and from the International Atomic Energy Agency (project # SLR/1001).

This study has also been supported by the Slovak Science Grant Agency under the contract # VEGA 1/0891/17 and by the Slovak Research and Development Agency under the contract # APVV-16-0148.

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