Stable Isotope Production

—a distillation process

by Nicholas A. Matwiyoff, Berthus B. McInteer, and Thomas R. Mills

A 70(-foot distillation column is far from ordinary, but the Laboratory was forced to this length to separate the rare but stable isotope carbon- 13 from the common isotope carbon- 12 by distillation of carbon monoxide. In somewhat shorter distillation columns the Laboratory also enriches the even rarer stable isotopes nitrogen-15, oxygen-17, and oxygen-18. Produced in the western hemisphere only at Los Alamos, these enriched stable isotopes are used as tracers in many research areas ranging from metabolism to agriculture to atmospheric circulation.

Distillation. one of the oldest and most effective methods for separating mixtures, exploits differences in the boiling points of the components. Two familiar examples of its beneficial applications are the separation of hydrocarbons from crude oil and the production of a beverage with a high alcohol content and a distinctive flavor from sour mash.

Distillation is most often applied to mixtures whose components have boiling points that are above 50 degrees Celsius and that differ from one another by more than 10 degrees. Separation of such mixtures can then be carried out in relatively short (10- to 30-foot) columns. In contrast, the boiling points of the various isotopic forms of carbon monoxide or nitric oxide are very low [only slightly higher than the boiling point (-196 degrees Celsius) of liquid nitrogen] and, more important, differ by only tenths of a degree. Small as these differences may be, they are larger than those exhibited by most other compounds of carbon, oxygen, and nitrogen.

The boiling points of ^{12}CO and ^{13}CO differ by less than 0.1 degree Celsius (more precisely, their vapor pressures differ by less than 0.8 percent). To separate the two, the Laboratory built a 700-foot distillation column—probably the longest in existence. This engineering marvel. developed by B. B. McInteer, T. R. Mills, and J. G. Montoya, produces 20 kilograms per year of 99+ atomic percent carbon-13. The problem of supporting such a long column was solved economically by lowering it into a cased, 15-inch-diameter hole in the ground, but this method of support prevents access for repair. Design and construction of the system therefore required the greatest care. The length changes (about 2 feet) that occurred when the system was initially cooled to the temperature of liquid nitrogen for operation (or that will occur should shutdown be necessary) demanded particular accommodation. The welds joining the thirty-five 20-foot sections were thoroughly tested for soundness, and the gas feed lines were fitted with expansion joints, Proof that the system met the highest standards of design and construction is its continuous operation without incident since 1978.

Since the vapor pressures of the various isotopic forms of nitric oxide differ more from each other (2.7 percent in the case of $^{14}N^{16}O$ versus $^{15}N^{16}O$) than do those of carbon monoxide, the two distillation columns at Los Alamos for separating them are "only" 150 and 270 feet long. These systems are complicated, however, by the large number of product streams ($^{14}N^{16}O$, $^{14}N^{17}O$, $^{14}N^{18}O$, $^{15}N^{16}O$, $^{15}N^{17}O$, and $^{15}N^{18}C$)) and by the necessity, since liquid nitric oxide is a high explosive, for barricades and remote controls. Nonetheless the two columns have been operated routinely since 1975 with an annual production capacity of 18 to 20 kilograms of nitrogen-15 at enrichments of up to 98 percent and 1 kilogram of oxygen- 17 and 13 kilograms of oxygen-18 at enrichment of better than 40 and 95 atomic percent, respectively.

Under the guidance of McInteer and Mills, research has been directed recently to development of distillation methods for separating stable isotopes of heavier elements that are used as targets in the production of short-lived radioisotopes for nuclear medicine. ■

Schematic drawing of the Los Alamos distillation column for separating the naturally occurring mixture of ^{12}CO and ^{13}CO. The 700-foot column is supported over most of its length within a hole in the ground. The packing (spherical or saddle-shaped pieces of ceramic or metal provides surface area for contact between liquid and vapor phases. The column is cooled initially to liquid nitrogen temperature as natural carbon monoxide enters, rises to the condenser, and liquefies. The liquid flows downward and cools the column as it vaporizes. When liquid carbon monoxide collects at the bottom, it is vaporized by carefully regulated heat. At equilibrium a downflow of liquid repeatedly contacts upstreaming vapor, and the more volatile component accumulates at the condenser.