*Journal of Radioanalytical and Nuclear Chemistry, Articles, Vol. 133, No. 2 (1989) 217–226*

# NEUTRON ACTIVATION ANALYSIS AND RADIOACTIVITY MEASUREMENTS OF AUSTRALIAN COALS AND FLY ASHES

J. J. FARDY, G. D. McORIST, Y. J. FARRAR

*Centre for Advanced Analytical Chemistry, CSIRO, Division of Fuel Technology, PMB 7, Menai, NSW, 2234 (Australia)*

(Received February 27, 1989)

Forty elements were determined by instrumental neutron activation analysis in a wide range of Australian coals and fly ash to update and extend earlier measurements. The natural radioactivity content of selected samples were analysed by high resolution gamma-ray spectrometry and low level radiochemistry. The results indicated a marked disequilibrium of the $^{232}$Th decay series in some samples while a general enrichment of $^{210}$Pb in most fly ash samples disrupted the $^{238}$U equilibrium.

## Introduction

The combustion and liquefaction of coal produce a variety of emissions and effluents ranging from particulates in flue gases to solid and liquid wastes. Included in these wastes are traces of the natural occuring radionuclides, $^{40}$K, $^{87}$Rb and the radioactive daughters from the decay series of $^{238}$U and $^{232}$Th. During the last decade, increased interest in the concentration and fate of these inorganic elements has been reflected in the large number of analyses performed at these laboratories on a wide range of Australian coals and fly ashes, both independently and in collaborative research projects with industry and other government departments. The aim of these studies was to verify, update and extend earlier measurements of trace elements in Australian black[1-6] and brown coals[7] and to assess the levels of natural radioactivity in these coals.[8,9] Trace elements in coal and fly ash samples were determined by instrumental neutron activation analysis (INAA); selected samples were analysed for natural radioactivity using high resolution gamma-ray spectrometry with lithium-drifted germanium, hyperpure germanium and low energy photon detectors and the ultra-sensitive technique of low level radiochemistry.

*Elsevier Sequoia S. A., Lausanne*
*Akadémiai Kiadó, Budapest*

## Experimental

The trace element profiles of coal and fly ash were measured by INAA techniques involving two irradiation periods and four counting intervals. All irradiations were performed in the self-service facilities of the X-176 tube (short period irradiation, 1 to 3 min.) and the X-6 tube (long-period irradiation, 5 to 24 hrs.) of HIFAR, a 10-MW DIDO-type reactor system located at the Lucas Heights Research Laboratories, at thermal neutron fluxes of $5 \cdot 10^{13}$ and $5 \cdot 10^{12}$ $n \cdot cm^{-2} \cdot s^{-1}$, respectively.

For short irradiations, samples and NBS standard coal (SRM1623) and/or NBS fly ash (SRM1633) were weighed (70–100 mg) into pre-washed (nitric acid, demineralised water, A. R. ethanol, air-dried) polythene vials (0.13 ml) and individually irradiated. After the irradiation, samples were transferred to inactive polystyrene containers (6 ml). Samples were then allowed to decay for 20 minutes before counting at an appropriate geometry to limit the dead time to less than 20% on a Ge(Li) detector coupled to a 4096-channel analyser fitted with a pileup rejector amplifier. The detector has a resolution of 2.1 keV for the 1332 keV peak of $^{60}Co$, and a relative efficiency of 15%. Each sample was counted for 10 minutes. Gamma-ray analysis was repeated after 24 hours.

Long irradiations were performed simultaneously for samples and standards (100–200 mg) in similar containers as described above, but only transferred to the polystyrene vials after 3–4 days cooling period to allow the major $^{24}Na$ to decay significantly. High resolution gamma-ray spectrometry measurements were made after 7 and 28 days cooling intervals on a hyperpure germanium detector having a resolution of 1.8 keV for the 1332 keV peak and a relative efficiency of 20%.

The gamma-ray spectra for samples and standards were stored on a floppy disc or disc cartridge and were processed on an IBM 370 computer via a series of FORTRAN programs for NAA comparative analysis. Where a sample produced a gamma peak for a element not present in the standard, elemental concentrations were determined directly from the peak, using a computer program based on the nuclear parameters of HEFT[10] and the mathematical approach of HEFT and MARTIN[11] for element determination by absolute NAA.

Naturally occurring radionuclides were measured by a number of techniques including NAA, high resolution gamma-ray spectrometry and low level radiochemistry. For gamma-ray spectrometry, a low energy photon detector (resolution of 378 eV at 6.4 keV and 568 eV at 122 keV) was used in addition to those described above. The concentrations of $^{226}Ra$, $^{210}Pb$ and $^{40}K$ in coals and fly ash were determined by high resolution gamma spectrometry, the samples being sealed in plastic Petri dishes (60 × 15 mm) for 21 days and then counted for 24 hours. Quantitative determinations were made by comparing these results with data obtained on uranium ore stand-

ards whose daughters were in equilibrium (New Brunswick Laboratories reference materials 42–1 to 42–4) and which were mixed with either high purity graphite to approximate the composition of coals or high purity silica to duplicate the composition of fly ash.

Low level radiochemistry was also used to determine the concentration of $^{230}$Th, $^{226}$Ra, $^{226}$Ra, $^{210}$Pb and $^{210}$Po in coals and fly ash. Solids were dissolved by digestion with $HNO_3/H_2O_2$. If undissolved silica remained, the solutions were transferred to platinum dishes and evaporated to dryness with $HNO_3$/HF. Measurements of $^{230}$Th were performed by alpha spectrometry after successive separation and coprecipitation with $BaSO_4$, solvent extraction with Aliquat 336, then electrodeposition from sulphate medium onto stainless steel planchets.[12] The emanation technique of BLANCHARD[13] was used to determine $^{226}$Ra. The $^{228}$Ac daughter of $^{228}$Ra was used for the measurement of this radium isotope on a low level beta counter after its separation by solvent extraction with di(2-ethylhexyl) phosphoric acid followed by Aliquat 336.[14] Low levels of $^{210}$Pb were analysed by measuring the beta emission from the $^{210}$Bi daughter after solvent extraction with Aliquat 336, adding bismuth carrier, then precipitating from $H_2SO_4/HNO_3$.[15] Alpha gas-flow proportional counting was used to determine the $^{210}$Po after spontaneous deposition on a silver disc.[16]

## Results and discussion

Fifty samples of bituminous coals from Queensland and New South Wales, ten sub-bituminous coals from South Australia, thirty-five brown coals from Victoria and twenty fly ash samples from New South Wales and Queensland were analysed for trace elements.

### *Black coals*

The range and mean of the concentration of 37 elements found in bituminous coals and 36 elements in sub-bituminous coals are listed in Table 1. Results show that the average figures for Sc, V, Cr, Co, As, Se, Sr, Cs, Ba, La, Ce and Th in bituminous coals are up to 50% greater than previously published values,[1–6] whereas concentrations of the other 25 elements did not differ significantly.

Based on the results and recommendations of earlier studies and recommendations on the ecological and health aspects of trace elements in combustion,[17,18] the elements were classified into four categories: (a) of greatest concern – As, Se; (b) of moderate concern – V, Cr, Zn; (c) of minor concern – Na, Cl, Co, Br, Sr, Sb, Ba; and (d) radioactive elements – U, Th.

Table 1
Trace element concentration ranges and means in Australian bituminous and sub-bituminous coals ($\mu g \cdot g^{-1}$)

| Element | Bituminous coals | | Sub-bituminous coals | |
|---|---|---|---|---|
| | Range | Mean | Range | Mean |
| Na | 110 – 3600 | 570 | 3600 – 8100 | 5900 |
| Al (%) | 0.79 – 5.5 | 2.8 | 0.6 – 3.76 | 2.6 |
| Cl | 24 – 1900 | 360 | 2600 – 4900 | 3800 |
| K | 110 – 6900 | 1800 | 750 – 4000 | 2400 |
| Ca (%) | 0.04 – 1.8 | 0.4 | 0.81 – 1.63 | 1.09 |
| Sc | 2.4 – 13.9 | 5.1 | 1 – 7 | 4.9 |
| Ti | 480 – 4200 | 1700 | 650 – 3700 | 2600 |
| V | 13 – 90 | 32 | 4.3 – 49 | 28 |
| Cr | 2 – 56 | 12.5 | 54 – 41 | 29 |
| Mn | < 5.4 – 410 | 120 | 31 – 240 | 150 |
| Fe (%) | 0.09 – 3 | 0.83 | 0.49 – 1.88 | 1.4 |
| Co | 1.6 – 20 | 5.9 | 2.9 – 7.1 | 4.8 |
| Zn | 15 – 45 | 27 | 15 – 40 | 28 |
| Ga | 2.2 – 12 | 6.5 | 9.9 – 12.4 | 11.2 |
| As | 0.19 – 16 | 2.4 | 2.1 – 3.7 | 2.8 |
| Se | 0.19 – 2.6 | 0.8 | < 0.3 | |
| Br | 0.34 – 30 | 3.6 | 1.0 – 8.9 | 5.3 |
| Sr | 34 – 270 | 140 | 400 – 700 | 510 |
| Sb | 0.1 – 4.7 | 0.84 | 0.08 – 0.9 | 0.5 |
| Cs | 0.16 – 4.9 | 1 .2 | < 0.7 – 2.8 | 1.8 |
| Ba | 33 – 980 | 210 | 220 – 440 | 315 |
| La | 4.2 – 24 | 12 | 6.3 – 30 | 19 |
| Ce | 9 – 46 | 23 | 12.8 – 56 | 34 |
| Nd | < 3.5 – 23 | 11 | 4.3 – 21 | 15 |
| Sm | 0.8 – 4.6 | 2.3 | 0.8 – 3.2 | 2.2 |
| Eu | 0.17 – 0.80 | 0.4 | 0.12 – 0.48 | 0.24 |
| Tb | 0.11 – 0.78 | 0.34 | 0.09 – 0.43 | 0.3 |
| Dy | < 1.6 – 3.5 | 2.2 | 0.57 – 2.63 | 1.8 |
| Yb | 0.42 – 2.5 | 1.2 | 0.37 – 1.6 | 1.1 |
| Lu | 0.07 – 0.5 | 0.23 | 0.06 – 0.29 | 0.21 |
| Hf | 0.63 – 5.6 | 2.2 | 0.62 – 5.2 | 2.9 |
| Ta | 0.05 – 0.8 | 0.27 | 0.18 – 2.1 | 1.1 |
| W | 0.8 – 13 | 2.5 | 1.1 – 8.1 | 3.8 |
| Au | < 0.001 – 0.01 | 0.005 | <0.001 – 0.008 | 0.003 |
| Th | 0.57 – 7.9 | 3.7 | 2.1 – 17.4 | 9.2 |
| U | 0.29 – 2.5 | 1.3 | 0.4 – 3.8 | 2.1 |

The average level of As in these coals (2.4 $\mu g \cdot g^{-1}$) is markedly lower than the average values of 15 $\mu g \cdot g^{-1}$ for US coals[18] and 18 $\mu g \cdot g^{-1}$ for British coals.[19] Similarly, Australian bituminous coals are lower (0.8 $\mu g \cdot g^{-1}$) in Se than US (4.1 $\mu g \cdot g^{-1}$) and British (2.8 $\mu g \cdot g^{-1}$) coals.

For those elements of moderate concern, both Cr and Zn contents are lower than overseas coals whereas the concentrations of V are higher than for US but lower than the published values for British coals.

Only one element of minor concern, Sb, is present in Australian coals at concentrations lower than overseas coals. Although Cl and Ba concentrations are higher than for US coals, they are less then published data for the average world coal.[18] Australian black coals contain similar concentrations of Co and Br to overseas coals whereas Sr is similar to US coals but lower than the average world coal. Uranium concentrations in Australian coals are similar to overseas coals but the thorium values are higher than published data for either US coals[18] or British coals.[20]

*Fly ash from bituminous coals*

Table 2 summarises the trace element data for twenty fly ash samples obtained from NSW and Queensland bituminous coals. The average composition of the earth's crust[21] is included for comparison. Half of the 33 elements measured are concentrated with respect to the elemental composition of the earth's crust; these are Al, Ti, Zn, As, Se, Sb, Ba, La, Ce, Sm, Eu, Tb, Yb, Hf, W, Th and U. Of these, the largest enrichment fators are obtained for As, Se, Sb, Th and U; two being classified as those of greatest concern and two being radioactive.

*Brown coals*

Data for 35 brown coals samples from Victoria are given in Table 3. One fly ash sample from Victoria has been analysed for trace elements and is included in this table. Most brown coals are low in ash and this is reflected in the low concentrations of trace elements detected in these samples. Only Na and the halogens, Cl and Br, exceed the levels in bituminous coals. These data are in excellent agreement with earlier published data of BONE and SCHAAP.[7]

Although the trace element levels in brown coals are low, there are high concentrations of these elements in their fly ash. This is indicated by the data for the single fly ash sample which produced higher concentrations of Na, Cr, Mn, Co, Zn, As, Sr and Ba than the mean values for fly ash from the black coals listed in Table 2.

*Activity in black coals and their fly ash*

Sixteen samples of coal and fly ash from a number of coal-fueled power plants in Australia were analysed for their natural radioactivity. The results of these measurements are summarised in Table 4.

Table 2
Trace element concentration ranges and means in Australian fly ash from bituminous coals ($\mu g \cdot g^{-1}$)

| Element | Range | Mean | Crystal average[21] |
|---|---|---|---|
| Na | 580 – 1500 | 4000 | 28300 |
| Al (%) | 7.5 – 16.5 | 12.7 | 8.1 |
| K (%) | 0.55 – 2 | 1.3 | 2.6 |
| Ca (%) | 0.2 – 2 | 1.1 | 3.6 |
| Sc | 13 – 27 | 19 | 22 |
| Ti (%) | 0.31 – 0.96 | 0.64 | 0.44 |
| V | 82 – 180 | 130 | 135 |
| Cr | 32 – 61 | 50 | 100 |
| Mn | 55 – 2000 | 630 | 950 |
| Fe (%) | 0.45 – 10 | 3.1 | 5 |
| Co | 7.3 – 28 | 16 | 25 |
| Zn | 45 – 180 | 110 | 70 |
| Ga | 22 – 50 | 35 | |
| As | 2.7 – 8 | 5.1 | 1.8 |
| Se | 0.75 – 2.2 | 1.5 | 0.05 |
| Sr | 140 – 810 | 280 | 375 |
| Sb | 1.2 – 6.4 | 3.4 | 0.2 |
| Cs | 2.4 – 7.6 | 4.8 | 3 |
| Ba | 340 – 730 | 520 | 425 |
| La | 51 – 74 | 62 | 30 |
| Ce | 71 – 150 | 110 | 60 |
| Nd | 28 – 50 | 45 | |
| Sm | 8.9 – 14 | 11 | 6 |
| Eu | 1.5 – 3 | 2.2 | 1.2 |
| Tb | 0.7 – 2.3 | 1.7 | 0.9 |
| Yb | 4.2 – 9.8 | 7.8 | 3.4 |
| Lu | 0.62 – 1.3 | 1.1 | |
| Hf | 5.4 – 18 | 12 | 3 |
| Ta | 0.4 – 2.4 | 1.5 | 2 |
| W | 2.7 – 9.4 | 5.5 | 1.5 |
| Au | 0.003 – 0.04 | 0.01 | |
| Th | 14 – 29 | 24 | 7.2 |
| U | 5.2 – 9.3 | 7.3 | 1.8 |

Of the coal and fly ash samples subjected to radiochemical analysis (from three NSW power stations), two coals indicated some degree of disequilibrium between $^{238}U$ and $^{230}Th$. However, the lower members of this decay series were in secular equilibrium with the $^{238}U$ parent. This contrasted with the significant disequilibrium in fly ash samples. Measurement of the $^{232}Th$ and $^{228}Ra$ contents from the thorium chain revealed a marked disequilibrium for all three coal samples and a single fly ash sample.

Table 3
Trace element concentration ranges and means in Australian brown coals and fly ash ($\mu g \cdot g^{-1}$)

| Element | Brown coals | | Fly ash |
|---|---|---|---|
| | Range | Mean | |
| Na | 370 – 2200 | 950 | 16400 |
| Mg | < 700 – 4700 | 1900 | 52900 |
| Al | 70 – 13100 | 3000 | 109000 |
| Cl | 380 – 1300 | 780 | |
| K | 20 – 120 | 75 | 5750 |
| Sc | 0.01 – 5.5 | 0.4 | 12 |
| Ti | 45 – 2000 | 400 | 4760 |
| V | < 1 – 25 | 3.5 | 120 |
| Cr | 0.08 – 19 | 2.2 | 96 |
| Mn | 0.45 – 55 | 21 | 870 |
| Fe | 55 – 4900 | 1750 | 97000 |
| Co | 0.05 – 2 | 0.6 | 224 |
| Zn | 0.5 – 13 | 3.5 | 600 |
| As | < 0.1 – 1.3 | 0.2 | 80 |
| Se | 0.3 – 1.5 | 0.6 | |
| Br | 6.2 – 19 | 13 | |
| Sr | 2 – 250 | 82 | 740 |
| Sb | < 0.02 – 0.5 | 0.1 | 2.2 |
| Cs | < 0.02 – 0.06 | 0.04 | 6.3 |
| Ba | 1.7 – 170 | 63 | 2240 |
| La | 0.03 – 14 | 1.3 | 77 |
| Ce | 0.06 – 50 | 10 | 140 |
| Nd | < 0.3 – 13 | 1.8 | 35 |
| Sm | 0.004 – 2.9 | 0.3 | 11 |
| Eu | 0.002 – 0.6 | 0.06 | 2.4 |
| Tb | < 0.002 – 0.4 | 0.08 | 1.6 |
| Dy | < 0.02 – 2.1 | 0.2 | 11 |
| Yb | 0.005 – 1.3 | 0.2 | 5.7 |
| Lu | 0.001 – 0.3 | 0.03 | 0.7 |
| Hf | 0.006 – 1.1 | 0.3 | 5 |
| Ta | < 0.03 – 0.7 | 0.1 | 1.2 |
| W | < 0.1 – 1.2 | 0.5 | 3.5 |
| Th | < 0.008 – 3.5 | 0.3 | 9.9 |
| U | < 0.06 – 1.2 | 0.4 | 3.6 |

These results are not consistent with the assumption that there is always secular equilibrium between the radioactive daughters of each decay series. Therefore, the source terms used in most assessments of radioacitve exposures to the general public from coal-fired stations are not applicable.

Table 4
Concentrations of natural radionuclides in Australian coals and fly ash ($mBq \cdot g^{-1}$)

| Sample | $^{238}U$ | $^{230}Th$ | $^{226}Ra$ | $^{210}Pb$ | $^{210}Po$ | $^{232}Th$ | $^{228}Ra$ | $^{40}K$ |
|---|---|---|---|---|---|---|---|---|
| Vales Point | | | | | | | | |
| Coal | 28 | 68 | 24 | 30 | 28 | 27 | 64 | 72 |
| Fly ash | 114 | 130 | 110 | 150 | 85 | 130 | 78 | 434 |
| Liddel | | | | | | | | |
| Coal | 26 | 21 | 21 | 33 | 18 | 29 | 11 | 112 |
| Fly ash | 78 | 70 | 68 | 85 | 58 | 85 | 67 | 170 |
| Tallawarra | | | | | | | | |
| Coal | 22 | 39 | 19 | 20 | 16 | 17 | 48 | 81 |
| Fly ash | 64 | 77 | 59 | 60 | 33 | 57 | 58 | 263 |
| Munmoorah | | | | | | | | |
| Coal | 21 | | | | | | 23 | 67 |
| Fly ash | 107 | | | | | | 126 | 430 |
| Wallerawang | | | | | | | | |
| Coal | 25 | | | | | | 27 | 140 |
| Fly ash | 89 | | | | | | 113 | 615 |
| Wongawilli | | | | | | | | |
| Coal | 21 | | | | | | 20 | 63 |
| Fly ash | 81 | | | | | | 77 | 220 |
| Leigh Creek | | | | | | | | |
| Coal | 47 | | | | | | 69 | 23 |
| Fly ash | 102 | | | | | | 138 | 430 |
| Blackwater | | | | | | | | |
| Coal | 9 | | | | | | 11 | 43 |
| Fly ash | 76 | | | | | | 57 | 350 |
| Earth's Crust | 22 | | | | | 29 | 803 | |

Although controversy still surrounds these source terms and the models used for dispersion, inhalation and ingestion,[22,23] most researchers agree that dose rates from power plants are much lower than recommended guidelines. However, more accurate measurements of the individual radionuclides are necessary if the long-term environmental effect of radioactive emissions are to be determined.

## Conclusion

Trace element data for a wide range of Australian coals and fly ash gathered over the last eight years both independently and in collaborative research projects have been summarised. Results show that average figures for Sc, V, Cr, Co, As, Se, Sr, Cs, Ba, La, Ce and Th in bituminous coals are up to 50% higher than previously published. However, the data for brown coals are in close agreement with literature values. For those elements of greatest concern in power station emissions, As and Se, their levels in Australian coals are markedly lower than for US and British coals.

Although the trace element content of brown coals is significantly lower than levels found in bituminous coals, fly ash concentrates elements to similar or higher levels than found in fly ash derived from black coals.

Radiochemical and instrumental analyses of selected radionuclides in the uranium and thorium decay chains for some coals and fly ash samples from Australian coal-fired stations indicates marked disequilibrium for some sections of the decay chain.

## References

1. H. R. BROWN, D. J. SWAINE, J. Inst. Fuel, 37 (1970) 422.
2. R. E. PORRITT, D. J. SWAINE, Mercury and Selenium in Some Australian Coals and Fly Ash. Preprint Inst. Fuel Conf., Sydney, 1976, Paper 18, p. 9.
3. D. J. SWAINE, Combustion of Pulverised Coal – The Effect of Mineral Matter (Conf.), I. M. STEWART, T. F. WALL (Eds), Newcastle, 1979, p. W3.
4. D. J. SWAINE, Energ. Res. Technol., 4 (1982) 18.
5. R. A. CAHILL, J. C. MILLS, Proc. Australas. Inst. Min. Metall., 285 (1983) 39.
6. L. DALE, J. J. FARDY, E. J. CLAYTON, Trace Element Abundance Data for Coals using Instrumental Methods of Analysis., Proc. Intern. Conf. on Coal Science, Sydney, 1985, p. 857.
7. K. M. BONE, H. SCHAAP, Trace Elements in Latrobe Valley Brown Coal: Environmental and Geochemical Implications, 30/80/15, State Electricity Commission of Victoria, 1983.
8. R. J. BAYLISS, H. M. WHITE, Air Wat. Pollut. Int. J., 10 (1966) 819.
9. M. B. COOPER, I. S. LEITH, Radiological Impact of the Operation of Coalfired Power Stations within Australia, ARL/TR-50, Australian Radiation Laboratory, 1983.
10. R. W. HEFT, A Consistent Set of Nuclear Parameters for INAA, UCRL-80286, Lawrence Livermore Laboratory, California, 1978.
11. R. W. HEFT, NADAC and MERGE – Computer Codes for Processing Activation Data, UCRL-52249, Lawrence Livermore Laboratory, California, 1977.
12. C. W. SILL, K. W. PUPHAL, F. D. HINDMAN, Anal. Chem., 46 (1964) 1725.
13. R. L. BLANCHARD, An Emanation Method for Determining Small Quantities of Radium-226, Environmental Health Series, Environmental Health Series 999-RH-9, U. S. Department of Health, 1964.
14. H. G. PETROW, A. COVER, W. SCHIESSLE, E. PARSONS, Anal. Chem., 36 (1964) 1600.
15. H. G. PETROW, A. COVER, Anal. Chem. 37 (1965) 1659.
16. W. W. FLYNN, Anal. Chim. Acta, 43 (1968) 221.

17. M. HEJT, A Review of Current Information and Some Ecological and Healt Related Aspects of the Release of Trace Elements into the Environment Associated with the Combustion of Coal, HASL-320, New York Health and Safety Laboratory, New York, 1977.
18. Trace-element Geochemistry of Coal Resources Development Related to Environmental Quality and Health, National Academy Press, Washington, D. C. 1980.
19. M. Y. LI, Trace Element from Coal Combustion – Atmosphere Emissions, ICTIS/TROS, International Energy Agency, 1979.
20. L. SALMON, A. E. R. TOUREAU, A. E. LALLY, Sci. Total Environ., 35 (1984) 403.
21. S. R. TAYLOR, Geochim. Cosmochim. Acta, 28 (1964) 1273.
22. W. C. CAMPLIN, Coal-fired Power Station – The Radiological Impact of Effluent Discharges to Atmosphere, NRPB-R107, National Radiological Protection Board, Harwell, 1980.
23. J. O. CORBETT, The Significance of Lead-210, Polonium-210 and Proactinium-231 in Emissions from Coal-fired Power Stations: A Comparison with National Environmental Source, RD/B/506, Central Electrity Generating Board, Berkeley Nuclear Laboratories, 1981. [PB]