PHYSICS IN THE WEST

Stable Isotope Abundance Studies in W.A.

P. M. Jeffery, Department of Physics, University of Western Australia

INTRODUCTION

In 1950 a visit to the Physics Department of the University of Western Australia by the present Governor of South Australia, Sir Marcus Oliphant, triggered off a research program which altered the careers of many of the students who were subsequently trained in the Department. During his visit, Sir Marcus stressed the need for an Australian research program in geochronology, pointing out that this country was very backward in this regard considering that it was believed to contain some of the oldest exposed rocks on earth.

Two of the younger staff members at that time, A. H. Morton (ANU)* and the author were so impressed with Sir Marcus' suggestion that they decided to initiate a research program whose objective was to solve radioactive dating problems using mass spectrometric techniques.

It seems incredible, in retrospect, that this project could have been started when neither equipment nor funds were available, but the enthusiasm of youth made it possible to ignore such problems as having to build a mass spectrometer from water pipe and copper tube, build diffusion pumps to evacuate the spectrometer and construct all the electronics. Before embarking on what was regarded as the difficult research area of radioactive dating it was decided to select a 'simpler' problem for investigation in order to acquire skill in mass spectrometric measurements. The first problem selected was a study of the variation of the $^{12}C/^{13}C$ ratio in samples of carbon from various natural sources. Possible variations in this ratio had been previously reported. *Nier and Gulbransen, 1939.* And so mass spectrometry research began in Western Australia!

THE 1950's

In the early 1950's information transfer between scientists in Perth and the rest of the world was somewhat meagre so that it is not surprising that the Perth mass spectrometry group did not know that the carbon isotope abundance problem was also being studied by Harmon Craig at the University of Chicago under the watchful eye of Nobel prize winner, Harold C. Urey. This piece of information was not available until Craig published his classic carbon paper *Craig, 1953.*

The home-made mass spectrometer first produced a reasonable ion beam on 27 October, 1952 and shortly afterwards an attempt was made to distinguish, on isotopic grounds, between samples of CO_2 from the local brewery and from limestone. Despite some initial successes the data from the spectrometer was not satisfactory because of continual vacuum problems ('glyptal' covered water piping is not a high grade vacuum housing!) and general beam instability, so that very reluctantly it was decided to abandon the entire project. Luckily an eleventh hour bid to obtain funds by the new head of the Department of Physics, Professor C. J. B. Clews, was successful when the Carnegie Institution of Washington granted an amount of $7500 to the Department for research into age determination problems.

This grant was made on the recommendation of the Director of the Carnegie Institution's Department of Terrestrial Magnetism, Dr Merle A. Tuve, and a senior member of his staff, Dr L. T. Aldrich. Tom Aldrich had already played an important part in the development of the Rb/Sr method of age determination and was well aware of the need for isotope abundance studies to be supported in Australia.

The Carnegie funds provided the Perth group with a new sixty degree Nier type mass spectrometer and also permitted the original home-made spectrometer to be upgraded by the replacement of its waterpipe sections with fabricated stainless steel. Such exotic devices as a chart recorder, to replace a wall galvanometer, and commercial diffusion pumps were all very acceptable. In spite of these improvements in equipment however, ion currents were still being measured using 'acorn' 954s as electrometers (for those who can remember).

With another spectrometer at their disposal the group was able to proceed with two distinct lines of research in the latter half of the fifties.

The new Carnegie mass spectrometer was commissioned by D. Greenhalgh (University of WA) who then applied it to the measurement of the ages of selected Australian rocks and minerals using the uranium to lead decay scheme. The isotopic composition of the lead was measured by ionising it in the form of lead-tetramethyl *Greenhalgh and Jeffery, 1950.* Some years later some of the data in this paper provided fuel for an interesting argument in 'Nature' concerning the age of the base of the Cambrian period in the geological time scale *Holmes, 1960; Wetherill, 1960.*

W. Compston (ANU) took over the carbon investigation from C. G. Cole and with the other members of the group completed the initial study of the isotopic composition of carbon in nature thus verifying the results of Craig who had published his results some two

years earlier *Jeffery, et al., 1955*. Compston continued with the carbon work and during a post doctoral fellowship at the Californian Institute of Technology, published again on this topic *Compston, 1960*.

The award of a Carnegie Fellowship to allow the author to carry out research at the Department of Terrestrial Magnetism in Washington DC in 1955 provided an important means of diffusing knowledge of the latest techniques in geochronology between Perth and the United States. Indeed the first Rb/Sr ages to be made on Australian rocks resulted from this visit *Jeffery, 1955*. It became apparent, even at this early stage, that the Rb/Sr method would surpass the U/Pb technique as a means of determining accurate ages in Precambrian rocks so it was not surprising to find the direction of research in Perth turning away from U/Pb dating to the Rb/Sr and K/Ar dating methods.

Whilst the Rb/Sr method was being developed, a series of K/Ar ages were measured on Australian rocks so that ages found by the two different decay schemes could be compared.

By this time (latter part of the 1950's) the validity of the Rb/Sr method was being seriously questioned by geologists since again and again discordant ages were being reported from mineral concentrates extracted from a single rock sample. In an attempt to solve this problem, rock samples from Boya Quarry near Perth were collected and careful Rb/Sr studies made on them, both as total rock samples and on each rock's mineral concentrates. The Rb/Sr group at this time comprised Compston, who had returned to the Physics Department early in 1959 as a staff member, G. H. Riley (CSIRO) and the author, together with the able technical assistance of J. F. Smith (La Trobe University). Riley later presented a very interesting thesis on the theory and techniques of Rb/Sr geochronology *Riley, 1961*.

The data from the Boya study led to the development of a theoretical model to account for the effects of metamorphism on the Rb/Sr ages determined from rock sampels and their mineral concentrates. This model *Compston and Jeffery, 1959* represented a major advance in the understanding of discordant Rb/Sr age patterns.

[Interested readers are referred to the editor's comments in 'Geochronology' (1973, pp 244-247) published by Dowden, Hutchinson and Ross in the 'Benchmark Papers in Geology Series'.]

In the latter part of the 1950's, in addition to radioactive age studies obtained by using the Carnegie mass spectrometer, the original instrument was rebuilt so that the isotopic composition of potassium in nature could be measured. Interest in potassium arose from an early report *Lasnitzki and Brewer, 1942* that carcinogenic tissue contained potassium which differed isotopically from normal potassium. Using a triple source solid sample mass spectrometer B. R. F. Kendall (Penn. State University) was able to carry out high precision measurements on potassium, demonstrating that for the range of samples he investigated the isotopic composition of potassium was constant to within a few permils *Kendall, 1960*. This study was supported in part by Dr K. W. Starr (NSW) and the NSW State Cancer Council.

Two aspects of Kendall's work are of particular interest. First, the controversy which arose over whether he had in fact observed isotopic variations in the potassium from carcinogenic tissue [see for example, comments by *Starr, 1962 and Kendall, 1963*] and second, how unfortunate he was not to have had the opportunity to study potassium in meteorites. Anomalous potassium in meteoritic material was observed shortly after Kendall had completed his potassium study *Voshage and Hintenberger, 1959*. An attempt to obtain an iron meteorite sample for Kendall failed due to administrative difficulties!

Problems of this nature resulted in the formation, in the early 1960's of the Western Australian State Meteorite Advisory Committee which advises the Board of the Western Australian Museum on all matters concerning meteorites, including request from investigators for samples.

THE 1960's

By the late 1950's geologists were beginning to ask for radioactive ages 'on demand' and it was becoming obvious that the modest facilities in Perth would not be able to provide the data which would be needed. At a meeting at Lucas Heights sponsored by the Australian Institute of Nuclear Science and Engineering it was decided informally that a major age determination centre should be established in Australia and that the Australian National University was probably the best venue. The centre was, in fact, established at the ANU with assistance from the Bureau of Mineral Resources.

By 1961 research in the area of geochronology had ceased in the Perth group which now focused its attention on other isotopic problems associated with stable isotope abundances.

A new noble gas mass spectrometer was obtained from Nuclide Analysis Associates and a project aimed at detecting the reaction products of cosmic ray negative muons in surface rocks commenced. The specific reaction involved was: alkali target nucleus (μ^-, x n) noble gas isotope product. Due to the fortuitous installation of a PDP-6 computer in the Physics Department building it was possible for J. Hagan, who was working on the muon project, and J. de Laeter (WAIT), to bring into operation the first on-line computing system to be used for natural isotope abundance studies *Hagan and de Laeter, 1966*. Concurrently with the muon work, a solid source mass spectrometry project was begun to study the isotopic composition of elements with high ionization potentials.

Tin was the first element chosen for investigation due to its clear cut mode of formation by the R, S and P nuclear processes. The current theoretical model implied that inhomogeneous mixing might have occurred between the products of these three processes so it was possible that variations in the isotopic composition of tin might be present in tin samples from different environments. No isotopic variations were observed in a wide range of meteoritic and terrestrial samples *de Laeter and Jeffery, 1967*.

Following the tin study, K. Rosman (WAIT) examined the isotopic composition of zinc in a somewhat similar manner. Rosman, however, was able to introduce a high degree of precision into his data by using a liquid nitrogen cooled ion source to suppress the general mass spectro-

meter background in his instrument. Zinc was of special interest since it was hoped that it might be possible to observe isotopic fractionation effects which had been caused by the particular mode of introduction of zinc into ore bodies. Despite the fact that Rosman was not able to positively state that he had observed fractionation effects in the vicinity of a zinc ore body he concluded that only a factor of ten improvement in measuring precision was required to bring fractionation effects within the realm of measurable quantities. This gain in precision should be well within the limits of modern techniques *Rosman, 1972*.

THE 1970's

In 1968 the Physics Department of the Western Australian Institute of Technology introduced a program of isotope abundance studies under the direction of de Laeter (now Dean of Applied Sciences). At the instigation of Compston and A. F. Trendall (State Geological Survey) part of the WAIT program was directed towards Rb/Sr dating determinations which were by then badly needed to supplement those from the ANU.

An AEI MS 12 solid source mass spectrometer was installed for this work but shortly after its installation a disastrous fire in the mass spectrometer laboratory all but ruined the spectrometer so that a new MS 12 had to be installed in 1971. The original instrument was finally rebuilt and brought back into operation so that two MS 12's became available for training and research purposes.

Rosman joined the WAIT staff in 1971 and over the last few years the group has produced a steady flow of research papers on such topics as Rb/Sr dating, Cd isotope abundance studies, fission product studies and the role of lutetium in S–process chronology *McCulloch, 1974*.

During the 1970's the WA University group directed its efforts to studying the isotopic composition of the noble gases in terrestrial materials in an attempt to understand how the noble gases evolved in the earth during its early hisotry and how the present day atmospheric isotopic composition of the noble gases was produced. Xenon and neon have come in for most attention since these two elements appear to offer the best opportunity of detecting ancient changes in isotopic composition. In principle, neon should be the best stable element to use since its isotopic composition varies more than any other stable element when samples from selected sites are considered [see for example: *Jeffery and Zadnik, 1975*]. Neon trapped in a variety of rock types, such as oceanic basalts and ultramafic rocks are being studied and a model of neon evolution during the history of the earth is being developed.

Neon isotopic measurements have been facilitated by the use of a cryostatically cooled 'cold finger' which is coupled directly to a static noble gas mass spectrometer. By varying the temperature of the cold finger between 4 K and 30 K it is possible to 'manipulate' the neon and optimise the sample to background ratio.

CONCLUSION

Carrying out research in an undefined area of knowledge lying between geology and physics was not easy in the early days of isotope abundance research in this State. Most geologists looked askance at the 'numbers' that were being published in the literature as 'ages' and under these circumstances the active participation of geologist Professor A.F.Wilson (University of Queensland) was greatly appreciated by the physicists in the group *Wilson, et al, 1960*. On the other hand the Physics Department considered the whole research area as rather foreign to physics and perhaps a little *de trop* and in this academic climate it was sometimes rather difficult to maintain the group's enthusiasm.

The present day attitude is, of course, very different. Geologists are conscious of the important information concerning geological problems that can be obtained from isotopic studies, particularly in relation to orogenesis and the formation of ore bodies. Even physicists accept that isotopic data from natural samples has its uses when they see the vast quantity of information for nuclear studies that has resulted from isotopic studies on meteoritic and lunar material.

In more recent years the active participation of the Western Australian Institute of Technology in isotope abundance studies has increased the State's research output in this area considerably. In addition, local geologists will almost certainly take a more active part in future isotope studies as a result of the influence of Professor P. G. Harris, who has recently been appointed to the Chair of Geology in the University of Western Australia.

With the excellent cooperation that exists between the various people involved, it seems highly probable that the stable isotope abundance researches started in this State so long ago will develop into a major area of research in Western Australia.

An apology: In a brief review of this nature it is not possible to mention the many students and colleagues who have contributed so much to the success of mass spectrometry research in this State over the years, nor to mention the enormous amount of assistance that has been provided by the workshop staffs at both the Institutions involved. Generally only people who have gained higher degrees from research studies on isotope abundances have been mentioned while references to published works have been restricted to a few relevant papers.

References

Compston, W. (1960). *Geochim. Cosmochim. Acta* 18:1.
Compston, W. and Jeffery, P. M. (1959). *Nature* 184:1792.
Craig, H. (1953). *Geochim. Cosmochim. Acta* 3:53.
de Laeter, J. R. and Jeffery, P. M. (1967). *Geochim. Cosmochim. Acta* 31:969.
Greenhalgh, D. and Jeffery, P. M. (1959). *Geochim. Cosmochim. Acta* 16:39.
Hagan, J. and de Laeter, J. R. (1966). *J. Sci. Instr.* 43:662.
Holmes, A. (1960). *Nature, Lond.* 187:34.
Jeffery, P. M. (1955). *Geochim. Cosmochim. Acta* 10:191.
Jeffery, P. M. and Zadnik, M. G. (1975). *Proc. Third Australian Conference on Mass Spectrometry, A.N.U.*
Jeffery, P. M., Compston, W., Greenhalgh, D. and de Laeter, J.R. (1955). *Geochim. Cosmochim. Acta* 7:255.
Kendall, B. R. F. (1960). *Nature* 186:225.
Kendall, B. R. F. (1963). *Aust. J. Science* 25:437.
Lasnitzki, A. and Brewer, A. K. (1942). *Cancer Research* 2:494.
McCulloch, M. T. (1974). *Master's degree thesis. Western Australian Institute of Technology.*
Nier, A. O. and Gulbransen, E. A. (1939). *J. Amer. Chem. Soc.* 61:697.
Riley, G. H. (1961). *Ph.D. thesis. University of Western Australia.*
Rosman, K. J. (1972). *Geochim. Cosmochim. Acta* 36:801.
Starr, K. W. (1962). *Aust. J. Science* 24:375.
Voshage and Hintenberger (1959). *Zeits. Naturforschung* 14a:828.
Wetherill, G. W. (1960). *Nature* 187:35.
Wilson, A. F., Compston, W., Jeffery, P. M. and Riley, G. H. (1960). *Journ. Geol. Soc. Aust.* 6:179.