Klaus Clusius,
Marcel Benoist Prize Winner 1958

The awarding of the 1958 Marcel Benoist Prize to Prof. Klaus Clusius, Professor of Physical Chemistry at the University of Zürich, is an important event for friends and students. They want to express him their heartfelt congratulations on the occasion of the official recognition of an outstanding part of his scientific work.

The national prize is awarded for the "most useful discovery during the previous year in the topic of medicine or natural sciences, where the achievement should have a special significance for the life and the health of mankind". These criteria are satisfied to a high degree by a research published in the spring of 1958 by K. Clusius with his student K. Schleich on the topic of *stable isotopes*. The new information provided there about the difference in vapor pressure of isotopic nitric oxide molecules could, however, not have been obtained without the longtime preparations which, taken by themselves, represent an impressive life-long work. So, let us follow these roots!

The first research on isotopes by Clusius was done 1934 in Goettingen and dealt with two very relevant problems at that time : the properties of the most recently discovered heavy hydrogen isotope Deuterium, and their interpretation through quantum mechanics, then recently developed. The difficult determinations of the energy content of the molecules HD and D_2 in all their phases at temperatures of –250 ^{o} Celsius and up to high pressures, are the mark of a very gifted experimenter who succeeds overcoming very tough obstacles. With these investigations, a "Leitmotiv" emerges, always with new and surprising variations in a large part of the scientific work of K. Clusius : In which way do the micro- and macroscopic properties of matter change, when certain atoms are entirely replaced by lighter or heavier isotopes ? At our present state of knowledge, except for special systems, there are only rough trials to respond theoretically to these questions. For the performance of the experiments, it is necessary to isolate the isotopes of interest from naturally occurring sources, where they exist as isotopic mixtures. Deuterium, the abundance of which is 0.014 % in water, can be enriched relatively easily via electrolysis; this is work that in the thirties each author had to do by himself, as the industrial production of heavy water only started in 1945. The ortho and para states of D_2, heavy water, deuterated hydrides of carbon, sulfur and selenium are the first objects of the synthesis and investigation of their physical properties. Here Clusius supplies important proofs for the correctness of the new quantum statistics, he discovers similarities between the low-temperature phase changes in crystals and provides accurate measurements for the thermodynamic properties of many molecules with substituted deuterium. Among them, the observed vapor pressure differences, which also are evidenced in the boiling points of the isotopic fluids, are of specially practical importance. This is because they led him in 1940 to a procedure, which today is a large-scale technical process to obtain heavy water through distillation of liquid hydrogen.

The problem of the isolation of stable isotopes of heavier elements than hydrogen has fascinated a large number of scientists since 1920. No one succeeded, however, to achieve enrichments for of more than about 50 percent for larger quantities of material. In 1938 Clusius invented (with G. Dickel) the thermal diffusion separation column, which was based on an observation during experiments studying the explosion of hydrogen-deuterium-air mixtures. If one fills an isotopic gas mixture, for instance with ordinary nitrogen gas with molecules $^{14}N^{14}N$ and $^{15}N^{14}N$, into a vertically oriented tube cooled on the outside, but with a wire heated to a red glow at its axis, then the molecule of $^{15}N^{14}N$, which is only present to 0.74%, gets significantly enriched at the lower end of the tube , while practically disappearing at the top end. With this relatively simple apparatus, where the columns often have total lengths of 30 to 50 meters, Clusius succeeds, after several years of efforts, to produce the isotope ^{15}N with a purity state of 99.5 % in quantities of liters. This achievement is preceded by production of the separated isotopes of Chlorine (1939), Neon, Oxygen, Krypton, Xenon, Carbon and later Argon in their pure state.

Aside from further technical developments in the separation tube operation, isolated isotopes, produced for the first time, lead to a continuation of earlier research directions: The application of stable isotopes as probes into chemical reaction pathways is gaining a prominent place in the activities of the Physical-Chemistry Institute of Zürich University. In particular, the heavy nitrogen isotope ^{15}N is being inserted in a large number of inorganic and organic compounds. For this endeavor, almost always new synthetic procedures have to be developed. With these isotopomeric species in hand, some deep and often unexpected insights are being gained into the chemical reaction and molecular structures of important nitrogen compounds. The results of these investigations throw a new light on knowledge that has been frozen in textbooks: The phenomena often manifest themselves quite differently and more complex than they had been taught hitherto for several generations.

The scientific work for which the prize was awarded could not have been chosen more appropriately to emphasize all three sketched areas of investigations with stable isotopes. This work depended on all the relevant techniques which had been developed. During an extensive, very difficult investigation of the vapor pressure differences between isotopic molecules, among them $^{14}N^{16}O$, $^{14}N^{18}O$, $^{15}N^{16}O$ and $^{15}N^{18}O$ it appeared that for these nitrogen oxides there are large isotopic effects. The boiling points of $^{14}N^{16}O$ and $^{15}N^{18}O$ differ by 0.529 degrees C from each other. Measurements carried out meanwhile demonstrate the theoretically recognized possibility that, because of this large difference, a direct distillation of NO will permit isolating the two rare heavy isotopes of nitrogen and oxygen, ^{15}N and ^{18}O in one single procedure and inexpensively. Prototypes of industrial installations for the production of these isotopes are already operating. Thanks to this, for these so important elements in biology, which do not have radioactive isotopes with a long enough lifetime, there are now available safe, radiation-less tracer atoms available in large quantities. There are a great many applications, and only two typical ones are mentioned here: 1) the elucidation of many biological processes, in which absorption, transfer and elimination of nitrogen and oxygen are involved and 2) rendering possible the use in large quantities of Thorium-^{15}N-nitrate in homogenous breeding reactors for the production of new fission material during the atomic energy production.

Besides these practical successes there are a number of interesting theoretical problems that emerge with these discovered effects, the interpretation of which gives us important insights into the game rules of the chemical force impacts.

The astounding scientific achievements, which emerge from a university institute with very simple facilities and with only a few collaborators, deserve the highest admiration. They can only be understood when one is aware of the enthusiasm with which Prof. Clusius is able to galvanize his associates, and where he himself is involved in measurements, supervises directly the experiments, and notices so many small, but important peculiar features that escape eyes less skilled, and deduces correctly the consequences. One can hardly characterize better his Institute than was done by Prof. A.O. Nier at an international symposium, who stated : “All brains and almost no money”. May this productive investigator and amiable teacher remain in excellent health und give us further scientific knowledge over many more years.

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