DATING THE PAST WITH RADIOACTIVITY

Very soon after the discovery of radioactivity, measurements of the heavy radioactive elements existing in nature were undertaken to determine the age of rocks (Rutherford 1904), but only when Professor Willard Libby in an evening lecture during the first United Nations Conference on the Peaceful Uses of Atomic Energy in Geneva in 1955 described how he and his research team had evolved the carbon-14 dating technique did research on radioactive dating receive a real impetus. Since that time laboratories in many countries have made rapid progress and geologists, geophysicists, geochemists and archaeologists have joined forces with nuclear scientists to perfect the techniques and widen the range of its applications.

Professor Libby had first worked out the radiocarbon dating technique in theory; his team had found and extracted radiocarbon from sewage slushes in Baltimore, conducted a worldwide assay of woods and developed the necessary counting procedures and equipment. They then conducted actual datings, both of samples of known age, which proved that the method "worked", and of objects of hitherto unknown age. He first published his findings in 1952. Interest in the 1950's centred mainly round the Libby method using carbon-14 as the "chronometer" and the dating of archaeological and other relatively recent objects. Lately the interest and techniques have again shifted to the age determination of the very distant past down to the very origin of our solar system and to the application of the radioactive dating method to problems in the earth sciences, such as the movements of air and water masses, the origin and nature of rock formations and the history of meteorites.

This shift of interest was clearly discernible at a five-day international symposium organized by IAEA in Athens, 19-23 November 1962, which, although also dealing with carbon-14 and archaeology, mainly discussed geological and meteoritic problems. The symposium was co-sponsored by the Joint Commission on Applied Radioactivity of the International Council of Scientific Unions (ICSU); the Greek Atomic Energy Commission acted as host.

Methods and Possibilities

The general principle of radioactive dating is based on measuring the decay of certain given radioisotopes and/or the build-up rate of other radioisotopes in the decay chain. To obtain results with only small margins of error it is necessary to know as exactly as possible the half-life of the radioisotope concerned and to be sure that in the sample to be measured the activity of the radioisotope has not escaped by other means than the natural decay. One must also be able to measure small amounts of radiation, which is difficult because of the background radiation that is always present from other sources. In radioactive dating one uses both natural radioisotopes with very long half-lives which have survived since the formation of the earth and the solar system (nucleosynthesis) and - more recently - radioactive isotopes formed by the cosmic ray bombardment both directly on the surface of the earth and in the atmosphere and beyond.

The half-life of carbon-14, formed in the atmosphere by the neutrons from cosmic rays being absorbed by nitrogen and transformed into this radioactive isotope of carbon, had been assumed in the earlier days of radiodating to be about 5600 years. In a paper by two Swedish research workers from Uppsala University (Olsson and Karlén) it was pointed out that recent investigations at Uppsala, the US National Bureau of Standards and Aldermaston (UK) seemed to agree that the half-life value more correctly should be set at 5700 years. The determination of the half-life of carbon-14 is particularly important as this radioisotope is the main tool for measurements up to 50 000 years back and consequently the most used method in archaeology. For dating samples of much greater age, radioisotopes occurring in nature are used. A paper by a German and a Swiss scientist (Herr and Hirt) reported the use of rhenium-187 with a half-life of 48 million years to measure the age of iron meteorites. The results corresponded very well with the general assumption, based on the results from other radioactive dating methods, that meteorites and the crust of the earth itself were formed approximately at the same time, some 4500 million years ago. Before the use of the radioactive dating method there were no means of determining the age of the formation of the earth with any degree of reliability and even this method has only recently become reasonably precise.

Some of the elements in the uranium decay chain emit alpha particles which make up nuclei of helium. The helium can be measured by mass spectrometry and a report on this method was presented in a paper by two scientists from the University of Arizona (Damon and Green). One main difficulty in this method is to determine the loss of helium in the sample through reasons other than the natural decay, as for instance through "escape" caused by the high emission velocity of the alpha particles, radiation damage and heat diffusion.

The thermoluminescence method was discussed in a couple of papers. This is based on measuring the glow of a sample when exposed to heat. Certain elements undergo electronic rearrangement when exposed to radiation, with the electrons accumulating in "traps". This rearrangement is stable at the normal temperature of the environment of the element, but when subjected to heat the electrons recover their pre-radiation position and in this process they emit light which can be measured. The intensity of the light is in direct proportion to the original exposure to radiation.

The first commonly used method of dating uranium-bearing rocks was to measure the build-up of lead-206, which is a stable isotope at the end of the uranium disintegration chain, and to compare this with the amount of uranium-238 contained in the sample. The same principle can be used in comparing the amount of lead-206 with other preceding radionuclides in the chain. Experiments reported by Yugoslav scientists (Gojkovic, Deleon and Cervenjak) showed that by determining the lead-206/lead-210 ratio results were obtained which corresponded well with results from other methods.

One of the main tasks in present-day radioactive dating is to develop counting techniques and equipment which can be used to measure the very low levels of activity usually involved. H. Oeschger, of the Physical Institute of the Berne University, surveyed present low-level radiation detectors (gas and scintillation counters) and described methods for the suppression of the background radiation. He was of the opinion that although currently used techniques were adequate for samples not older than 50 000 years, improvements in techniques and devices - which seemed quite feasible - would open up new promising possibilities. In that case, one might, for instance, use carbon-14 for dating up to 60 000 to 70 000 years, one might not have to collect as large samples as at present, and the study of oceanic circulation processes and of radioactivity in terrestrial materials would be eased. It was also most probable, he said, that with higher sensitivity in counting techniques unknown radioisotopes with very long half-lives would be detected.

By using a sensitive mass spectrometer one can obtain reliable results with the potassium-argon dating method even when the samples have low concentration or are of relatively short age. This was reported by two German scientists (Gentner and Lippolt) who had experimented with fossil bones from the Black Forest and volcanic minerals from the Eifel Mountain.

Geology and Applications in Meteorology and Hydrology

It became clear in the course of the Athens symposium that in the last few years the radioactive dating method has not only sharpened its sensitivity, but also increased its time dimensions in both directions. The progress in methods to determine the ratio between uranium-234 and uranium-238, for instance, has made it possible to make more precise age determinations of samples aged 100 000 to more than one million years (Thurber). At the other end of the scale, it was shown that the use of lead-210, formed in the atmosphere through the decay of the short-lived gas radon-222, is useful for dating geochemical processes which have taken place within the last hundred years or so. This was shown in a paper from California University (Goldberg), which reported on studies of Greenland glaciers and the Colorado and Sacramento rivers. These studies also indicated that the composition of river waters is determined not only by the geology of the drainage basin, but also by inorganic or chemical reactions occurring in the waters.

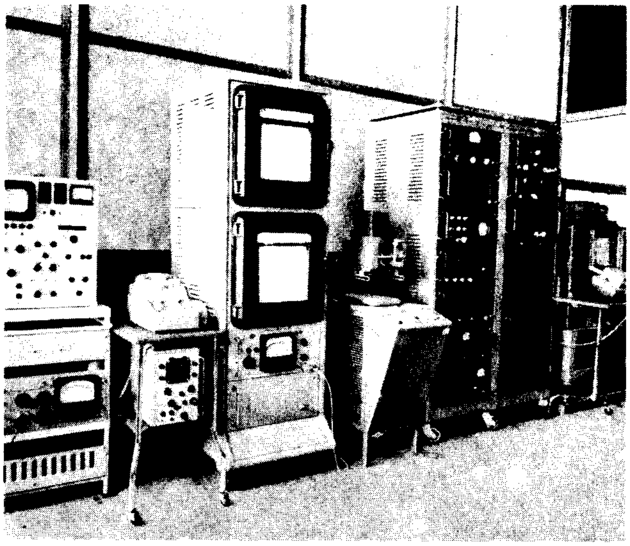
The use of carbon-14 and other radionuclides formed by cosmic radiation in finding out about the large-scale movements of air in the atmosphere were discussed. In an Indian paper (Lal) it was stated that it was now possible to determine the rates of accumulation of extra-terrestrial dust on the earth in the last few million years by measuring the aluminium-26 and manganese-53 concentrations in these sediments. At present it is estimated that one thousand tons of such dust reaches the earth every day.

According to the same paper, radioactivity formed on the surface of the earth by cosmic radiation can also be used for erosion studies, although normal erosion rates are so high that surfaces exposed at present would have been tens of meters underground some hundred thousand years ago. This method is therefore suitable only for measuring relatively recent erosion rates. Studies of cosmic ray-produced radioisotopes have also been found useful in the study of oceanic circulation processes and the chronology of oceanic sediments.

It was, for instance, reported (Bien, Rakestraw and Suess) that the estimated south-north speed of the water in the Pacific Ocean amounts to 0.04 cm/second, i.e. it takes approximately 400 years for the water at 40^o south latitude to reach the 40^o north latitude.

K.O. Münnich from Heidelberg University discussed the measuring of artificially produced carbon-14 in the high atmosphere; he was of the view that the old "box-model" to explain the slow intermixing of air masses both north/south and between altitudes should be replaced by a "continuous model" based on the diffusion theory.

The conventional method of determining rock ages has been to measure separated radioisotopes from a sample. This method has the defect that some decay products from the isotope to be measured have been lost during the cooling time of the rock. Strontium-87, a daughter isotope of rubidium, seems, however, to remain in the rock and a method of measuring a whole-rock sample, rather than the separated isotopes, for its concentration of rubidium



Multichannel spectrometers for alpha and gamma ray measurements used at the French nuclear centre Fontenay-aux-Roses

in relation to strontium therefore seems promising. A paper from the Massachusetts Institute of Technology (Hurley, Fairbairn, Faure and Pinson) reported on the development of this method. Its success depends partly on determining the initial strontium-87/strontium-86 ratio in the rock at the time of its formation. The authors reported that for most rocks this value is close to 0.708 and this value can be used without having to make individual measurements for each sample.

Results of age determination of uranium minerals in granite rock formations in Northern Bavaria showed that the uranium had been formed much later than the general deposits of the area, probably by uranium dissolved in acid water flowing through alkaline limestone and thus falling out as uranium salt (Lenz and Wendt).

The potassium-argon dating method has been used to determine the age of tektites (Zähringer), which are found in three main areas: North America, Czechoslovakia and Australia. The australites seem to be very young, about 600 000 years old, the Czech tektites about 15 million years, and those from North America more than 30 million years. Tektites are spheric, dark-greenish, glassy objects, commonly assumed to be of cosmic origin. Professor Vinogradov (USSR), however, pointed out in the discussion that available physical and chemical data, particularly the high potassium and uranium content similar to that in acid earth-formed rocks, would lead to the conclusion that tektites were of terrestrial genesis.

The discrepancies found in using the radioisotopic method for geological dating depend very largely on temperature changes and base exchange by ground water, stated J.L. Kulp of Columbia University. He reported on recent experimental work on the base exchange properties of biotite, where age determination results had been particularly discrepant. His conclusions were that thermal effects rarely caused discrepancies of more than 10 per cent, whereas ground water exchanges would explain the differences that have resulted from using the rubidium-strontium or potassium-argon methods. These discrepancies have their value, however, in providing valuable information on a region's geochemical history.

A.P. Vinogradov, of the Biogeochemical Institute of the Soviet Academy of Sciences, reported on carbon-14 dating of peat deposits along the lower Indighirka river in East Siberia and on the dating of mammoths (12 000 years) and the change from fresh to brackish water in the Black Sea (8 000 years).

Meteorites

The determination of the age of the formation of meteorites, the time of their fall to earth and the production of radioactivity in man-made satellites was the subject of a number of papers. Special attention was paid to determination of the intensity of cosmic rays in time and space and the formation of penetrating particles in space.

A United States paper (Fisher) stated that data obtained from using lead as a dating tool showed that stone meteorites had an age of 4500 million years, that is, the same as the crust of the earth. The evidence in respect of iron meteorites, using the potassium-argon ratio method, sets their age at up to almost twice as much. There are no other data that support the commonly held assumption that iron and stone meteorites and the earth itself were formed at the same time. With the results at present available one must conclude that iron meteorites are much older than stone ones.

The same techniques that are used for measuring the age of meteorites has also been applied lately to the fragments of man-made satellites. A paper from a research group at the Smithsonian Astrophysical Observatory (Fireman, de Felice and Tilles) gave a survey of the work carried out there. The paper stressed that the production of radioactivity in satellites is quite different from its production in meteorites. In meteorites radioactivity is uniform; in satellites considerable variations are observed because of their excentric trajectories and their interaction with the Van Allen radiation belt. Samples of some of the Discoverer satellites, it was reported, had been analysed for tritium in lead, steel and aluminium. These investigations have a direct bearing on the plans for manned space travel.

Other papers described techniques for the use of carbon-14 or other radionuclides produced by cosmic rays to date the time of the fall to earth of meteorites. The results so far available tend to prove that many meteorites have had longer "terrestrial lives" than has hitherto been assumed. It was generally agreed that carbon-14 provided a good tool for dating meteorites and that further developments in this technique would increase its accuracy and range, which might prove particularly interesting in dating prehistoric meteorite craters.

To obtain the same kind of information that has resulted from the study of recently fallen meteorites by "direct" observation would require a two-year long space travel in an appropriately constructed space vehicle.

The spatial and time distribution of cosmic radiation intensity were dealt with in several papers. In a paper from Brookhaven National Laboratory (Davis, Steenner and Schaeffer), a method of comparing the activity of a short-lived radioisotope with a long-lived one to establish the spatial density of cosmic radiation was described. Argon-37, with a half-life of 35 days, and argon-39, with a half-life of 325 years, were used. The results have led to the conclusion that the present cosmic ray intensity at one Astronomical Unit (the earth orbit) is within 15 per cent of the intensity at a distance of several Astronomical Units. The results also showed that the production of radionuclides by cosmic rays is independent of the meteorite's orbit. Special study had also been made of the effects of solar flares, as for instance the one that occurred in November 1960, comparing the argon-37 content of stainless steel samples from Discoverer 17, which was launched during the intense solar flare in November 1960, with samples from Discoverer 18 and 26.

Another paper from the United States (Goel and Kohman) dealt with variations in cosmic ray intensity in time. Cosmic rays are generally assumed to originate in stars or stellar systems and as these vary with time and position within the galaxy, long-term variations would be expected. The paper concludes, on the basis of chlorine-36 and argon-36 investigations, that cosmic ray intensity has been decreasing over the past few hundred million years. The research also indicates that erosion of the meteorite during its space travel has been "general and significant".

These conclusions were different from those reached in two other papers. In a paper by two scientists from the Max-Planck Chemical Institute, Mainz, Federal Republic of Germany (Voshage and Hintenberger), it was concluded that the cosmic ray intensity in the last million years had been greater than the average during the entire life-span of the meteorites. Another paper from the same Institute (Vilosek and Wänke) concluded that space erosion had been rather small. According to this paper, the relatively short life of stone meteorites is not due to erosion, but to collisions in space which would be catastrophic for stone but not for iron. A Belgian-US paper (Crèvecour and Schaeffer) also dealt with this question and stated that definite conclusions as to the changes in cosmic ray intensity in time could be made only after such problems as the erosion of meteorites in space and the production rate of radioisotopes in meteorites had been solved, although measurements of the aluminium-26/beryllium-10 ratio indicated that cosmic radiation intensity had been constant in the last ten million years.