EFFECTS OF HYDRATION ON ELEMENTAL SENSITIVITIES IN NEUTRON ACTIVATION ANALYSIS USING PROMPT AND DELAYED GAMMA RAYS

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A 1.85×10^{11} Bq (5Ci) Am/Be neutron source is being used to investigate the feasibility of applying the absolute method in the elemental analysis of bulk samples by instrumental neutron activation making use of prompt and delayed gamma rays. Among the different factors which might contribute to variations in sensitivity or in the uncertainty associated with the elemental concentration is the presence of hydrogen in the sample. It is the purpose of this study to examine whether the presence of water in bulk samples such as landfill waste produces significant variations in measured elemental concentrations and if so, requires samples to be dried, which can be a time consuming and expensive process. The effect of hydration of samples on the sensitivity of ten elements has therefore been investigated. Samples with different water concentrations in a fixed volume and shape were prepared and used in the analysis in order to obtain a measure and an understanding of the effects involved.

In performing neutron activation analysis of voluminous samples using the absolute method, a number of correction factors should be applied in order to obtain a valid estimate of the elemental concentrations. Factors such as neutron self shielding, gamma ray attenuation within the sample and detector efficiencies for distributed sources are of great importance and have been studied before[1,2]. Another factor considered in this study, which might contribute to the analytical error, is the effect of hydrogen on the elemental sensitivities. Bulk samples which are supplied to the laboratory for environmental monitoring contain different amounts of hydrogen, and if analysed as such, might produce erroneous values of elemental concentrations when comparing results from similar type of samples due to the scattering by hydrogen atoms within these samples. Bulk sample analysis using neutron activation techniques have been performed previously in different fields including oil logging[3], water pollutant analysis[4,5], in-vivo analysis[6] and on-line analysis of coal[7]. The issue of neutron thermalization due to the presence of large volumes of hydrogen and carbon within the sample was investigated[7].

The ideal case in neutron activation experiments is when the target material experiences the same neutron flux with the same energy distribution throughout the target volume. This should result in a linear response curve between the reaction rate and the mass of analyte. Practically, it is not possible to meet this situation with relatively large targets because the

neutron flux strongly depends on the scatterers and absorbers in the sample and the sample shape as well[8,9]. The neutron flux is affected by the amounts and the cross-section values of the absorbers and scatterers present in the sample. It is particularly sensitive to the amounts of hydrogen (e.g. as water) due to its ability to slow down the neutrons. The size and the shape of the sample also affect the reaction rate. The increased probability of escape of scattered neutrons from the sample results in a decrease of the reaction rate; on the other hand if the probability of neutron escape is sufficiently small, this results in an increase in the reaction rate[10,11,12]. The presence of elements with large neutron absorption cross section can also decrease the neutron flux through neutron self shielding, but corrections can be made using a simple absorption law[13].

Experimental

Three to four kilogrammes were a typical weight for the different sample matrices. The samples were dried in an oven at a temperature of 60°C for a week to ensure that all water had evaporated. The samples were put in polyethylene containers of 156 mm dia. and 275 mm length. The neutron facility shown in Figure 1 accommodates an Am/Be neutron source (1.85×10^{11} Bq), with an emission rate of 10^7 n/s for irradiation. A perspex tube of about 1070 mm length and an inner diameter of 50 mm runs through the centre of a large water tank. The neutron source can be transferred within the tube from shielding position to irradiation position using compressed air. The source stop at the irradiation end consists of a metal plate with a circular hole in it; the diameter of which is 10 mm less than that of the perspex tube. Powdered Li_2CO_3 in a perspex box is placed in front of the neutron source with a hole of 50 mm diameter through the box's centre to allow neutrons to pass through it. In addition a perspex box containing boron loaded clay is placed round the perspex flight tube in order to further reduce the number of neutrons travelling towards the detector. Paraffin wax is also used to reduce the number of neutrons scattered by the water shield towards the detector. Lead is used as a shield against the dynamic gamma rays background.

Irradiation and counting was performed for 17 to 24h (long irradiations are required since the neutron flux is very low) in order to obtain good counting statistics. Known amounts of double distilled water were added to each sample, and each time the sample was subjected to the above mentioned irradiation and counting scheme. A background spectrum was measured with an empty container at the irradiation position for the same irradiation/counting time adopted for the samples, and used to correct the full energy peak areas resulting from the samples. One can argue that the background with and without the sample is different, due

to the increase in the number of neutrons scattered by the sample, which in turn interact with the detector and shielding materials and therefore contributing more counts in the sample background[14].
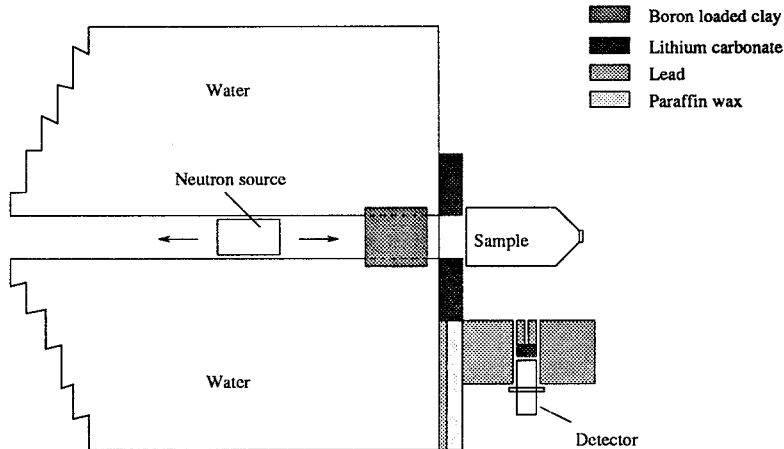

Figure 1: Geometrical configuration of the experiment (plan view-not to scale)

Results and discussion

Bulk samples of landfill waste and coal were used to study the effects of water on the elemental sensitivities. The relative sensitivity enhancement, RSE is defined as:

$$RSE = [(NH_i/ND_i)-1]$$

where NH_i is the full energy peak area of radionuclide i in the hydrated samples, and ND_i is the full energy peak area of the radionuclide i in the dried sample. Sensitivities of B, C, Na, Al, S, Cl, Ti, V, Fe and In have been measured in a range of hydration from (0 to 20%) of the sample weight(w_s). Only V and In were measured through their delayed gamma rays, all other elements through their prompt gamma rays. Double distilled water was used to hydrate the samples. As shown in Figures 2 to 7, all the elements studied showed an increase in the sensitivity in the first few percent of added water. This is interpreted as the ability of the added hydrogen as a large scatterer to thermalize the remaining epithermal neutrons, since the major part of this component was already thermalized by the constituents of the dried sample. So the increase in sensitivity continues until the majority of the epithermal neutrons have been thermalized. The sensitivity then tends to remain steady, even with extra additions of water, and may even fall as more neutrons are captured in water. Comparing the increase in the elemental sensitivities between the landfill and coal samples, it was found that the sensitivity
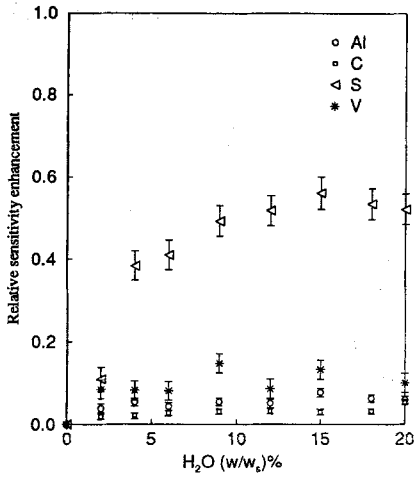
Figure 2: Measured relative sensitivity enhancement for
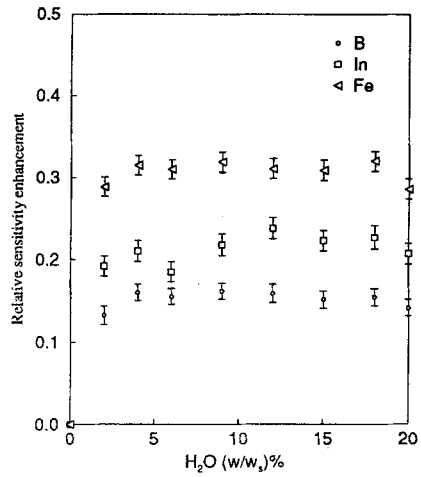Al, C, S and V in bulk coal sample.

Figure 3: Measured relative sensitivity enhancement for
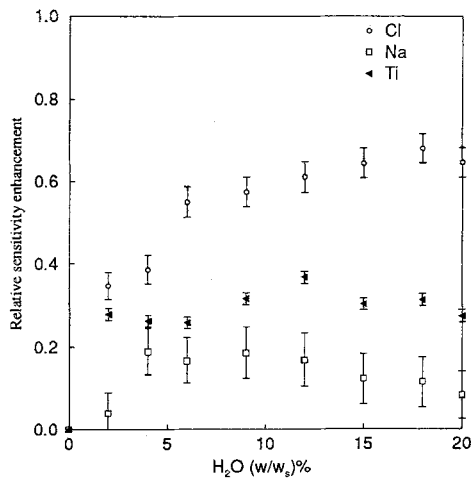B, In and Fe in bulk coal sample.

Figure 4: Measured relative sensitivity enhancement for
Cl, Na and Ti in bulk coal sample.

enhancement was more pronounced in the landfill sample than in the coal, and a calculation of the macroscopic absorption and scattering cross-sections using the major constituents of both samples, reveals that there is more scattering and absorption in the landfill sample. The sensitivity reaches a maximum value in these samples with a lower percentage of water added.
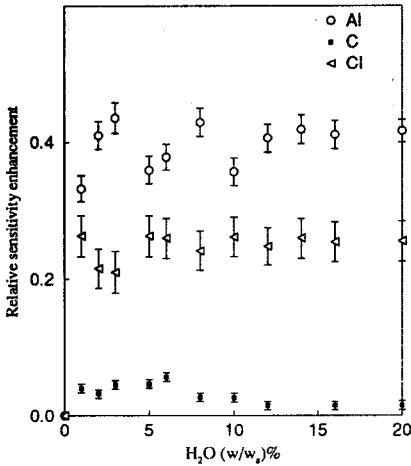
Figure 5: Measured relative sensitivity enhancement for
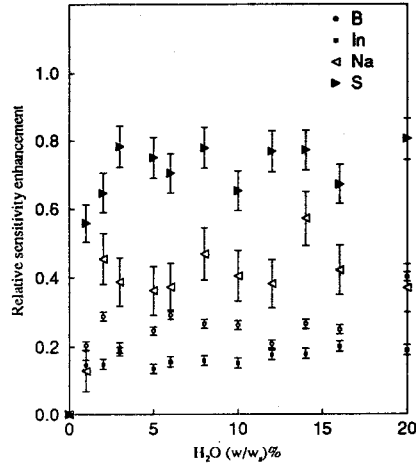Al, C and Cl in bulk landfill waste sample.

Figure 6: Measured relative sensitivity enhancement for
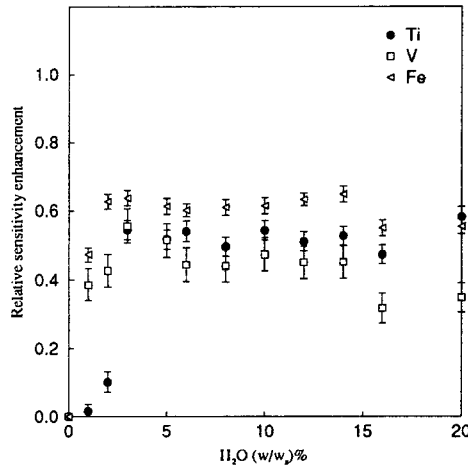B, In, Na and S in bulk landfill waste sample.

Figure 7: Measured relative sensitivity enhancement for
Ti, V and Fe in bulk landfill waste sample.

Conclusions

The results obtained when an increasing amount of water is added to a sample matrix show an increase in elemental sensitivity up to a certain value after which it remains more or less constant. However, the amount by which the sensitivity was enhanced seems to vary from element to element and from matrix to matrix. This therefore suggests that by hydrating

all samples by a few percent of water will produce not only an increased sensitivity (up to seventy percent) in the detection of elements but also allow intercomparisons of elemental concentrations to be made from sample to sample. It also obviates the need for time consuming and probably expensive drying procedures.

References

1. R.M.W. OVERWATER, P. BODE, J.M DE GOEIJ, Nucl. Instr. & Meth. in Phys. Res., A324 (1993) 209.

2. M.B. ALAMIN, N.M. SPYROU, Simultaneous cyclic and prompt gamma-neutron activation analysis in large landfill and sewage samples analysis., in Proc. Int. Conf. on Nuclear Analytical Methods in the Life Sciences, Prague, 13-17 Sep. 1993 (in press).

3. R. HERTZOG, IEEE Trans. Nucl. Sci., 35,1 (1988) 827.

4. C. CHUNG, T.C. TSENG, Nucl. Instr. & Meth. in Phys. Res., A267 (1988) 223.

5. J.H. CHAO, C. CHUNG, Nucl. Instr. & Meth. in Phys. Res., A299 (1990) 651.

6. I.P. MATTHEWS, N.M. SPYROU, Int. J. Appl. Radiat. Isot., 33 (1982) 61.

7. M.R. WORMALD, C.G. CLAYTON, Int. J. Appl. Radiat. Isot., 34,1 (1983) 71.

8. E.A. MACKEY, G.E. GORDON, R.M. LINDSTROM, D.L. ANDERSON, Anal. Chem., 63 (1991) 288.

9. D. TRUBERT, G. DUPLATRE, J.Ch. ABBE, Int. J. Appl. Radiat. Isot., 42,8 (1991) 699.

10. J.R.D. COPLEY, C.A. STONE, Nucl. Instr. & Meth. in Phys. Res., A281 (1989) 593.

11. J.R.D. COPLEY, Nucl. Instr. & Meth. in Phys. Res., A307 (1991) 389.

12. K.H. BECKURTS, K. WIRTZ, Neutron Physics, Springer, Berlin, 1964.

13. R.F. FLEMING, Int. J. Radiat. Isot., 33 (1982) 1263.

14. D.L. ANDERSON, E.A. MACKEY, J. Radioanal. Nucl. Chem. 167,1 (1993) 145.