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Instrumental photon and proton activation analysis of some elements in stony meteorites

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Chapter I

Introduction

I.1 Introduction into activation analysis

I.1.1 *General principles*

Although the first activation analysis experiment was carried out already in 1936 (*de Hevesy, 1936*), it was not until after the second world war that this technique became more extensively used. This can easily be explained by the fact that since then a large number of reactors and accelerators has become available. Nowadays activation analysis is used in a great variety of disciplines, where its special features appear to full advantage.

As a consequence a vast amount of literature on the subject has appeared, including handbooks on the techniques (*Bowen, 1963; Lyon, 1964; Lenihan, 1965*), lists of nuclear data of interest (*Koch, 1960*) and bibliographies on applications (*Euratom, 1964; Lutz, 1970-a, -b, 1971-a*). Moreover the proceedings of several international conferences on the developments in and the applications of activation analysis (*Texas, 1965; Salzburg, 1965; Amsterdam, 1967; Liège, 1968; Gaithersburg, 1969*) provide more detailed information as do publications in scientific magazines.

The principle of activation analysis is rather simple. If a sample is irradiated with nuclear particles, nuclear reactions can occur, some of which may lead to unstable nuclei as reaction products. By determination of the

type, the energy and the half-life of the emitted radioactivity, these radioisotopes may be identified. If necessary, a chemical separation is used to isolate the element of the required radioactive product. From this the parent element, which is frequently identical with that of the radioisotope, is deduced. Moreover, if the intensity of the radiation, emitted by the radioactive product, can be measured, this quantity constitutes a measure of the amount of parent nuclides present in the sample.

The first stage in an activation analysis is the activation (sometimes preceded by some special preparation of the sample). This can be achieved by bombardment with neutrons (thermal or fast), charged particles or high-energy photons. The choice of irradiation will depend on several circumstances, such as the element to be determined, the matrix, possible interferences, needed sensitivity and accuracy, and of course the available irradiation facilities.

In general most activations are performed with thermal neutrons in reactors. The large cross sections for (n,γ) reactions and the high flux densities make possible determinations at very low concentration levels. However, this type of activation does not always offer a solution, as for some nuclei the cross sections for (n,γ) reactions are very low (especially for elements with $Z < 10$), while in some other cases the activity of the product nuclides can not be measured conveniently.

Fast neutrons, in most cases of 14 MeV produced by T+D generators, find wide applications too. As in general the cross sections for fast neutron reactions are small compared to those for thermal neutrons, and the fluxes obtainable are smaller than those of thermal neutrons, the detection limits will be appreciably higher.

Charged particles, mostly from cyclotrons or van der Graaff-accelerators, offer another possibility. Especially in the determination of light elements their application has proved very successful, as very low detection limits can be reached (e.g. *Nozaki, 1970*).

High-energy photons, occurring in the bremsstrahlen produced by electrons from an electron accelerator (lineac, betatron or microtron) can be used to induce reactions, leading to product nuclides which in some cases are the same as those, produced by fast neutron reactions. As the flux densities available greatly surpass those of fast neutrons, the detection limits will be lower in those cases. Recently a general survey on this technique has been published (*Lutz, 1971-b*).

Between activation and activity measurement a chemical treatment of the sample is frequently required to remove activities interfering with the analysis. However, in recent years the use of high resolution semiconductor gammaspectrometers has in many cases provided means to avoid these separations. For this technique the name "instrumental activation analysis" is used.

The last step, the measurement of the activity, is mostly performed by gamma-ray spectrometry. In some cases beta-counting is applied, but, as a chemical separation is nearly always required for this purpose to obtain a counting sample of sufficient radiochemical purity, this method is only used if gamma-ray spectrometry is not possible.

A special kind of activation analysis is the one where the number of nuclear reactions is recorded, by measurement of the product particles emitted during the irradiation. This technique has not yet found many applications, although the introduction of track registration in dielectrics is making it attractive for the determination of small amounts of fissile elements or nuclides.

Activation analysis has some features which make it a very suitable technique for certain special purposes. In the first place there are the low detection limits, especially for thermal neutron activation analysis. In many cases this provides a means for quantitative determinations at concentration levels where other analytical techniques are not applicable. A second feature is the possibility of non-destructive analysis, a property which it shares with only a limited number of other techniques (e.g. X-ray fluorescence spectrometry, X-ray diffractometry). This makes activation analysis a powerful tool for the investigation of very valuable or irreplaceable objects (e.g. products of art, archaeological finds, etc.).

I.1.2 Applications of activation analysis in geochemistry and cosmochemistry

For geochemical and cosmochemical studies activation analysis has become an essential technique, especially for the analysis at levels of very low concentrations. This could not be illustrated better than by the fact that recently a special conference was held on the applications of activation analysis in these disciplines (*Kjeller, 1971*).

For the determination of trace elements the thermal neutron activation offers the best solution for most elements. Many elements can be determined simultaneously by one irradiation when use is made of specially developed methods for the chemical separation of elements into groups (*Morrison, 1969*), as well as of instrumental analysis (*Gordon, 1968*).

Fast neutron activation has been applied extensively too, especially for the determination of major constituents (e.g. *Morgan, 1971; Kuykendall, 1971*), where it provides

a fast analysis of several elements, often simultaneously.

The role of other kinds of activation in these disciplines has been rather limited. Photon activation was applied in a few cases, which are listed in the recently published survey on photon activation by *Lutz (1971-b)*.

With respect to charged particles, their applications in this field are limited to the Russian work with α -sources for the determination of Be in ores (*Plaksin, 1960*), and the determination of Pb in meteorites with α -particles from an accelerator (*Cobb, 1960, 1964*).

I.2 Meteorites

"A meteorite is a solid body which has arrived on Earth from outer space. Its size can range from microscopic to a mass of many tons" (*Mason, 1962*).

Between Mars and Jupiter there is located the asteroid belt. Here the asteroids travel along their orbit around the sun, and by collisions of one with another, pieces of them are thrown out of the orbit. If such a meteoroid heads for the Earth and survives the journey through our atmosphere, a new meteorite has fallen.

The main constituents of meteorites are a nickel-iron alloy and the silicate minerals olivine and orthopyroxene (both Fe-Mg-silicates). According to the contribution of these constituents to the total composition, a meteorite can be classified in the following scheme (*Prior, 1920; Mason, 1962*):

1. Siderites or irons. Meteorites of this class consist mainly of nickel-iron, with small amounts of accessory minerals. A subdivision is made, reflecting on the crystal structure and the Ni content.
2. Siderolites or stony-irons. Here the amounts of nickel-iron and of silicate minerals are about equal. Of the

subclasses two types are the most important:

a. mesosiderites (or pyroxene-plagioclase stony-irons), in which the metallic constituent is present as small or large grains, more or less connected with another, while the interstices are filled with silicate minerals;

b. pallasites (or olivine stony-irons), in which the metallic phase forms a definite reticulum, of which the meshes are filled with silicate minerals.

3. Aerolites or stony meteorites. These meteorites have for their main constituent silicates, generally with some nickel-iron. Their subclassification is rather complicated:

a. chondrites. Stones of this class contain the so called "chondri" or "chondrules", i.e. spheroidal aggregates of the silicate minerals olivine and/or pyroxene, usually about 1 mm in diameter. According to increasing contents of oxydized iron and decreasing free iron contents they are subdivided into enstatite, olivine-bronzite, olivine-hypersthene, olivine-pigeonite and carbonaceous chondrites.

b. achondrites. Any stone which is no chondrite belongs to this class. Therefore they constitute a rather inhomogeneous population. They are also much less numerous than the chondrites. Their subclassification is:

i. Ca-poor achondrites, subdivided into enstatite, hypersthene, olivine and olivine-pigeonite achondrites;

ii. Ca-rich achondrites, subdivided into augite, diopside-olivine and pyroxene-plagioclase achondrites.

With respect to the origin of meteorites, many theories exist. They certainly belong to our solar system, and therefore it is generally accepted that their original material was the same as that from which the rest of our solar system was formed. This makes the research on meteorites so interesting, because the Earth is a geoche-

mically disturbed system, while the meteoritic parent system has broken up before it could be spoiled. The parent body and the history leading to the eventual formation of the meteorites is the subject of many theories. Some of these (e.g. *Ringwood, 1961*) assume a broken up planet as parent body. Others (*Urey, 1959*) suppose two successive generations of parent bodies, primary and secondary; while still others (*Fish, 1960*) assume the formation of the meteorites to have taken place in bodies of asteroidal size. As far as the results of this work have to do with these theories, they will be summarized in the discussion (chapter IV). A survey of the different theories is given by *Mason (1962)*.

I.3 Introduction to this work.

As meteorites are subjects of great scientific value, non-destructive analytical techniques offer advantages in the research on this material. Moreover it is possible to perform different kinds of analysis on the same sample.

Many kinds of analytical techniques have been applied in this field. Conventional wet chemistry has been applied in many cases (e.g. *Wik, 1956*). However, instrumental techniques, like X-ray fluorescence spectrometry (e.g. *Nichiporuk, 1967*) and, above all, neutron activation analysis, have provided many results, especially on trace element concentrations. Most activation analysis results were obtained with thermal neutron activation, some with fast neutron activation.

This work deals with the, instrumental, determination of some elements by activation with high-energy photons and with charged particles. The techniques have been developed and applied to a large number of stony meteorites and some stony-irons, almost all of which belong to the collection present at IKO.