

Memorial to Henry G. Thode 1910–1997

DENIS M. SHAW

McMaster University, Hamilton, Ontario, Canada L8S 4M1

SAM EPSTEIN

California Institute of Technology, Pasadena, California

JOHN M. HAYES

Woods Hole Oceanographic Institution, Woods Hole, Massachusetts

Leading geochemist, nuclear chemist, former president of McMaster University, farmer, Member of the British Empire, Companion of the Order of Canada, and 1980 Arthur L. Day medalist of the Geological Society of America, Harry Thode played many diverse roles and was warmly esteemed and respected in each of them.

Many knew him not only from his service as president of McMaster University, but as the principal developer of that institution. Another group of scientists would probably identify him mainly as a nuclear physicist. People from many walks of life in Hamilton, Ontario, would think of Harry as a very successful and creative businessman and contributor to civic life.

Henry Thode completed a B.Sc. in honors chemistry at the University of Saskatchewan in 1930, stayed to complete an M.Sc. two years later, and, just two years after that, at the age of 24, completed his Ph.D. in physical chemistry at the University of Chicago. In 1934 it was difficult to find a job. Harry found one at the Pennsylvania College for Women, in Pittsburgh, and he began teaching there in 1935. In September 1936, the American Chemical Society held its national meeting in Pittsburgh, and Harry took the opportunity to talk to Harold C. Urey and ask whether he needed a postdoctoral research associate. Urey's work on deuterium, the heavy isotope of hydrogen, had brought him the Nobel prize in chemistry in 1934, and he was then on the faculty of Columbia University in New York City. Harry's informal application fit Urey's style perfectly, and it yielded a job within 24 hours, since it was easy for Urey to check promptly with chemists who had known Harry in Chicago. He said yes the next day.

Harry Thode's two-year stay in Urey's laboratories yielded seven papers dealing mostly with the design and operation of systems for the separation of nitrogen-15 from nitrogen-14, carbon-13 from carbon-12, and sulfur-34 from sulfur-32. In the hands of Urey's biochemical colleagues, the newly available separated isotopes were driving a revolution by revealing the dynamic nature of biochemistry. For example, the availability of useful quantities of nitrogen-15 led to the discovery that proteins were constantly torn apart and rebuilt rather than synthesized and then degraded only when they were no longer needed. It is almost inconceivable to us now that this was not known only 60 years ago.

During those years at Columbia, Thode thought constantly about how to achieve not total separation of one isotope from another but at least substantial enrichment of the rare isotopes, so that the enriched material could serve as an isotopic tracer. The key was to find a reaction that



led to the greatest possible isotopic fractionation in a single step and then to devise some practical means of repeating it thousands of times.

Finishing up at Columbia, Harry first took a job with U.S. Rubber Company, across the Hudson River in New Jersey, but found his way back to Canada when, in 1939, he accepted an assistant professorship in chemistry at McMaster University. It was then a small, church-affiliated liberal arts college with two buildings. The director of research at U.S. Rubber—who would have thought of Harry as the hotshot from Urey's group—could hardly believe it when he described why he was leaving. It was, however, clearly a declaration of independence that any American could appreciate: a breathtaking combination of self-confident intelligence, foresight, and personal courage.

At Columbia, mass spectrometers had been part of the landscape, but when Harry moved to McMaster there were none anywhere in Canada, and they were not commercial products. So Harry found a magnet and fabricated his own spectrometer tube from Pyrex glass, rather than the customary stainless steel. Its difficult birth notwithstanding, that mass spectrometer went on to demonstrate convincingly that the operator and the choice of problems are always more important than the instrument. With it, Harry produced the first analyses of the gases resulting from the fission of uranium.

For part of the war Harry Thode was seconded to carry out war research. One of us, Sam Epstein, first encountered him at the University of Montreal, which at that time housed the Canadian atomic war effort. Harry was working in one of the small English groups whose objective was to determine the nature and concentrations of the rare gases in neutron-irradiated uranium metal. He was able to identify all of the isotopes of xenon and krypton which were produced by slow neutron uranium fission. These measurements provided wartime physicists with their first information about the relative proportions of the different isotopes produced by this fission. This was a technical and scientific contribution of huge significance and probably explains his early election to the Royal Society of Canada. When the University of Montreal laboratories closed down, this collaboration continued at McMaster University, but not for long. Thode acted in a typically unselfish manner and recommended that Epstein accept an offer to go to a job with Harold Urey at the University of Chicago.

When the dust of war settled, Harry had (1) the knowledge of processes of isotopic fractionation that he had gained in Urey's lab, (2) a view from that same era of biology and biochemistry at their best, and (3) the mass spectroscopic and analytical capabilities that he had developed at Hamilton. In the late 1930s, Al Nier had shown that the ratio of carbon-13 to carbon-12 varied among materials at Earth's surface, and evidence was accumulating that distributions of the isotopes of other elements were similarly uneven. The isotopic variations were tiny in absolute terms: 1.112% carbon-13 in a limestone and 1.087% carbon-13 in some organic material. Measuring them was an interesting challenge. At that stage even the founding fathers of isotopic geochemistry were publishing accounts of exploration rather than explanation.

Harry, on the other hand, was able to look at these natural fractionations and to know exactly what sort of processes might be causing them. While others explored, Harry explored *and* explained. He knew that Urey's new research group at Chicago was aggressively examining variations in the abundances of the isotopes of carbon and oxygen. Those in oxygen were viewed as particularly interesting because they seemed to be controlled by temperature, and that was something that could be understood by a physicist. No one seemed to be tackling sulfur, probably because its chemistry is so notoriously complex. Harry was attracted to it not only because it was unclaimed but because it had multiple isotopes that spanned a wide range of masses—32, 33, 34, and 36. It turned out that the fractionations were huge. Almost everywhere the McMaster group looked, sulfides were depleted in sulfur-34 and sulfates were enriched. Thode immediately identified these findings as an isotopic fractionation associated with the

action of sulfate-reducing bacteria. That insight stimulated the research that has led to our present recognition that fully half of the organic material reaching the seafloor is oxidized at the expense of sulfate rather than O_2 . A paper presented in 1951 at a meeting of the American Chemical Society had it all: a systematic consideration of the complete biogeochemical cycle of sulfur—its oxidation and reduction, erosion, burial, and volcanic emanations. This sort of thing showed up much later in papers from other groups—and a historical record of the isotope ratios of sulfate and sulfide backward in time, leading to an assessment of the antiquity of sulfate-reducing bacteria. Subsequent papers showed that the deposits of elemental sulfur that are so prominently associated with salt domes are systematically depleted relative to coexisting sulfate and must be of bacterial origin.

The titles of these papers are distinctive because they deal with what's going on as well as what's there. The first, in 1949, was "Natural variations in the isotopic content of sulfur *and their significance*." A 1950 paper in *Nature* was entitled "The distribution of sulfur-34 in nature *and the sulfur cycle*." These papers introduced what we would now call a "process orientation." They provided new information about how nature works, and they were recognized. Now everyone realizes that observed fractionations must be seen in the context of the global cycles of the elements.

Harry's wartime work on fission products and related nuclear topics led Harry Thode into many governmental and other national advisory scientific bodies, and attracted a growing group of students and colleagues at McMaster. Inevitably, although probably not by design, he became increasingly involved in administrative tasks. He nevertheless found time to interest himself in the projects of young colleagues, such as another one of us, Denis Shaw. His subsequent support in many ways really determined why many of us stayed at McMaster and turned down offers elsewhere. We never collaborated directly on any research project, but he would often come over to discuss what he was doing and to try to understand the strange reasoning of geologists.

These discussions continued even after he became heavily involved in administrative tasks, beginning in 1949 when he became principal of Hamilton College (a new entity, designed for some fiscal independence from the university's church affiliations) and continuing later as vice-president and then, from 1961 to 1972, as president of the university. Throughout that period, he was able to keep Saturdays free for science. It was in this period also that his work became more devoted to cosmochemical and medical topics, interests that continued to the end of his life. In the latter field he spearheaded several innovations in the application of radioisotopes as diagnostic tools.

One of Harry Thode's major achievements was to persuade the atomic energy authorities that there was a need in Canada for a nuclear reactor devoted to research for industry and academia, and to install such a facility at his home institution. The McMaster Nuclear Reactor has been in operation almost continuously since 1959.

This interest in applied science led Thode into several innovative enterprises. One of the most successful has been a local integrated circuit company, whose inauguration owed much to his encouragement and help.

He has received the Tory, Centenary, and Dawson Medals of the Royal Society of Canada, which he served as president in 1959. In 1980 he was the Arthur L. Day medalist of the Geological Society of America, an award that places him in the company of the most famous chemists and physicists that have contributed to the geological sciences. He was named a Fellow of the Royal Society of Canada in 1943, when he was 33 years old, and a Fellow of the Royal Society of London just 11 years later.

Although officially retired many years ago, he continued an active scientific career, going to his office on campus most days of the week, almost until his death in 1997. This was a man who was unselfish, loved science, and contributed enormously to the lives of many people.

SELECTED BIBLIOGRAPHY OF H.G. THODE

- 1938 (with Gorham, J.E., and Urey, H.C.) The concentration of N^{15} and S^{34} : *Journal of Chemical Physics*, v. 6, p. 296.
- 1940 The vapor pressures, heats and vaporization of melting points of N^{14} and N^{15} ammonias: *Journal of the American Chemical Society*, v. 62, p. 581–583.
- 1944 (with Smith, S.M., and Walkling, F.O.) The separation of the oxygen isotopes by the distillation of water: *Canadian Journal of Research*, v. B-22, p. 127–136.
- 1947 (and Graham, R.L.) A mass spectrometer investigation of the isotopes of xenon and krypton resulting from the fission of U^{235} by thermal neutrons: *Canadian Journal of Research*, v. A-25, p. 1–14.
- 1948 (with Macnamara, J., Lossing, F.P., and Collins, C.B.) Natural variations in the isotopic content of boron and its chemical atomic weight: *Journal of the American Chemical Society*, v. 70, p. 3008–3011.
- 1949 (with Macnamara, J., and Collins, C.B.) Natural variations in the isotopic content of sulphur and their significance: *Canadian Journal of Research*, v. B-27, p. 361–373.
- 1950 (with Macnamara, J.) Comparison of the isotopic constitution of terrestrial and meteoritic sulfur: *Physical Review*, v. 78, p. 307–308.
- 1951 (with Kleerekoper, H., and McElcheran, D.) Isotope fractionation in the bacterial reduction of sulphate: *Research*, v. 4, p. 581.
- Fission product yields and shell structure in atomic nuclei: *Transactions of the Royal Society of Canada*, v. XLV, p. 1–17.
- (with Lindsay, J.G., and Bourns, A.N.) C^{13} isotope effect in the decarboxylation of normal malonic acid: *Canadian Journal of Chemistry*, v. 29, p. 192–200.
- 1952 (with Johns, M.W., Gregson, J.H., Foster, G.C., and Jaimet, C.H.) Radio-iodine¹³¹ in the diagnosis of thyroid function: *Canadian Medical Association Journal*, v. 68, p. 132–137.
- 1953 (with Macnamara, J., and Fleming, W.H.) Sulphur isotope fractionation in nature and geological and biological time scales: *Geochimica et Cosmochimica Acta*, v. 3, p. 235–243.
- (and Wanless, R.K., and Wallouch, R.) The origin of native sulfur deposits from isotope fractionation studies: *Geochimica et Cosmochimica Acta*, v. 5, p. 286–298.
- 1954 (with Jaimet, C.H., and Kirkwood, S.) Studies and diagnostic tests of salivary gland and thyroid gland function with radio-iodine: *New England Journal of Medicine*, v. 251, p. 129–134.
- 1958 (with Monster, J., and Dunford, H.B.) Sulphur isotope abundances in petroleum and associated materials: *American Association of Petroleum Geologists Bulletin*, v. 42, p. 2619–2641.
- 1959 (with Harrison, A.G., and Monster, J.) Sulphur isotope fractionation in early diagenesis of recent sediments of northeast Venezuela: *American Association of Petroleum Geologists Bulletin*, v. 44, p. 1809–1817.
- 1962 (with Dunford, H.B., and Shima, M.) Sulfur isotope abundances in rocks of the Sudbury district and their geological significance: *Economic Geology*, v. 57, p. 565–578.
- 1963 Sulfur isotope geochemistry, *in* Shaw, D.M., ed., *Studies in analytical geochemistry*: Royal Society of Canada Special Publication No. 6, p. 25–41.
- 1964 (with Shima, M., Rees, C.E., and Krishnamurty, K.V.) Carbon-13 isotope effects in systems containing carbon dioxide, bicarbonate, carbonate and metal ions: *Canadian Journal of Chemistry*, v. 43, p. 582–595.
- 1970 (with Monster, J.) Sulfur isotope abundances and genetic relations of oil accumulations in the Middle East Basin: *American Association of Petroleum Geologists Bulletin*, v. 54, p. 627–637.

- (with Tracy, B.L.) Independent yields of ^{80}Br , ^{82}Br , ^{128}I , and ^{130}I from the thermal neutron fission of ^{235}U , ^{239}Pu , ^{233}U and ^{238}Np : *Canadian Journal of Physics*, v. 48, p. 1708–1715.
- 1971 (with Cragg, C.B., Hulston, J.R., and Rees, C.E.) Sulphur isotope exchange between sulphur dioxide and hydrogen sulphide: *Geochimica et Cosmochimica Acta*, v. 35, p. 35–45.
- (with Rees, C.E.) Measurement of sulphur concentrations and the isotope ratios $^{33}\text{S}/^{32}\text{S}$, $^{34}\text{S}/^{32}\text{S}$ and $^{36}\text{S}/^{32}\text{S}$ in Apollo 12 samples: *Earth and Planetary Science Letters*, v. 12, p. 434–438.
- 1976 (with Goodwin, A.M., and Monster, J.) Carbon, sulfur and isotope abundances in Archean iron formations and early Precambrian life: *Economic Geology*, v. 71, p. 870–891.
- (with Rees, C.E.) A ^{33}S anomaly in the Allende meteorite: *Geochimica et Cosmochimica Acta*, v. 41, p. 1679–1682.
- 1978 (with Monster, J., Appel, P.W.U., Schidlowski, M., Carmichael, C.M., and Bridgewater, D.) Sulfur isotope studies in early Archean sediments from Isua, West Greenland: Implications for the antiquity of bacterial sulfate reduction: *Geochimica et Cosmochimica Acta*, v. 43, p. 405–413.
- 1983 (with Goodwin, A.M.) Further sulphur and carbon isotope studies of late Archean iron formations of the Canadian Shield and the rise of sulphate-reducing bacteria: *Precambrian Research*, v. 20, p. 337–356.
- 1991 (with Ding, T., and Crocket, J.H.) Sulphur isotope and elemental geochemistry studies of the Hemlo gold mineralization, Ontario: sources of sulphur and implications for the mineralization process: *Canadian Journal of Earth Sciences*, v. 28, p. 13–25.

