Nuclear Instruments and Methods in Physics Research B 348 (2015) 165–169

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Application of PIGE, BS and NRA techniques to oxygen profiling in steel joints using deuteron beam

L. Csedreki *, R. Huszank

Institute for Nuclear Research, Hungarian Academy of Sciences, MTA Atomki, P.O. Box 51, H-4001 Debrecen, Hungary

ARTICLE INFO

Article history:
Received 10 July 2014
Received in revised form 1 December 2014
Accepted 3 December 2014
Available online 23 December 2014

Keywords:
DIGE
NRA
BS
Oxygen depth profile

ABSTRACT

In order to study the oxygen content and to characterize the oxygen depth profile on the surface of welded steel joints in the function of the applied shielding gases, particle induced gamma-ray emission (PIGE), backscattering spectrometry (BS) and nuclear reaction analysis (NRA) methods were used. The measurements were carried out at 1.0, 1.4 and 1.8 MeV deuteron energies. From the PIGE oxygen and carbon elemental maps ($1000 \times 1000\ \mu m^2$) taken with a beam of $2 \times 2\ \mu m^2$ beam size, oxygen rich regions were chosen for the depth profile analysis. The investigated depth was ~6 μm using particle detection (BS, NRA), which was extended to ~11 μm with the application of the differential-PIGE method, using the numerical integration of experimental cross-section data. The oxygen depth profiles show systematic discrepancy in the oxide layer thickness and composition between the two different kind of shielding gases.

1. Introduction

The selection and optimization of the proper welding technology is a key factor in construction industry, for which, the understanding and precise utilization of the physical processes is necessary. In the welding process, the presence of minor elements such as carbon, oxygen, sulphur and selenium are highly affecting the weld shape of stainless steel. The control of these elements is critical for the satisfactory welding process. The effects of the welding parameters on the weld shape and depth/width ratio strongly depend on oxygen content of the weld metal and the diffusion mixing [1]. The investigation of the oxygen depth profile at the surface of the welded joints in solid state helps to discover the possible effects of the applied shielding gases, moreover it may also affects the welding process.

Different surface analysis techniques are generally used in steel investigation such as scanning electron microscopy, energy dispersive X-ray spectroscopy, several ion beam analytical (IBA) techniques etc., but only a few are suitable in oxygen depth profile measurements. Among IBA techniques those which are using particle detection such as backscattering spectrometry (BS) and nuclear reaction analysis (NRA) are widely used in the determination of stoichiometry, areal density and the distribution of target elements as a function of depth. Moreover, NRA is complementary to BS technique due to its higher sensitivity in the detection of light elements on heavy substrate [2]. The advantage and limitation of oxygen analysis using charged particle bombardment was summarized in Cohen et al. [3]. Several papers deal with the measurement of oxygen depth profiles in different materials (see e.g. [4]), however only a few applications of IBA techniques to the investigation of steel surface using particle detection were found in literature [5–7]. The analysed depth varied between 1 and 5 μm in these applications and the energy of deuteron beam was low, 665 keV in Refs. [5,6] and 1200 keV in Ref. [7], respectively. Even the use of higher bombarding energy the investigated depth is limited by the overlap of the emerging p_o and p_1 oxygen peaks from the $^{16}O(d,p)^{17}O$, reaction, and the high yield from backscattered deuterons.

Particle induced gamma-ray emission (PIGE) technique – especially in the case of deuteron beam (DIGE) – is suitable for the determination of carbon, nitrogen and oxygen. Due to the high penetration of gamma-rays, the investigated depth depends on the penetration of bombarding particles. Therefore it would be useful to find a method based on the detection of gamma-rays from oxygen to extend the range of particle depth profiling also in ten μm scale. Similarly to the differential-PIXE introduced by Demortier et al. [8] for the analysis of gold artefacts with layered or depth profile structure and used also for the study of layered structure of paintings [9], the differential-DIGE method was introduced in this work. The development of the IBANDL database, which includes reliable and detailed gamma-ray production cross sections with suitable

* Corresponding author. Tel.: +36 52 509200; fax: +36 52 416181.
E-mail address: csedreki@atomki.mta.hu (L. Csedreki).

http://dx.doi.org/10.1016/j.nimb.2014.12.013
0168-583X/

accuracy for the IBA analysis, makes possible the introduction of this method. Besides, the main disadvantage of PIGE method in comparison to particle induced X-ray emission (PIXE) and backscattering spectrometry (BS) is that the reaction cross sections which are required to determine the elemental composition cannot be given easily due to the resonance feature (though the modern computer capacity is able to solve this problem). Performing measurements on the same area with various beam energies, the oxygen content of different depth layers in steel can be determined from the obtained gamma-ray yields, taking into consideration the differential cross section and stopping power data.

Due to the possible inhomogeneity of the surface of the welded joints on lateral and depth scale, the elemental mapping is essential for the selection of an area of interest (when the non-destructivity is necessary). These problems can be solved using nuclear microprobe.

The aim of this work was the optimization of differential-DIGE, BS and NRA techniques applied them in nuclear microprobes for the determination of oxide layer thickness and profile in steel joints, prepared with different shielding gases. These results help to understand the welding processes better and to reveal the advantages of different types of shielding gases.

2. Experimental

The experiments were carried out on the Oxford-type scanning nuclear microprobe facility at ATOMKI, Debrecen, Hungary [10]. The measurements were made at 1.0, 1.4 and 1.8 MeV deuteron energy, which were performed with a 5 MV single ended Van de Graaff accelerator. The beam was focussed to $2 \times 2\ \mu m^2$ while the scan size was set to $1000 \times 1000\ \mu m^2$. From the elemental maps, oxygen rich regions were selected and scanned during the measurement. The sample chamber is equipped with a high precision 5-axes goniometer. In order to position the samples to the appropriate geometry, two laser beams were used [11]. The accumulated charges were determined by a beam chopper, calibrated with a Faraday-cup on each deuteron energy. Typical beam current and collected charge were 500 pA and 1 μC, respectively. The uncertainty of charge was taken into account with ±5%.

The experimental set-up consisted of a coaxial type HPGe detector of 170 cm^3 volume positioned at an angle of 45° relative to the beam direction at a distance of 10 cm from the target, and an Ion Implanted Si detector with 500 μm thick depleted layer and 13 keV energy resolution placed at an angle of 135° relative to the beam direction at a distance of 2.65 cm from the target [12]. A copper collimator with a hole diameter of 3 mm was used in front of the Si detector. The solid angle of Si detector was determined with geometrical calculation and the calculated value was verified with RBS measurements carried out on thin palladium standards evaporated onto silicon substrate. The solid angel of Si detector was 9.3 ± 0.1 msr.

In order to determine the absolute efficiency (ε_{abs}) of the gamma-ray detector, radioactive sources (^{56}Co, ^{60}Co, ^{137}Cs, ^{152}Eu) were used. The uncertainty of the absolute efficiency (ε_{abs}) of the HPGe detector is ±4% based on the measured efficiency curve. The more detailed description of the calibration process can be seen elsewhere [13]. Lead shield was used to eliminate the gamma background from the materials of set-up and the laboratory background.

For the IBA investigations, two kinds of welded joints samples were used, which were prepared using two kind of shielding gases. In the case of the so called AC and IC samples, the compositions of shielding gases were 82% Ar + 18% CO_2 and 85% Ar + 10% CO_2 + 5% O_2, respectively. Based on the composition of applied shielding gases, we expect increase in the oxide layer by the AC sample compared with the IC sample. Moreover, due to the application of active shielding gases, "skin" did not grow up on the surface, thus, surface treatment could be avoided.

Optical microscopy was used for preliminary investigation to discover the possible surface inhomogeneity, which may affect the ion beam analysis and to choose suitable regions for further nuclear microprobe investigation.

The thickness of the studied steel samples caused significant reduction of gamma intensity, which was taken into account in the (DIGE) calculations. Supposed that the sample has iron matrix the reduction of gamma intensities of $^{16}O(d,p\gamma)^{17}O$-E_γ = 871 keV and $^{12}C(d,p\gamma)^{13}C$-E_γ = 3089 keV gamma lines were 9.5% and 5.5%, respectively.

2.1. Gamma-ray detection

Fig. 1 shows a typical gamma-ray spectrum of the AC sample measured at 1.8 MeV deuteron beam, included the 871 keV gamma-line of $^{16}O(d,p\gamma)^{17}O$ reaction, the 511 keV from annihilation, the 3089 keV gamma-line of $^{12}C(d,p\gamma)^{13}C$ reaction and several peaks as a background. The spectrum includes the gamma-ray peak from the $^{27}Al(d,n\gamma_{1\text{-}0})^{28}Si$ transition, which is caused by the interaction between the beam and the aluminium chopper wing.

The high intensity gamma-rays from $^{16}O(d,p\gamma)^{17}O$ and $^{12}C(d,p\gamma)^{13}C$ reaction (in comparison to the much weaker particle yields) were applied for the study the lateral distribution of oxygen in the welded joint part of the steel samples and to choose an appropriate region for the depth profile study. Fig. 2 shows the oxygen (a) and the carbon (b) elemental maps of the AC sample. We used oxygen maps for the selection of the measured area.

The oxygen and carbon concentration were determined based on the 871 keV and the 3089 keV gamma-lines, respectively. The accuracy of the yields of the determination was better than ±1% concerning the oxygen and under ±6% in the case of the carbon reaction.

In the calculation of oxygen and carbon concentration, experimental cross sections were used based on IBANDL database [14,15]. The relative errors of the applied experimental gamma-ray production cross section were below ±6%. In order to verify the reproducibility of the calculated concentration from DIGE, the samples were re-measured at E_d = 1.8 MeV and the discrepancy was less than 4% between the normalized yields at 1 μC.

2.2. Particle detection

Fig. 3 shows a typical particle spectrum of the IC sample measured at 1.8 MeV deuteron energy: the figure include different part

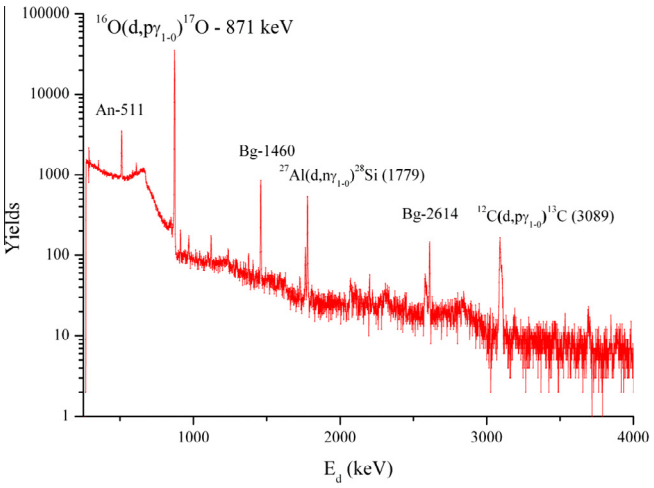

Fig. 1. Typical gamma-ray spectrum measured at 1.8 MeV deuteron energy on AC sample.