DETERMINATION OF MANGANESE IN STEELS USING PROTON-INDUCED NUCLEAR REACTIONS*

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The analytical use of prompt gamma-rays from the nuclear reactions on ^{55}Mn resulted from the bombardment of steels with 5.5 MeV protons was investigated. In this work a method for improving the limit of detection and for diminishing the errors of manganese determination in steels is described. It consists in the reducing of the background in the gamma spectra by the selection only of the (p,n) reaction channel, measuring the prompt neutron-gamma coincidences.

Key words: steels, manganese determination, PIGE analysis, nuclear reactions, proton irradiation.

1. INTRODUCTION

Manganese is usually introduced into steels to increase the tensile strength. Commonly, the concentration lies between 0.1% and 3% manganese by mass. Over this range of concentration particle-induced prompt gamma-ray emission (PIGE) technique [1–4] offers a rapid, precise and non-destructive method of analysis [5].

The analytical use of prompt gamma rays arising from the nuclear reactions ^{55}Mn(p, γ)^{56}Fe, ^{55}Mn(p,p′)^{55}Mn and ^{55}Mn(p, n)^{55}Fe [3,5] induced by 5.5 MeV protons during the bombardment of steel targets was investigated. Being an on-line technique, the sensitivity of PIGE critically depends on the level of background activities from the matrix nuclides under proton irradiation. In this paper a method for improving the limit of detection and for diminishing the errors of manganese determination in steels is described. It consists in the reducing of the background in the gamma spectra by the selection only of the (p,n) reaction channel, measuring the prompt neutron-gamma coincidences [2, 4].

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2. EXPERIMENTAL

The proton beam with an energy of 5.5 MeV has been generated with the aid of the 7 MV FN tandem accelerator of the “Horia Hulubei” National Institute of Physics and Nuclear Engineering (NIPNE) Bucharest. The beam current was below 10 nA. Prompt γ-rays produced in three steel plate samples (ST1, ST2, ST3) and one steel standard (E) provided by specialists from ISPAT-SIDEX Iron and Steel Works of Galatzy (Romania) were measured with a GeHP detector having an active volume of about 100 cm^3 and an energy resolution of 2 keV at 1.33 MeV, placed at 90° with respect to the beam. Coincident gamma spectra were measured simultaneously with the conventional ones, by means of a multiparameter analyzer system and processed off-line. The neutron scintillation detector, placed at 0° with respect to the beam, together with a pulse-shape discriminator provided the neutron-γ separation [2, 6]. The HPGe and the scintillation detectors have been coupled to a coincidence scheme of the type slow-fast and the resolution of the prompt coincidences curve in the gamma-ray energy range $E \geq 30$ keV was $2\tau = 17$ ns. The energetic and timing conditionings have been set with the aid of a multiparametric acquisition program. Neutron-gamma coincidences (E, n, Δt) spectra, obtained with the energetic window on the neutron spectrum, and singular gamma spectra were measured simultaneously and processed off-line.

3. RESULTS AND DISSCUSION

Details of the prompt n-γ coincidences and conventional γ-rays spectra obtained during the bombardment of the steel standard with 5.5 MeV protons are presented in Figs. 1 and 2, respectively, and in Table 1 are listed the γ-ray lines used to identify manganese in steel using both types of the spectra, labelled in accordance with the convention ^{A}X b(t, s) where ^{A}X is the target nuclide; b – the emitted nuclear product particle in the nuclear reaction induced by protons or deuterons; t and s – the level numbers in the heavy product nucleus between which the γ transition occurs.

Gamma energies corresponding to the transitions in the nuclei of interest were extracted from the nuclear level schemes [7] and from the yields of prompt gamma-rays resulted from the irradiation of pure elemental targets with protons [5, 8]. Gamma lines of 126 keV p(1,0), 858 keV p(2,1), 984 keV p(2,0), 1166 keV p(3,1), 1528 keV p(4,0) and 1883 keV p(5,0), reported in [5] for pure manganese target, arising from Coulomb excitation $^{55}Mn(p,p')^{55}Mn$, are not detected here in the steel target. The photon capture reaction leads to the population of the first two excited states to 2085 keV but the two possible gamma transitions of 846.8 keV γ (1,0) and 1238.3 keV γ (2,1) interfere with those

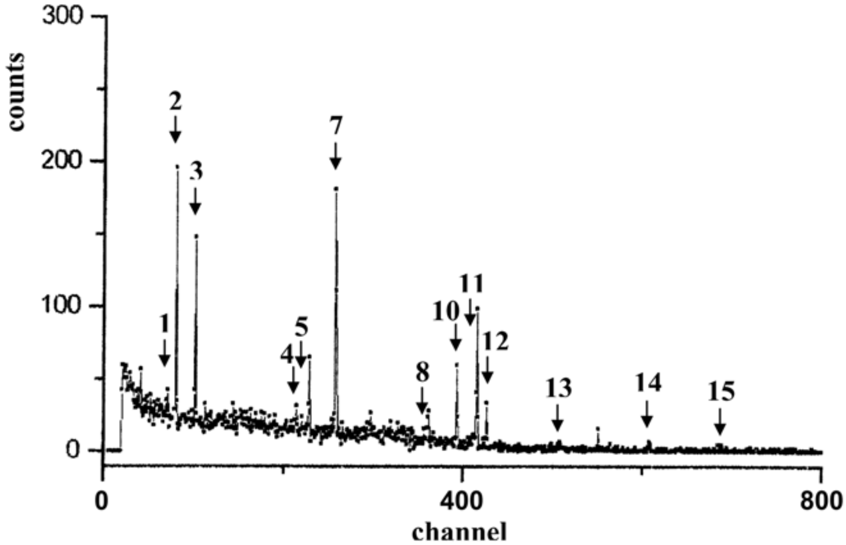

Fig. 1. – Detail of the prompt n-γ coincidences spectrum obtained during the proton bombardment of a standard steel sample.

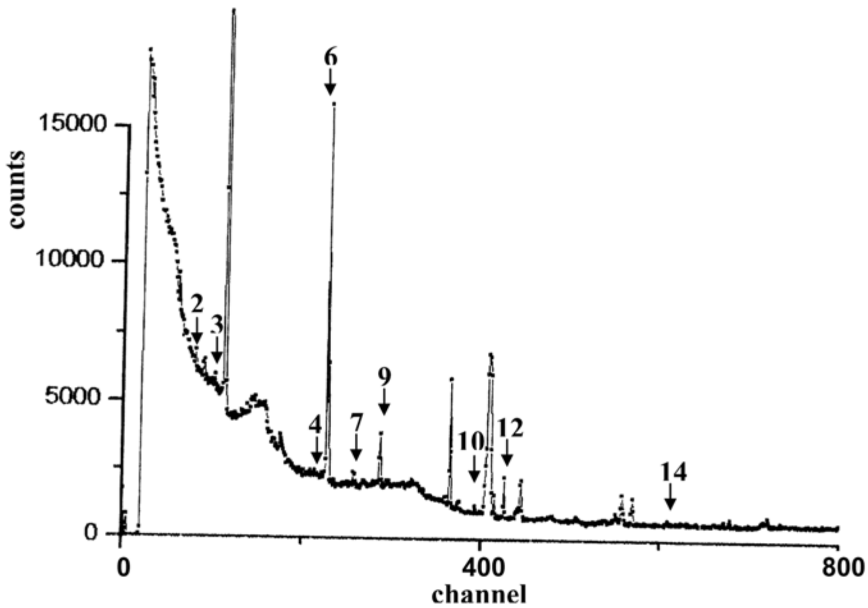

Fig. 2. – Detail of the conventional PIGE spectrum of a standard steel sample.

resulting from the reaction $^{56}Fe(p,p')\ ^{56}Fe$ taking place on the most abundant isotope of iron. Most gamma lines in the conventional PIGE spectrum (Fig. 2) originate from the $^{55}Mn(p,n)^{55}Fe$ reaction.

Table 1

Gamma lines of manganese in the analyzed steel standard

Peak no.	E_γ [keV]	Identity	Interference
1	385.4	^{55}Mn n (3,2)	
2	411.5	^{55}Mn n (1,0)	
3	477.1	^{55}Mn n (4,2)	
4	803.5	^{55}Mn n (8,4)	
5	827.4	^{55}Mn n (7,3)	
6	846.8	^{55}Mn γ (1,0)	^{56}Fe p (1,0)
7	931.4	^{55}Mn n (2,0)	
8	1212.8	^{55}Mn n (7,2)	
9	1238.3	^{55}Mn γ (2,1)	^{56}Fe p (2,1)
10	1316.8	^{55}Mn n (3,0)	
11	1368.2	^{55}Mn n (9,2)	
12	1408.5	^{55}Mn n (4,0)	^{54}Fe p (1,0)
13	1640.5	^{55}Mn n (6,1)	
14	1919.0	^{55}Mn n (5,0)	^{56}Fe γ (6,0)
15	2144.2	^{55}Mn n (7,0)	

From the conventional PIGE spectra we have obtained a detection limit of manganese determination in steel target of 115 ppm, which is comparable with the reported value of 109 ppm in the literature [1]. By the selection of the (p,n) reaction channel with the aid of n-γ coincidences measurement, the limit of detection was improved (18 ppm).

A comparison of the peak-to-background ratios in the conventional and coincident gamma spectra is given in Table 2 for the steel plate standard E

Table 2

Peak-to-background ratios for the most intense interference-free γ lines of manganese in the analyzed steel samples and the increase factor I for the coincident spectra

Sample	Spectrum type	Peak-to-background ratio		
		477 keV	931.4 keV	1316.8 keV
E	conventional coincident	0.22 2.33 I = 10.5	0.44 9.00 I = 20.5	0.615 7.14 I = 11.6
ST1	conventional coincident	0.181 1.176 I = 6.5	0.722 3.00 I = 4.15	0.714 2.50 I = 3.5
ST2	conventional coincident	0.718 1.183 I = 6.65	0.782 4.386 I = 3.43	0.709 2.70 I = 3.8
ST3	conventional coincident	0.208 1.189 I = 5.72	0.701 3.693 I = 5.28	0.734 2.359 I = 3.2

(containing 9770 ppm Mn) and samples ST1, ST2 and ST3 for the most intense interference-free γ lines of 477 keV (4,2), 931 keV (2,0) and 1316 keV (3,0) resulted from the $^{55}Mn(p, n)^{55}Fe$ nuclear reaction.

As can be seen from Table 2, the peak-to background ratios for the manganese peaks in the standard steel sample E at energies 477, 931.4 and 1316.8 keV have been calculated in this paper, being respectively 0.22; 0.44 and 0.615 in the conventional spectrum and 2.33; 9.00 and 7.14 in the coincident spectrum, determining an increase by a factor I of 10.5; 20.5 and 11.6. Also for the three steel samples the increase factors are: $I = 5.72 \div 6.65$ for 477 keV, $I = 3.4 \div 5.3$ for 931.4 keV and $I = 3.2 \div 3.8$ for 1316.8 keV.

These results are reflected in the improvement of the precision of manganese determination in the iron matrix. The values of the manganese determination using direct (conventional spectra) and coincidence measurements are presented in Table 3 and are in concordance but the relative errors of analysis are lower in the second case, for all the steel samples.

Table 3

Manganese concentrations in the steel samples obtained from the conventional and coincident PIGE spectra and the relative errors of analysis

Sample	Spectrum type	Manganese concentration [ppm]			Average concentration [ppm]	Relative error [%]
		477 keV	931.4 keV	1316.8 keV		
ST1	conventional	4128	4085	4697	4303 ± 342	8
	coincident	4197	4200	4151	4151 ± 27	0.7
ST2	conventional	2752	2706	3062	2840 ± 194	7
	coincident	2862	2671	3255	3007 ± 184	6
ST3	conventional	4130	4142	4118	4130 ± 413	10
	coincident	4542	4687	4489	4573 ± 102	2

4. CONCLUSIONS

An extension of PIGE method for manganese determination in steels is presented by measuring of prompt neutron-gamma coincidences with the selection of the (p, n) reaction channel. In this way the limits of detection and the precision of manganese analysis in iron matrix are improved by reducing the background in the gamma-rays coincident spectra.

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