

## Summary

We elaborated a uniform formulation for calculating estimated count rate errors in gamma spectrometry. Experimental results confirmed that our method gave accurate confidence limits combining both random and bias errors.

## *Accuracy in CPAA for C, N and O and in ERDA and NRA for H*

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### 1. Introduction

Trace amounts of H, C, N and O can be determined by only a few methods, often with rather poor accuracies. Charged particle activation analysis (CPAA) is highly reliable for C, N and O, and is used for calibration of other methods. Elastic recoil detection analysis (ERDA) and nuclear reaction analysis (NRA) have recently been utilized for H and D near the surface. Accuracies in these analyses are discussed on the basis of our experimental data.

### 2. Accuracy in CPAA

#### 2.1 Advantage and Error in CPAA

The advantage of CPAA for C, N and O consists of inherent high sensitivity, freedom from various contaminations, and reliability in the use of comparators. The following reactions are used for activation:  $^{12}\text{C}(\text{He},\alpha)^{11}\text{C}$  or  $^{12}\text{C}(\text{d},n)^{13}\text{N}$ ;  $^{14}\text{N}(\text{p},\alpha)^{11}\text{C}$ ; and  $^{16}\text{O}(\text{He},\text{p})^{18}\text{F}$ . Uncertainty in the measurement of bombarding particle beam current frequently becomes a major cause of error in CPAA. Also, some bombardment damage of the sample often results in noticeable overestimation of C and O.

#### 2.2 Precision for O and C

We analyzed O in a silicon wafer repeatedly nine times from 1982 to 1985 by the nondestructive

measurement of the  $^{18}\text{F}$  annihilation radiation with a Ge(Li) detector [1]. The results gave the mean value of 10.04 ppm wt with  $\sigma=0.18$  ppm.

We examined chemical separation of  $^{11}\text{C}$  in Si, using  $^{11}\text{C}$ -containing Si prepared by proton bombardment of B-doped Si [1]. The following separation method was selected: alkali-dissolution of the pulverized sample;  $\text{KMnO}_4$  oxidation of the  $^{11}\text{C}$  under microwave heating to 700 °C; and generation of  $^{11}\text{CO}_2$  and its conversion into  $\text{Li}_2^{11}\text{CO}_3$ . The measurement of  $^{11}\text{C}$  activity before and after the separation and of carrier recovery indicated that  $9.0\pm 2.0\%$  of the  $^{11}\text{C}$  apparently disappeared. Our results obtained by this separation has thus been corrected for this 9%.

For the separation of  $^{13}\text{N}$ , dry fusion into  $^{13}\text{N}_2$  and wet distillation as  $^{13}\text{NH}_3$  are used [1]. Carbon in 27 plates of GaAs made from a single rod were analyzed by the dry method in different machine times of two cyclotrons. The mean results were 24.6 ppb wt with  $\sigma=2.6$  ppb. For the last five plates taken from adjacent parts of the rod, the results were  $23.25\pm 0.84$  ppb. We separate  $^{18}\text{F}$  by precipitation as  $\text{KB}^{18}\text{F}_4$  [2]. This method has shown satisfactory precision.

#### 2.3 Calibration of IR Spectrophotometry

Our calibration curve for IR spectrophotometry of C in Si is shown in figure 1 [1]. About 70 carefully prepared samples were submitted to round-robin IR measurement in 24 organizations. About 1/3 of them were then analyzed by CPAA; some of them were also analyzed by SIMS. The differences between the results of CPAA and SIMS are probably caused by the presence of background in SIMS. Satisfactory calibration curves were obtained also for O and N in Si [3,4]. Efforts are now being made to obtain a reliable calibration curve for C in GaAs.

### 3. Accuracy for ERDA and NRA for H

Figure 2 shows ERDA spectra of a H- and D-containing amorphous silicon film on a Si wafer [5]. The detection efficiency is seen to depend highly on the detection angle. No suitable substances are found as comparators for H and D, and their quantities are usually obtained in comparison with an internal standard element by the use of the theoretical relationships and experimental parameters. The H content of amorphous silicon films obtained by ERDA and vacuum fusions are given in

table 1 [5]. At present, the accuracy for H by ERDA is notably lower than for C, N and O by CPAA. The depth profiling of H and D is possible by ERDA, as is seen in figure 2; but further studies are necessary.

In the depth profiling of H by the  $^1\text{H} + ^{15}\text{N} \rightarrow ^{12}\text{C} + \gamma(4.44 \text{ MeV})$  reaction, it is easy to get the shape of the profile reproducibly but difficult to obtain an absolute value for H within a 20% uncertainty. We now use D as an activatable tracer, determining it by the reactions  $\text{D} + ^3\text{He} \rightarrow \alpha + \text{p}(14 \text{ MeV})$  and  $\text{D} + ^{15}\text{N} \rightarrow ^{16}\text{O} + \text{n}(\text{or } \text{p} + \beta^-) + \gamma(6.13, 7.11 \text{ MeV})$  [6]. Experimental data are required concerning the accuracy of these methods.

**Table 1.** Hydrogen content of amorphous silicon films measured by three methods ( $10^{17}$  atoms/cm<sup>2</sup>): ERDA, vacuum extraction with the measurement of volume (VOL) and heat conductivity (HC)

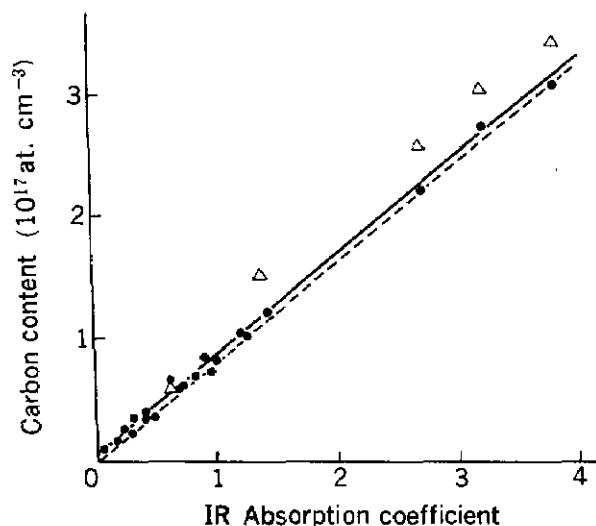
Film thickness (nm)	ERDA	VOL <sup>a</sup>	HC <sup>b</sup>	VOL/ ERDA	HC/ ERDA
1065	7.36	7.92	7.38	1.08	1.00
830	5.77	6.54	5.90	1.17	1.06
672	4.25	5.60	5.11	1.32	1.20
650	3.78	5.26	4.40	1.39	1.16
526	4.53	4.40		0.97	
510	3.96	3.63		0.92	
310	2.40	3.07	2.67	1.28	1.11
270	1.85	2.19	1.89	1.18	1.02
Average				1.16 $\pm 0.17$	1.09 $\pm 0.08$

<sup>a</sup> By Prof. Ogawara, Science Univ. of Tokyo.

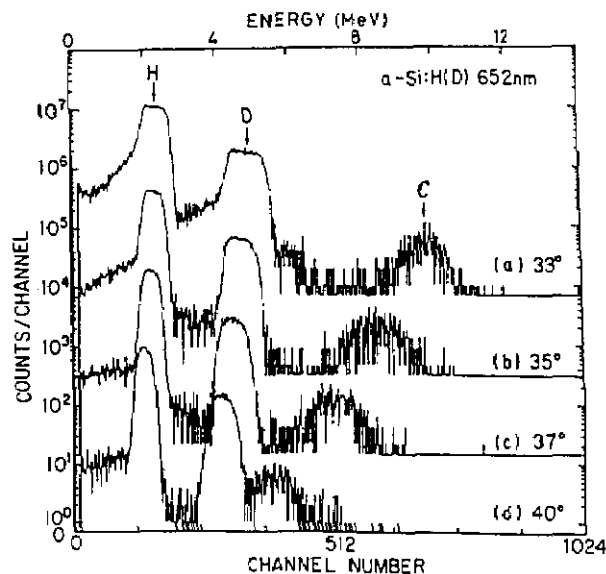
<sup>b</sup> By Dr. Nomura, Mitsubishi Metals Co.

#### 4. Conclusion

At present, accuracy can be determined for the CPAA of C, N and O. For the ERDA and NRA determination of H and D, experimental data are still too scarce for consideration of accuracy.



**Figure 1.** Calibration curve for IR spectrophotometry of C in Si ( $605 \text{ cm}^{-1}$ , at room temperature). Solid line, before correction for C in IR reference Si; (C concentration,  $4.3 \times 10^{15} \text{ at. cm}^{-3}$ ); broken line, after the correction;  $\Delta$ , results of SIMS.



**Figure 2.** ERDA spectra for various detector angles. Sample: H- and D-containing amorphous silicon film 652 nm thick; incident particle: 42.3 MeV Ar; target angle: 30 °C; absorber: 10  $\mu\text{m}$  Al foil.

#### References

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### *A Fast-Neutron Diagnostic Probe*

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A novel, nuclear method for instrumental analysis of elements has been developed. The system, called a "neutron diagnostic probe (NDP)," has several unusual capabilities that are especially applicable to nondestructive, 3-dimensional analysis in solid materials or inaccessible spaces. The prototype instruments that have been assembled and tested demonstrate that the NDP system can penetrate several inches of solids, locate inaccessible inhomogeneities by time-of-flight ranging, and perform elemental analysis of the located volume of interest. This neutron diagnostic probe utilizes a combination of associated-particle, neutron time-of-flight spectroscopy; inelastic gamma-ray spectroscopy; and a recently developed sealed-tube neutron generator. The NDP interrogates a material or inaccessible space with an electronically collimated and timed beam of fast neutrons which have great penetrating power. Highly penetrating, inelastic gamma rays produced in the interrogated volume by the neutrons are detected and analyzed. The spatial production of these gamma rays is analyzed to "image" the distribution of materials within the volume of interest. The energies of the gamma rays from each substance within the space examined are analyzed to determine the concentrations of the elements in each volume element. An inelastic gamma-ray energy spectrum characteristic of every element except hydrogen can be obtained.

The name of this associated-particle technique [1] is derived from the alpha particle "associated" with a 14-MeV neutron produced by the  $T(d,n)He^4$  reaction in the neutron generator. Because the associated particle is produced simultaneously with the fast neutron and is emitted in the opposite direction, its detection specifies the subsequent trajectory of the individual neutron. This capability makes it possible to define a timed and directed "beam" of 14-MeV neutrons traveling away from a continuous  $T(d,n)He^4$  generator at about 5 cm per nanosecond. By using time-of-flight, range gating, only gamma rays generated by neutron reactions within a known sensitive volume at a predetermined distance from the generator are recorded.

The key element in the instrumentation is the sealed-tube neutron generator (STNG) shown schematically in figure 1. The basic design of the neutron generator is similar to that described by Reifenschweiler [2,3], using the  $T(d,n)He^4$  reaction, a getter-controllable mixture of deuterium and tritium gases and a self-loading target. The innovations in the STNG used for this work are: 1) the inclusion of an internal alpha detector to supply time and direction information by the associated-particle technique and 2) provision for focusing the ion beam on the target to ensure a "point source" of 14-MeV neutrons.

In operation, the radiation detectors and nuclear electronics are arranged as shown in the block diagram of figure 2. Light signals from the ZnS alpha scintillator are amplified by a photomultiplier (PM) and preamplifier, shaped by a constant-fraction discriminator (CFD) and routed to the "start" input of a time-to-amplitude converter (TAC). The alpha signals are also counted and stored in the multi-channel analyzer (MCA) data acquisition system for subsequent normalization of the spectral information. The gamma rays generated by inelastic neutron interactions in the interrogated sample are converted to energy spectra with a 15 cm diameter by 36 cm long NaI(Tl) scintillation spectrometer. These signals are shaped (CFD) and routed to the "stop" input of the TAC to provide a range-proportional, time-of-flight spectrum at the TAC output. The time-of-flight spectrum is proportional to range because the 14-MeV neutrons travel from the STNG to the interrogated sample at a constant 5 cm/ns and the gamma rays produced in the sample travel at 30 cm/ns to the NaI(Tl) detector. Signals from the portion of the time-of-flight spectrum that corresponds to the range position of the sample are selected to open a gate circuit (linear gate)