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IN-BEAM NEUTRON ACTIVATION ANALYSIS OF STAINLESS STEEL AND IRON ORE

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Neutron capture gamma-ray activation analysis technique has been used for the non-destructive analysis of Fe, Cr, Ni, Mn and B in stainless steel and iron ore samples. It has been shown that the use of low energy capture gamma-rays, ranging from 0.2 to 1.4 MeV, helps considerably in reducing the time of analysis. The problem of congestion of peaks due to compton continuum and the double and single escape peaks in this region of the spectrum has been overcome by using a Ge(Li) detector in conjunction with a 15.25 cm thick bifurcated NaI(Tl) annulus in anticompton mode. The results obtained by this technique have been compared with those of the chemical analysis.

Introduction

Neutron activation analysis due to its inherent sensitivity and accuracy has become an important analytical technique. In this technique[1] the sample to be analyzed is irradiated with neutrons to produce radioisotopes and the amount of the desired element is determined by measuring the resultant delayed activity. The basic requirement for an element to be analyzed by this technique is that the neutron irradiation of the element should result in the formation of a reasonable amount of the radioactive nuclide which can be conveniently measured. Certain elements such as Fe, Ti and Cr have relatively large neutron capture cross-sections but are still not very suitable for radio-activation analysis as the product nuclides are either stable or have inconvenient half-lives. Further the neutron irradiation of certain other elements such as S, P and Tl results in the production of pure beta-emitting isotopes, which are unsuitable for non-destructive analysis.

Several workers have mentioned the use of neutron capture gamma-rays as an alternative technique for non-destructive elemental analysis.[2-26] Since this technique

involves the simultaneous irradiation and measurement of neutron capture prompt gamma-rays, it is independent of the nuclear characteristics of the product nuclides. Neutron capture gamma-ray activation analysis, however, has not received much attention and is still in the developmental stage. Very little work has been done on the utilization of low energy (<5 MeV) gamma-rays for analysis despite the fact that it would help to reduce the analysis time.[12,16,24] (The detection efficiencies are higher for low energy gamma-rays.[24]) The lack of interest in the utilization of low energy gamma-rays is apparently due to the complex nature of the spectrum. In this region of the spectrum there is a general congestion of peaks due to the presence of compton edges, electronic noise and the background caused by the inelastic scattering of fast neutrons and gamma-rays from the neutron source. In order to avoid the complicated part of the spectrum, the lower cut-off limit of 5.0 MeV was chosen by some workers.[26,30]

The present study has been undertaken to demonstrate the usefulness of lower energy gamma-rays (from 0.2 to 1.4 MeV) for neutron capture gamma-ray activation analysis. In this study stainless steel and iron ore samples have been used. The full energy peaks, in the region of 0.1 to 1.4 MeV, are expected to be more prominent than the double or single escape peaks as the escape peaks here will arise only from gamma-rays of energy less than 2.4 MeV while the pair production cross-section below this energy is only a small fraction of the total cross-section. The compton continuum and the escape peak intensities can be further reduced by using the Ge(Li) detector in conjunction with a NaI(Tl) annulus in anticompton mode.

Experimental

The neutron capture gamma-ray facility installed at the 5 MW swimming pool research reactor at PINSTECH has been used in the present study. Full details of the system have been reported elsewhere.[28] A brief description of the experimental arrangement is given below.

Experimental set-up

A collimated beam of neutrons 2 cm in diameter has been extracted out of the through tube of the reactor. A 10 cm thick bismuth single crystal placed in the path of the beam provides preferential discrimination against fast and empithermal neutrons and also helps to reduce the intense low energy gamma-ray background from the reactor. The thermal neutron flux at the target position is $1.2 \cdot 10^7$ $n \cdot cm^{-2} \cdot sec^{-1}$ with cadmium ratio of 18. A double walled tube of internal diameter 4.1 cm with the spacing between the two walls (9.5 mm) filled with lithium

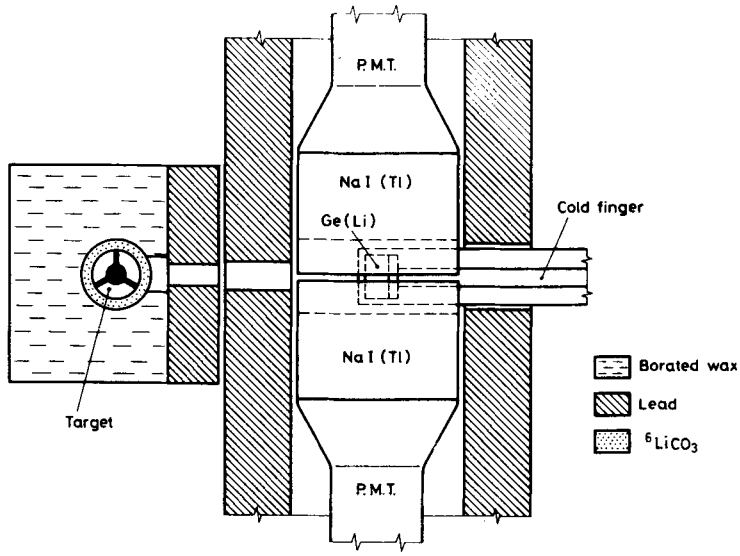

Fig. 1. A vertical section of the experimental set-up. The direction of the neutron beam is normal to the plane of the paper

carbonate enriched in ^{6}Li to 90% is used as target holder. A beam dump is situated 23 cm from the centre of the target holder tube.

A vertical section of the set-up is shown in Fig. 1. A 30 cm^3 Ge(Li) detector has been placed inside the 6.35 cm diameter slot of a 15.25 cm dia X 15.25 cm thick bifurcated NaI(Tl) annulus. Only the gamma-rays coming directly from the target are seen by the Ge(Li) detector through a lead collimator of aperture 2.22 cm. Each of the two NaI(Tl) halves is shielded from the target by a lead wall. The NaI(Tl) detectors coupled together are used as an anticoincidence shield for discriminating against compton scattered events in the Ge(Li) detector which lead to the detection of the scattered photons in the NaI(Tl) shield. The linear signals from the Ge(Li) detector which are not in coincidence (2τ = 100 nsec) with the output of either of the two NaI(Tl) detectors are recorded in a 4096 channel analyser. A digital spectrum stabilizer has been used to compensate for any gain or bias shifts.

The efficiencies for the full energy peaks in the anticompton mode remain the same as in the free mode whereas those for single and double escape peaks are reduced by factors of 2.7 and 5.6, respectively. The compton continuum is reduced on the average by a factor of 2 in the energy range covered by this spectrum.[28]

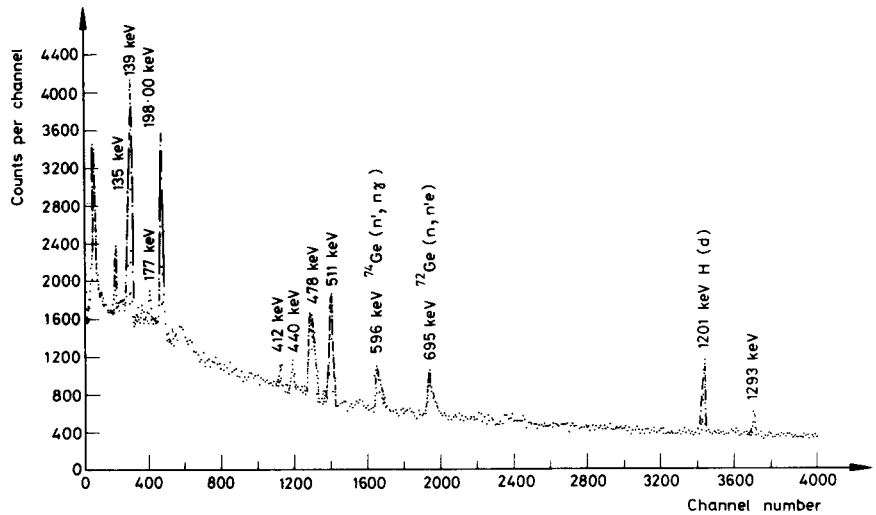

Fig. 2. The background spectrum taken with a polyethylene scatterer in the target position. (d) indicates double escape peak

Target samples

The stainless steel sample was machined into a cylindrical disc of diameter 1.8 cm × 0.12 cm thick. After cleaning, the sample (weighing about 2 g) was encapsulated in a thin polyethylene container. The powdered ore sample weighing about 9 g was filled into a similar container. The sample container was placed in the centre of the target holder tube and was supported on thin polyethylene fins.

Results and discussion

A list of the prominent gamma-rays along with their intensities,[22] which are expected to be seen as a result of neutron capture by Fe, Cr, Ni and Mn, the four main constituents of stainless steel, and B, is given in Table 1. The gamma-ray energy calibration standards used in this study are listed in Table 2.

A background spectrum, taken with a polyethylene scatterer in the target position, is shown in Fig. 2. Most of the low energy gamma-background can be seen to lie below 0.2 MeV. The 478 keV gamma-line is due to boron present in the shielding material. The excited nucleus ^{7}Li, which is produced as a result of the neutron capture in boron, emits this gamma-ray while in motion resulting in the

Table 1
Energies and intensities of the prominent neutron capture gamma-rays from different elements**

Element	Thermal neutron capture cross-section, barn	Energy, keV	Intensity, photons/100 neutrons captured
Fe	2.62	230*	1.53
		353*	10.85
		367*	1.47
		479	1.77
		692*	4.91
		898*	1.72
		1019*	2.29
		1261*	2.38
		1358*	1.03
Cr	3.10	564*	1.93
		749*	9.88
		835*	24.04
Ni	4.6	283*	3.69
		340*	3.07
		465*	14.32
		847	1.17
		878*	4.26
		1189	1.16
		1302	1.52
Mn	13.3	213*	6.02
		271	2.81
		314	3.55
B	752.0	258	1.65
		281	1.26
		453	1.20
		478*	470.0
		498	2.22
		502	1.91

* These lines have been used in the present analysis.
** Ref.[22]

broadening of the line. Another line at 695 keV which is due to an electron conversion transition in ^{72}Ge following inelastic neutron scattering, is broadened on account of the recoiling germanium ions.[27] A similar peak is observed at 596 keV due to the ^{74}Ge(n, n'γ) reaction. These peaks originate within the detector, as the polyethylene scatterer did not contain any Ge and are thus a measure of the neutrons scattered from the target.

The neutron capture gamma-ray spectrum of stainless steel sample is shown in Fig. 3. Full energy gamma-lines due to Fe, Cr, Ni and Mn are clearly seen. Relative masses of the elements present in the sample were obtained using the formula:

$$\frac{dN}{dt} = N \Phi \sigma I_\gamma \epsilon_\gamma = \frac{N_0 m}{A} \Phi \sigma I_\gamma \epsilon_\gamma$$

where $\frac{dN}{dt}$ – count rate due to a particular gamma-ray;

N – number of atoms of the element present in the target;

Φ – thermal neutron flux;

σ – neutron capture cross-section of the element;

m – mass of the element;

N_0 – Avogadro's number;

A – atomic mass of the element;

I_γ – number of the gamma-rays emitted per neutron capture in the element;

ϵ_γ – detection efficiency of the system for the gamma-ray.

All the lines listed in Table 1, however, could not be used to compute the masses due to either the low intensity or the interference from other gamma-rays. The gamma lines used for analysis are indicated in Table 1. The percentage of each constituent was calculated assuming the absence of any other element in the sample in significant quantity. The results obtained from the present measurements along with those from chemical analysis are presented in Table 3. The values quoted are the averages obtained using different gamma-lines and the errors indicate the standard deviations. The uncertainties in the values of Φ, σ, I_γ and ϵ_γ give rise to rather large errors in the results. These uncertainties can be removed by using the standard comparison method. Since the present study was principally directed to demonstrate the usefulness of lower energy gamma-rays for in-beam neutron activation analysis, detailed evaluation of the comparison method was not undertaken. Further work with the proper standard comparison technique is planned.

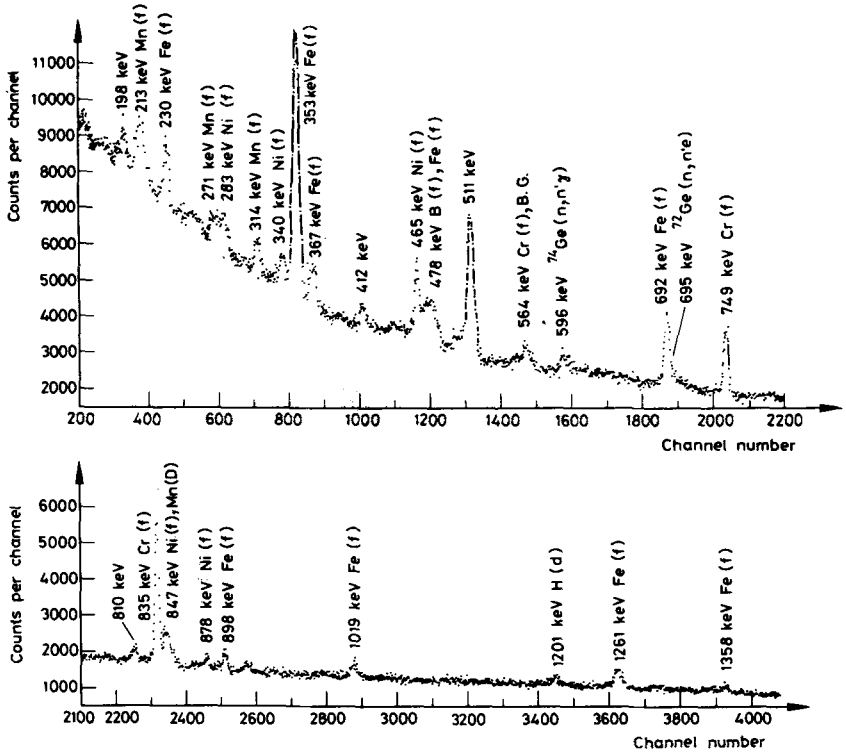

Fig. 3. The neutron capture gamma-ray spectrum of the stainless steel target. The prominent gamma-lines due to Fe, Cr, Ni and Mn are indicated. f: full energy peak, s: single escape peak, d: double escape peak and D: decay gamma-line

The iron ore spectrum is shown in Fig. 4. Since the composition of the ore was not exactly known, the amounts of Fe and Mn in the ore were obtained by using a different approach. This was done by comparing the peak areas of the relevant gamma-lines in the ore spectrum with the corresponding lines in the stainless steel spectrum. As the densities of the two samples were not the same, the neutron flux within the two samples is not expected to be the same. The relevant correction factor was determined by irradiating gold foils on each face of the two samples. The boron peak in the ore spectrum is much stronger than that in the background or stainless steel spectrum. After accounting for the contribution due to boron present in the shielding and the overlap by 479 keV Fe(n, γ) line, the amount of boron in the ore was estimated by internal comparison with Mn and Fe lines. The results for iron ore are listed in Table 4.

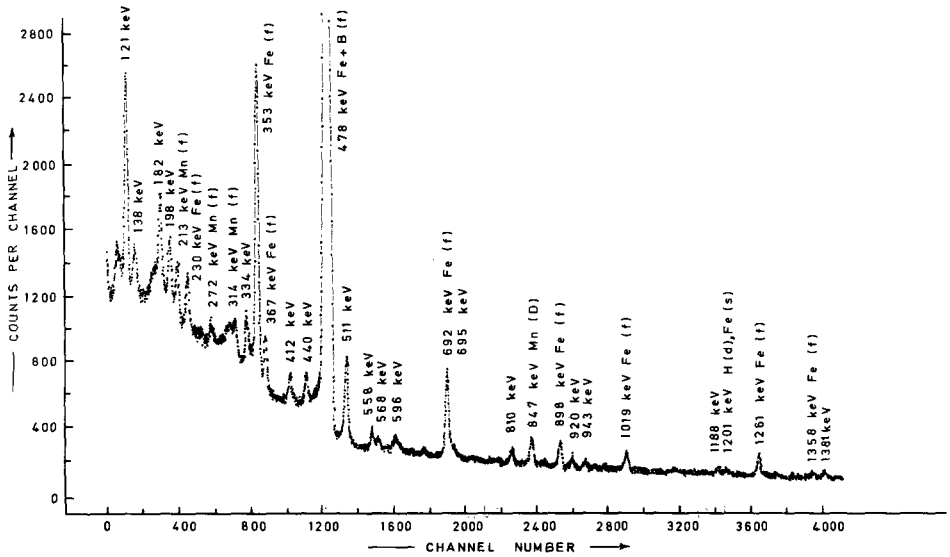

Fig. 4. The iron ore (n, γ) spectrum. f: full energy peak, s: single escape peak, d: double escape peak and D: decay gamma-line

Table 2
Gamma-ray energy calibration standards

Source	γ-Ray energy, keV
^{182}Ta	100.105 ± 0.001[a]
^{182}Ta	222.106 ± 0.002[a]
^{192}Ir	295.949 ± 0.006[a]
^{192}Ir	316.497 ± 0.007[a]
^{192}Ir	468.062 ± 0.010[b]
^{137}Cs	661.635 ± 0.076[c]
^{54}Mn	834.81 ± 0.03[c]
^{60}Co	1173.23 ± 0.04[c]
^{182}Ta	1221.376 ± 0.027[b]
^{60}Co	1332.49 ± 0.04[c]

[a] R. C. GREENWOOD, R. G. HELMER, R. J. GEHRKE, Nucl. Instr. Meth., 77 (1970) 141.
[b] R. G. HELMER, R. C. GREENWOOD, R. J. GEHRKE, Nucl. Instr. Meth., 96 (1971) 173.
[c] J. B. MARION, Nucl. Data, A4 (1968) 301.

Table 3
Results of the stainless steel sample analysis

Element	Amount,** %	
	In-beam analysis	Chemical analysis
Fe	66.1 ± 3.0	68.4 ± 1.1
Cr	22.8 ± 2.0	20.7 ± 0.8
Ni	8.3 ± 3.0	7.9 ± 0.3
Mn	2.3*	1.9 ± 0.1

* Only one line was used.
** Errors quoted are the standard deviations.

Table 4
Results of the iron ore sample analysis

Element	Amount, ** %	
	In-beam analysis	Chemical analysis
Fe	42.8 ± 4	43.3 ± 0.5
Mn	1.0*	1.1 ± 0.1
B	0.006*	–

* Only one line was used.
** Errors quoted are the standard deviations.

A comparison of the present work with that of ZWITTLINGER[26] indicates that with almost identical neutron flux and using much smaller (a factor of 25) amounts of the target material, the analysis with the present technique can be carried out in a much shorter time i.e. one quarter of that taken by ZWITTLINGER. This is a considerable advantage particularly when one has to deal with a large number of samples.

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