ACTIVATION ANALYSIS WITH CHARGED PARTICLES: THEORY, PRACTICE AND POTENTIAL

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Abstract

Activation analysis using charged particles (CPA) from cyclotrons and other similar accelerators is presented. Its advantages over neutron activation analysis using thermal neutron (NAA) is presented. Also some of the difficulties involved with the CPA are also highlighted along with the practical solutions to overcome these difficulties. Our "special theory" [1] is presented that makes the CPA as simple as the NAA. A practical example is given where the Effect of French Atomic Tests of the 197's in the Pacific on the Australian East Coast was studied with CPA. By making use of empirically constructed excitation functions of Lange and Muenzel [2] we have estimated the detection sensitivities of all the elements and isotopes with Z from 20 to over 90, through 1, 2 &3 particles-emission under irradiation with protons, deuterons and alpha particles of up to 36 MeV and have presented these in graphical form [3].

INTRODUCTION

Activation analysis with slow neutrons from a reactor is the most commonly used radio-analytical method. However, certain elements such as Al, Si, Ti, Cd, Tl, Pb, Bi, etc. can not be conveniently or at all activated with slow neutrons. Furthermore, for biomedical samples, due to the presence of large quantities of Na, radiochemical separation is required before NAA can be carried out. Particle induced X-ray emission Analysis (PIXE) is another accelerator based technique which is simple and has multi-elemental measurement capabilities. But PIXE is not so convenient for measurement for elememts lighter than F. Furthermore, its detection sensitivity is limited to around ppm (or µg/g) range.

Charged particle activation analysis tends to overcome many of these short comings. It can be non-destructive and elements lighter than F can be conveniently and accurately determined and sensitivities of as low as ppb (µg/kg) range can be achieved in favourable circumstances. Furthermore, this technique has multi-elemental detection capability.

METHOD

Mostly energetic protons energy, ca. 8-10 MeV have been used for CPA, although other charged particles, such as deuterons, He-3 and He-4 also offer a great deal of potential in this field.

However, unlike the NAA there are a number of practical difficulties in CPA which have to be overcome. These are:

1. Energy loss and heat generation in the sample under irradiation with charged particles. This can be minimised by cooling the sample holder with water, air or even liquid Nitrogen. In the case of powdered samples the heat generation problem can also be overcome by mixing the sample with analytical grade Graphite and thus making it conductive.
2. Simultaneous and controlled "identical" irradiation .of the sample and the standard is absolutely essential in CPA. It can generally be achieved by keeping the irradiation conditions energy, beam-intensity, etc., stable and monitoring the beam carefully. Alternatively, one can also mount the sample and the standard on a special target holder and rotating it at a constant speed during irradiation. This assures that the sample and the standard have been irradiated under identical conditions.

The trace element to be determined has to be uniformly distributed in the matrix. This condition is generally met in many types of samples, especially in biological fluids and organs, which are freeze-dried and /or ashed and then homogenized.

Chaudhri et al. [1], have presented a simplified theory of CPA which reduces it to the simplicity of NAA. According to this theory the induced activity in an unknown sample A is related to the induced activity in the standard (with known concentration of the element being determined) A_s with the following equation:

$$A_s / A = Cs\ Qs / C\ Q\ \mathbf{X}\ (dE/dx) / (dE/dx)s. \qquad [1]$$

Where C is the concentration, in appropriate units in the sample, Q is the charge collected to induce activity A and dE/dx is the stopping power of the incoming beam in the sample-matrix at the "mean-energy" of the incident particles. This "mean-energy" is given by $[E_i + E_0] / 2$ where Ei and E_0 are respectively the incident energy and the lowest energy for which an appreciable amount of induced activity would be produced. Various terms with the subscript "s" refer to the standard. In Eq. (1) all quantities are measured or can be taken from the published tables with the exception of C, which can then be calculated.

Chaudhri et al. [4] have recently shown, through extensive calculations, that this theory produces the best results as compared to any other approximation for CPA and it's a lot simpler.

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As a practical application of CPA we present our results on the study of the effect of French Atomic Tests series of 1974 in the Pacific on the atmosphere of the Australian Eastern Coast. High Volume Air Samplers, operated by a government organization, had collected air samples from East coastal cities from Port Moresby to Hobart. This was done by passing 5000m^3 of air through 0.2 micron-Polystyrene filters. The particulate matter, greater than 0.2μ would be collected by the filter. The air samples were collected just before the atomic tests started and just after the tests finished. Our hypothesis was that the concentrations of elements and isotopes, both radioactive and stable, would change between the two sets of air particultes, if there was any "Fall-Out" on the Australian East Coast.

The Polystyrene Filters were analyzed by activation with 8 MeV protons. The filters were cut into small pieces and mounted on a target holder, which was cooled with liquid nitrogen. The intensity of the proton beam was 1 μA. It was found that the cooling of the targets was satisfactory as none of the filter pieces showed any charring signs after irradiation. A number of elements, S, Ca, Ti, Cr, Fe, Ni, Cu, Zn, Se and Hg, ranging in concentrations from 0.001μg/m^3 to upto 3,27 μg/m^3 were found. However, no differences in the concentrations of various elements determined in the particulate matter collected just before and just after the atomic tests indicated that there was no Fall-Out on the Australian East Coast cities due to te French Atomic Tests of 1970's in the Pacific.

POTENTIAL

It has already been mentioned that CPA has the potential to achieve detection sensitivities of ppb levels in favourable circumstances. There are quite a few publications in the literature giving the sensitivities of detecting different element with CPA. The work of Debrun and Barrandon [5] is worth mentioning. They estimated the detection sensitivities of many elements activated by 10 MeV protons at 1 μA for 1 hour radiation time. Under these conditions they give achievable sensitivities of:

10^{-3} – 10^{-2} ppm for Ca, Ti, Cr, Ni, Cu, Zn, Ga, Ge, Se, Br, Rb, Y, Zr, Mo, Ru, Pd, Cd, Sn, Te;
10^{-2} – 10^{-1} ppm for Li, Va, Fe, As, Sr, Nb, Ag, Sb, I, Pt, Hg, Nd, Er;
10^{-1} – 1 ppm for B, S, Re, Ir, Au, Tl, Pb, La, Pr, Sm, Gd, Dy, Yb;
1 -30 ppm for In, Ba, W, Eu, Rb, Tb.

These sensitivity values might be improved by using other nuclear reactions and other projectiles, such as He^3, deuterons and alpha particles.

We have estimated the activation sensitivitiers for 1,2 and 3 particle emission reactions for all of the elements / isotopes with z from 20 to 92, activated by protons, deuterons and alphas of up to 35 MeV for a thick-target. The reaction cross sections are taken from Lange and Muenzel [2].The results are presented in graphical form from which the induced activities under any bombarding conditions, for a thick or thinner targets can be directly read [6]. Some examples of such sensitivity curves for 1 and 2 particle emission reactions, induced by protons and and deuterons are presented in Fig. 1-3.

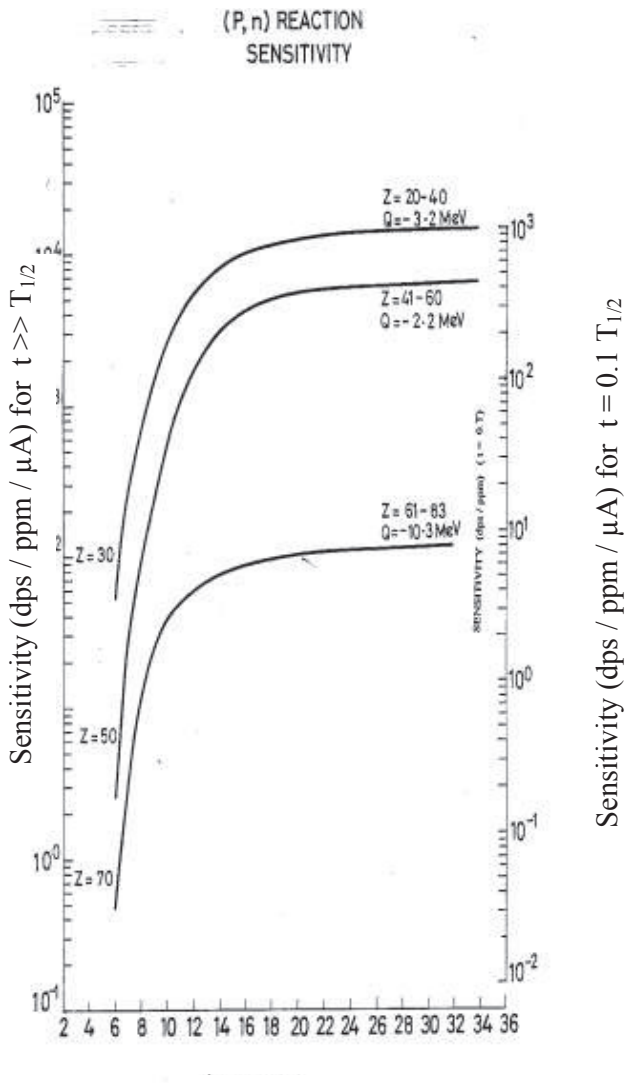

Figure 1: Activation-sensitivity curves for detecting anyelement/isotope, with Z=20 to Z > 90, through (p,n) reaction.

Due to the way in which the excitation functions are presented by Lange and Muenzel [2], we have also presented the thick-target activation curves in similar

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fashion. That is grouping a number of elements/isotopes in different sets, such as Z = 20-40; 41-60; 61-83 and Z > 90. The average of Q-values of the set of elements/ Isotopes for a particular reaction type, were used to convert the excitation functions of Lange et al [2], in a useful form. The results are presented in a graphical form from which the induced activities under any bombarding condition. Full details of these estimations are published by Chaudhri et al. [3]. Some examples of such sensitivity curves for 1 and 2 particle emission reactions, induced by protons and deutedrons are shown in Figs. 1-3.

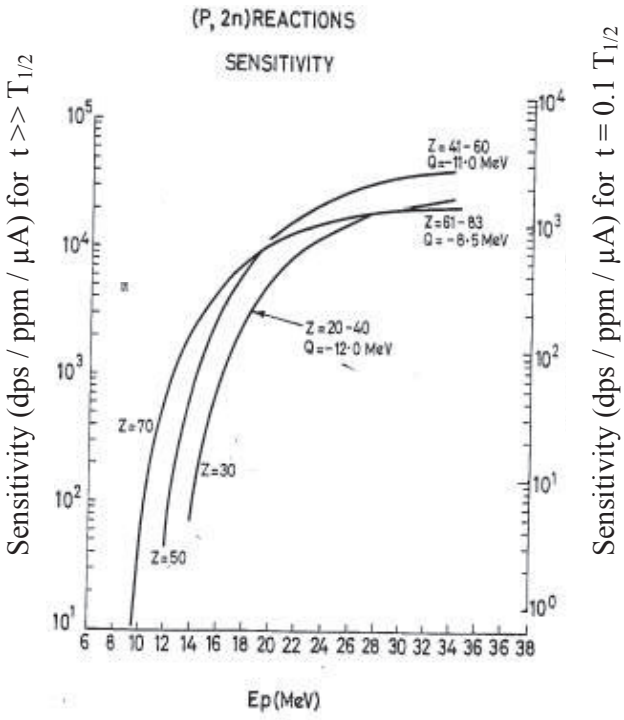

Figure 2: Activation-sensitivity curves for detecting any element/isotope, with Z=20 to Z > 90, through (p,2n) reaction.

As matrices (the material/compound) where the element being determined) the means of Z values of the elements invarious groups are chosen. For example the matrix for the group Z=20-40 is the element/compound with Z = 30. This is clearly indicted on the curves. The left hand ordinate gives the activation sensitivity in dps/ppm/μA at irradiation time which is much longer than the half-life of the induced activity A (approximately 5 half lives or greater). The right-hand ordinate gives the same information for an irradiation time which is one-tenth of the half-life of the induced activity. For any other irradiation time t, the induced actuivity A_t can be obtained from the well-known equation

$$A_t = A[\, 1 - \exp(-\lambda t)\,],$$

where λ is the "Decay Constant" of the induced activity.

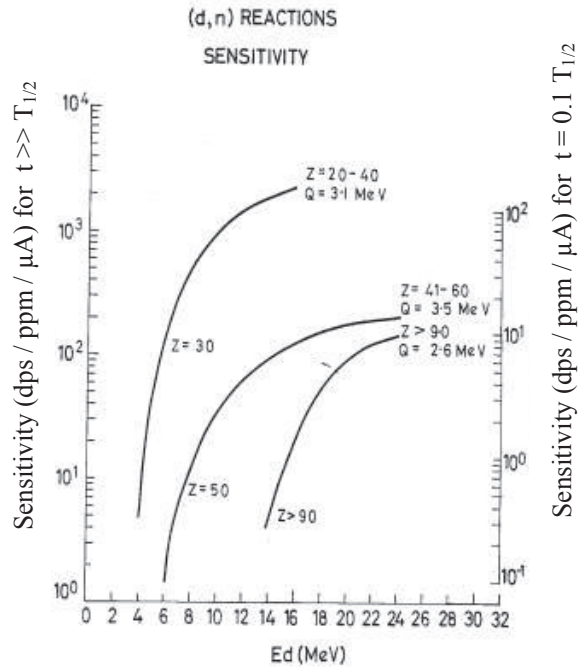

Figure 3: Activation-sensitivity curve for detecting any element/isotope, with Z = 20 to Z > 92, through (d,n) reaction.

It can be easily seen from the curves that sensitivities of the ppb levels, or even lower, are easily obtainable.

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