Indian J. Phys. **65A** (6), 551-553 (1991)

An external beam irradiation system for trace elemental determination in liquids

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Abstract : An external beam irradiation facility for direct trace elemental analysis of liquid samples has been developed at the Variable Energy Cyclotron Centre, Calcutta. The system has been standardized primarily for transition metals, in liquid samples. The system has been found to behave lineraly over a wide range of concentrations. The minimum detection limits (MDL) were found in the order of 15-160 ppm in presence of interfering elements and 0.6-10 ppm in the absence of interfering elements The results were validated by analysing NIST standards.

Keywords : Trace elements, external beam, charged particle activation analysis, liquid samples.

PACS No : 29.90.+r

1. Introduction

Charged Particle Activation Analysis (CPAA) (Khathing 1988) has been mostly used for the analysis of light elements ($Z < 10$) in high purity metals, semiconductors etc. Recently the trend of extending CPAA in analysing medium and high Z elements has been seen. The extraction of particle beam from accelerator vacuum into air offers several advantages for irradiation and subsequent elemental analysis (Katsanos *et al* 1976, Seaman and Shane 1975). This aspect has been incorporated in Particle Induced X-ray Emission (PIXE) and Particle Induced Gamma ray Emission (PIGE). While external-beam PIXE and PIGE have received wide attention, not much seems to have been done on the external beam CPAA system, in particular for liquid sample analysis. The role of transitional trace metals (V, Cr, Mn, Fe, Co, Ni, Cu, Zn) individually or as group in various fields such as clinical studies, industry, agriculture etc, is well established. These elements (except Mn, Co and Cu) have poor sensitivity in thermal Neutron Activation Analysis (NAA). Inspite of the fact that external beam PIXE and PIGE can measure these elements effectively in ppm levels, they have the inherent problems

associated with sample preparation, and mounting of samples on fragile backings. These facts led us to study the feasibility of a system for rapid, direct analysis of liquid samples with minimum sample preparation. We report the studies carried out on aqueous and crude oil samples.

2. Experimental

A detailed study on reaction channels, their cross sections, product half lives, and beam energy degradation was performed and specific reactions were identified. Tables 1 and 2 show the selected reactions in presence and in absence of interfering elements along with the observed detection limits. A steady beam collimated

Table 1. Nuclear reactions with irradiation energy and MDL values in presence of interfering elements.

Element	Reaction	Can thickness (μm)	Final energy on target (MeV)	MDL (ppm)
V	$^{51}V(\alpha n)\ ^{54}Mn$	400	14	28
Cr	$^{50}Cr(\alpha pn)\ ^{52}Mn$	150	25	90
Mn	$^{55}Mn(\alpha n)\ ^{58}Co$	400	14	14
Fe	$^{54}Fe(\alpha n)\ ^{57}Ni$	400	14	15
Ni	$^{64}Ni(\alpha p)\ ^{67}Cu$	400	14	158
Ga	$^{71}Ga(\alpha n)\ ^{74}As$	300	19	17

through a set of collimators was extracted through a Havar foil (25.4μ) and used for the irradiation. Precise irradiation parameters to avoid interferences from neighbouring elements were optimised. Elemental interferences were solved by

Table 2. Nuclear reactions with irradiation energy and MDL values in the absence of interfering elements.

Element	Reaction	Can thickness (μm)	Final energy on target (MeV)	MDL (ppm)
V	$^{51}V(\alpha n)\ ^{54}Mn$	200	30	32
Cr	$^{50}Cr(\alpha pn)\ ^{53}Mn$	200	30	9
Mn	$^{55}Mn(\alpha n)\ ^{58}Co$	200	30	5
Fe	$^{56}Fe(\alpha pn)\ ^{59}Co$	200	30	5
Ni	$^{58}Ni(\alpha 3pn)\ ^{59}Co$	200	30	9
Zn	$^{64}Zn(\alpha 3n)\ ^{65}Ge$	200	30	4
Cu	$^{63}Cu(\alpha 2n)\ ^{67}Ga$	200	30	0.06
Ga	$^{71}Ga(\alpha n)\ ^{71}As$	200	20	4

irradiating foils deposited with the interfering pair of elements on Al backings. The system linearity was checked by irradiating spiked samples of different concentrations. The irradiated samples were transferred into glass vials and counted on Ge(Li) detector of resolution 1.8 keV at 1332 keV using standard nuclear

instrumentation. Quantitative calculations were made using the method of average cross section (Ricci and Hahn 1967).

3. Results

The system standardisation involved optimisation of irradiation parameters viz, beam energy and current, counting and cooling times, interference studies, etc. In the liquid sample irradiation it was observed that isotopes with halflives less than 6 to 8 hours could not be used for measurements owing to the strong 511 keV annihalation γ-ray originating from the matrix (mainly C, N and O) of liquids or organic samples. The system was found to be linear over a wide concentration range.

The MDL values shown in Table 1 are for an irradiation of two hours at selected energies with beam current of about 250 nA, while in Table 2 they were calculated after irradiation of two hours at selected energies with beam current of about 120 nA. The MDL values at 45 MeV irradiation for ten minutes with beam current of about 250 nA was also determined (Rao *et al* 1988).

The results were validated by analysing NIST SRM No 8505 (Vanadium in Crude oil) and SRM No 1634b (Trace elements in Fuel oil). The Vanadium values obtained for SRM 8505 (certified 390 $\pm$ 10 ppm) was 384 $\pm$ 33 ppm while for SRM 1634b (certified 55.4 $\pm$ 1.1 ppm) was 55.1 $\pm$ 4.5 ppm. The errors involved in the measurement when added in quadrature was about 5-15%.

4. Conclusion

This investigation shows that external beam CPAA in the easily avaiable range of cyclotron energy can be effectively used for trace level quantitative estimation of transition metals in liquids. The system though surveyed primarily for transition elements can be extended to other elements too.

Acknowledgment

One of us (VRR) is thankful to Council of Scientific and Industrial Research, New Delhi for providing financial help in the form of individual fellowship. The authors wish to acknowledge Mr D K Bandopadhay of Variable Energy Cyclotron Centre, Calcutta for extending technical help in designing and fabricating the sample holder.

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