II. 2 Charged Particle Activation Analysis of Oxygen in Iron

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Purification of iron by anion exchange method and floating zone-refining was
confirmed effective to remove most metallic impurities.1-3) Carbon and oxygen
are considered to be major remaining impurities,1) though melting in dry hydrogen
and wet hydrogen treatment are adopted in order to minimize and keep constant the
concentration of non-metallic impurities. It is very important to examine the
distribution and the concentration of carbon and oxygen, because these elements
seriously affect the properties of iron and iron-base alloy such as hydrogen
diffusion,4) corrosion resistant property,5) electrical properties6) and vacancy
migration.7)

Charged particle activation analysis (CPAA) is well known to be one of the
most sensitive methods for the analysis of light elements. Many authors have
reported on CPAA experimentally8-10) and theoretically.11) But there is few
paper on CPAA of light elements in transition metals. Blümel et al.12) reported
the contents, obtained by CPAA, of carbon, nitrogen and oxygen in iron specimen
annealed in an atmosphere of dry hydrogen without giving detailed analytical
procedure.

In this paper, a method of activation analysis of oxygen in iron using
$^{16}O({}^3He,p)^{18}F$ and $^{16}O({}^3He,n)^{18}Ne$ ($^{18}Ne + {}_{\text{He}}^1 + {}_{\text{F}}^8 + {}_{\text{e}}^0$) reactions is described.
This method has the advantage of extremely lower detection limit and less amount
of sample compared with a conventional extraction method through vacuum fusion.
Furthermore, influence of surface oxide layer can be avoided by etching away of
surface portion after bombardment.13)

Iron specimens were commercial high purity iron with nominal purity of 99.99
% supplied by Jonson-Matthey Co. (J-M iron) and twice purified electrolytic iron
(T-E iron) with fairly high oxygen content (see table 1). The latter was used
as a standard for oxygen concentration. The specimens 0.5 mm in thickness were
polished chemically and some of them were annealed in wet or dry hydrogen at
800°C for 24 h. The specimen mounted on target holder was bombarded through
the collimator (10 mm in diameter) for 20-40 min. The energy and current of $^3He$
ion beam was 16 MeV at the surface of specimen and 1.5-2.0 μA, respectively.

In the case of activation analysis, special attention must be paid to the
interference nuclear reactions that produce the same radio nuclide as marked one.
Engelman10) examined the interference reactions in detail. The interference can
be estimated negligibly small in this experimental condition from his results.

After bombardment, surface was etched away chemically by 20 μm to remove
the surface oxide layer and the portion contaminated through recoil.9) And then
specimens were dissolved in nitric acid (2:1) with carrier NaF and $^{18}F$ was
separated as PbFCl precipitate by the process shown in fig. 1. The time required for the separation is usually within 1 h from the finish of bombardment. Radioactivity was measured by detecting 511-keV γ-rays coming from the positron annihilation of $^{18}$F. The recovery yield of carrier was determined as follows. PbFCl precipitate was dissolved in nitric acid(1:9). The known amount of $\text{AgNO}_3$ larger than stoichiometry was added to precipitate $\text{AgCl}$. After filtration, the excess $\text{AgNO}_3$ was determined by titration using KSCN. About 50% of carrier was recovered and recovery yield of the carrier was determined precisely. Gamma-ray spectrum of the iron specimen showed that $^{57}$Ni was produced through the bombardment with $^3$He ion beam. Chemical separation is known to be very important, since the presence of $^{57}$Ni gives high level of background through similar $\beta^+$ decay.

Figure 2 shows decay curves of 511-keV γ-ray from PbFCl precipitate and iron hydroxide. Their half lives agree with $^{57}$Ni(36.0 h) and $^{18}$F(109.8 m). It is confirmed that $^{57}$Ni is well separated. Final results are tabulated in table 1. The values in bracket are the results obtained by the infrared absorption method with vacuum extraction and in good agreement with the data of activation analysis.

Judging from the radioactivity of the specimens and the background level of the surroundings, detection limit is estimated to be 100 ppb in this experimental condition. The process developed in this paper is expected to be easily applied to other metals. Examination of the relation between oxygen content of iron and the condition of hydrogen treatment is under experiment.

References

10) Engelman Ch., J. Radioanal. Chem. 7 (1971) 281.
Table 1. Analytical results of oxygen in iron specimens

<table>
<thead>
<tr>
<th>Sample</th>
<th>Treatment</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>as rolled</td>
<td>Standard (366)</td>
</tr>
<tr>
<td>T-E</td>
<td>wet $H_2$</td>
<td>64.8</td>
</tr>
<tr>
<td></td>
<td>dry $H_2$</td>
<td>70.5</td>
</tr>
</tbody>
</table>

|        | as rolled | 26.9 (26) |
| J-M    | wet $H_2$ | 11.7       |
|        | dry $H_2$ | 11.7       |

$^3$He ion bombardment

Etching away of surface layer

Carrier NaF

Dissolution in nitric acid

NaOH

Centrifugal separation of hydroxide

$\text{HNO}_3$

pH : 3.6-5.6

NaCl, Pb(NO$_3$)$_2$

PbFCl precipitate

Measurement of activity

Determination of recovery yield of F

Fig. 1. Process for separation of $^{18}$F.

Fig. 2. Decay curves of 511 keV gamma-ray.