MULTIELEMENTAL ANALYSIS OF STEELS VIA ATOMIC AND NUCLEAR METHODS*

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The capabilities of atomic and nuclear methods – Particle-Induced X-ray Emission (PIXE), Particle-Induced Gamma-ray Emission (PIGE) and Neutron Activation Analysis (NAA) – for the determination of minor and trace constituents of steels have been investigated. The steel samples and standards were obtained from the ISPAT-SIDEX Iron and Steel Works of Galatz. The minor and trace elements identified in the iron matrix of the steel samples were: K, V, Mn, Cr, Ti, W, Zn, Co, Cu, Ni, As, Mo, Rb, In, Rh, Pb (by PIXE); S, P, Si, Al, Mn, C, Ti, Ni, Na, O (by proton-PIGE) and Co, Zn, Mn, V, Sb, Ti, As, Cr, Mo, Cu, P selecting only (p,n) reactions; As, Pb, Sb, P, Cr, V, Ti, Cu, Fe, Ni, O, Co, Mo, Zn, Si, S, Al, Mn, C (by deuteron-PIGE) and Mo, Cr, Ni, Pb, Fe, Co, Ti, Zn, Sb, Mn, O, S, Cu, Si, V, P, Al selecting only (d,n) reactions. Application of NAA technique followed by gamma spectrometry, has led to the identification of Mn, Al, V, As, Cu, W, Ni, Mo, Cr, Sb, Co, Na, K, Ce, La, Sm, Sc, Zr, Zn, Au, Ga, Hf, Ta, Te, Se, Ba, Rb, Yb, Tb and Ir in steels.

Key words: PIXE, PIGE, neutron activation analysis, analysis of steels, minor and trace elements.

1. INTRODUCTION

The attractiveness of non-destructive methods and the ability to perform simultaneous multi-elemental determinations has led to an extensive application in steel industry of atomic and nuclear techniques such as neutron activation analysis (NAA) [1, 2], particle-induced X-ray emission (PIXE) [3, 4] and particle-induced gamma-ray emission (PIGE) using protons (p-PIGE) [5, 6] or deuterons (d-PIGE) [7]. Despite the high accuracy, precision and sensitivity of the three methods, all do suffer from one drawback or another [5, 6, 8, 9]. Among the factors affecting the sensitivity of PIGE frequently discussed in the


literature, the background in the \( \gamma \)-ray spectrum holds a prominent place. The enhancement of sensitivities of some minor elements in steels can be possible reducing the background by the selection of the neutron formation reaction channel, measuring the neutron-gamma coincidences following the nuclear reactions of charged particles on steel target [10]. Steels represent a good system in which this method could be tested because most steels contain a variety of minor components in a matrix consisting largely of iron.

In this work the capabilities of atomic and nuclear methods – Particle-Induced X-ray Emission (PIXE), Particle-Induced Gamma-ray Emission (PIGE) and Neutron Activation Analysis (NAA) – for the determination of minor and trace constituents of steels have been investigated.

2. EXPERIMENTAL

The steel samples and standards were obtained from the ISPAT-SIDEX Iron and Steel Works of Galatz. The used standards were British Chemical Standards BCS Nos. 320 (mild steel: Mo-0.22%; Ni-0.022%; Cr-0.131%; W-0.17%; Sn-0.085%; As-0.031%; Ti-0.021%; Al-0.013%;) and 402/1 (low alloy steel: C-1.2%; Si-0.13%; V-0.15%; Mn-0.16%; Cu-0.21%; Mo-0.17%; Ni-0.81%; Cr-0.77%; P-0.011%; S-0.032%), EURONORM CRM No. 085-1 (C-670 ppm; Si-80 ppm; S-3360 ppm; V-21 ppm; Mn-9770 ppm; Co-190 ppm; Cu-2910 ppm; Zn-25 ppm; Sb-73 ppm and Pb-10 ppm), available from Chemical Laboratory of ISPAT-SIDEX Iron and Steel Works at Galatz (Romania), ash standard EOP [11] (Cr-183 ppm; Mn-440 ppm; As-79.1 ppm; Na-3700 ppm; K-6700 ppm; Ce-322 ppm; La-164 ppm; Sc-36.7 ppm; Sm-21.9 ppm) from AIEA Vienna, metallic gold and Al_2O_3. Simultaneous PIXE–PIGE [4] analyses have been carried out using a 3 MeV proton beam generated with the aid of the 7 MV FN tandem accelerator of the National Institute of Physics and Nuclear Engineering (NIPNE) Bucharest. The detection system included a GeHP detector with an energy resolution of 200 eV at 5.9 keV for the X rays and a GeHP detector with an energy resolution of 2 keV at 1.33 MeV for the \( \gamma \) rays. The steel target was mounted in the irradiation chamber at 45° with respect to the beam and the both detectors’ direction. A thin surface barrier silicon detector was also placed in the chamber, at 135° with respect to the beam direction, in order to detect the backscattered protons for spectra normalization. The X-ray, \( \gamma \) ray and particle spectra were simultaneously collected and processed off-line. Separate PIGE determinations have also been done using 5.5 [6, 8]; 6.5 and 8 MeV protons [4] and 5 MeV deuterons [7]. Also, in the case of PIGE analyses using 5.5 MeV proton and 5 MeV deuteron beams, there were selected the (p,n) and (d,n) reaction channels by neutron-prompt gamma ray coincidences [6, 7, 10, 12] in order to improve the limit of detection of minor
elements in steels. The complete experimental arrangement and the general procedure have already been published for NAA at VVR-S Nuclear Reactor of NIPNE Bucharest [1, 2].

3. RESULTS AND DISCUSSION

Details of the PIXE spectrum obtained during the bombardment of a standard steel sample with 3 MeV protons are presented in Fig. 1 a,b,c and a detail of the PIGE spectrum of a steel sample using 6.5 MeV protons in Fig. 2, respectively, together with a table listing the $\gamma$-ray lines used to identify the component elements in the steel sample (Table 1), labelled in accordance with the convention [12]. Gamma energies corresponding to the transitions in the nuclei of interest were extracted from the nuclear level schemes [13] and from the yields of prompt gamma-rays resulted from the irradiation of pure elemental targets with protons [14]. The entire PIGE spectra of a steel standard EURONORM CRM No. 085-1 using 5.5 MeV protons is presented in [12] and using 5 MeV deuterons in [7]. In Fig. 3 the $\gamma$-ray spectrum from neutron activation of a steel sample is presented.

The minor and trace elements identified in the iron matrix of the steel samples were: a) K, V, Mn, Cr, Ti, Zn, Co, Cu, Ni, W, As, Mo, Rh, Pb (PIXE); b) S, P, Si, Al, Mn, C, Ti, Ni, Na, O (proton-PIGE) and Co, Zn, Mn, V, Sb, Ti, As, Cr, Mo, Cu, P selecting only (p,n) reactions; c) As, Pb, Sb, P, Cr, V, Ti, Cu, Fe, Ni, O, Co, Mo, Zn, Si, S, Al, Mn, C (deuteron-PIGE) and Mo, Cr, Ni, Pb, Fe, Co, Ti, Zn, Sb, Mn, O, S, Cu, Si, V, P, Al selecting only (d,n) reactions.

Application of NAA technique in three irradiation steps at VVR-S Nuclear Reactor of NIPNE Bucharest, followed by gamma spectrometry using four source-detector distances (1 cm, 5 cm, 6 cm and 12 cm), has led to the identification of Mn, Al, V, As, Cu, W, Ni, Mo, Cr, Sb, Co, Na, K, Ce, La, Sm, Sc, Zr, Zn, Au, Ga, Hf, Ta, Te, Se, Ba, Rb, Yb, Tb and Ir in steels.

The disadvantages of NAA are: i) there is a prolonged delay between the irradiation of the samples and the completion of the analysis; ii) some elements present in steel, such as the light ones, cannot be analysed because of the short half-life of the generated radioactive nuclei, or because the corresponding neutron-capture cross section is too small.

The limitations of PIXE are the following: i) in steels, iron is the major component so that the X-rays emitted from this element will dominate the energy spectrum, as can be seen from Fig. 1 a; ii) In the case of the analysis of the elements with $Z \leq 30$ an interference if frequently encountered between the $K_\alpha (Z+1)$ X-ray and the $K_\beta (Z)$ X-ray, which have virtually the same energy (Fig. 1 a) or between the X-K lines of medium elements and X-L lines of heavy elements (Fig. 1 b).
Fig. 1. – Detail of PIXE spectrum of a steel standard irradiated with 3 MeV protons.
In PIGE, on the one hand, the $\gamma$-ray yield is dependent on the type of the incident particle, its energy and on the cross-section of the nuclear reaction and, on the other hand, the bombardment of a complex target, such as steel, with...
charged particles may open many reaction channels so that the spectrum becomes extremely complicated (Fig. 2), making the identification and analysis very difficult. The nuclear interferences due to adjacent elements in the periodic table present in the steel target could be eliminated and the background lowered using prompt neutron-gamma coincidences measurements [12].

4. CONCLUSIONS

All the three methods – PIXE, PIGE and NAA – are non-destructive, multielemental, precise, sensitive and complemented each other. In this work are also presented their limitations in the particular case of steels analysis.

REFERENCES