Chapter 3

Stable Isotope Enrichments

Isotopes are defined as atomic nuclei having the same number of charges, or protons, but different number of neutrons. As nuclear charge determines the atomic properties, different isotopes with the same charge, or the same chemical element, would have the same chemical properties, and they cannot be separated by chemical means. Many isotopes are radioactive, but many are not. In nuclear medicine, they often confuse all isotopes with radioactive elements, which is wrong. Oxygen-18 is a perfectly stable element and can be used as a target material to create fluorine-18 for fluorodeoxy-glucose for PET (positron emission tomography). The heaviest stable element is Uranium, beyond which the electrostatic repulsion would overcome the nuclear binding force until the gravitational force comes into play, and that will start from a white dwarf as one giant atomic solid evolving to a neutron star to compose the stellar matter as one giant nucleus.

There are nine schemes of isotope enrichment. Three of them were developed during the Manhattan Project, since then one was developed in Canada, one more in U.S. which was unsuccessful, but became successful in Australia, and one unsuccessfully in Brazil. I participated to initiate a few more. The most well known method is the use of magnetic bending. First developed at Berkeley called Calutron (California University tron); it is an early version of the mass spectrometer where ionized elements of various masses would enter into certain spatial windows according to their mass. Using silicon-28 as an example, if a particular window or channel would take in 10,000 atoms per second, a single gram of silicon would require 60 billion years to accumulate, more than 4 times the age of the universe. Calutron today is used mainly to separate radioactive elements where the amount of material under processing is extremely small and of very high value. In the early days of the Manhattan Project, in order to assure the success of Calutron that utilizes large magnetic coils, and copper was the key metal for munition shells, the silver holding of the U.S. Treasury was utilized for silver wires at the Oak Ridge National Lab in order to process the Uranium

isotopes. This is a good example of useless messy tech turned semi-useful in a different operation.

The second method is called "diffusion", made famous because of the Manhattan Project. In a given temperature for a gaseous fluid, all molecules have the same kinetic energy, which is the thermal energy. Light particles in this fluid would have more random velocity and would diffuse slightly faster than heavy particles. Passing the fluid under a barrier, light particles would move slightly ahead of the heavy ones and thereby cause separation over the lengthy barrier. This separation provides also an entropy change and from the amount of energy required to deliver such an entropy change, a thermodynamic efficiency can be evaluated. The efficiency is one part in 48 million for UF_6 molecules under a diffusion process at Oak Ridge National Lab. This was the first successful method for large scale separation, and it became a standard method of enrichment to which all other methods are compared.

The third method is called "centrifugation", and it has been used most extensively in biomedical sciences. Christian Deduve, a foremost expert in separating cellular components, got a Nobel for isolating lysosomes that we are trying to use for localized tumor management (Chap. 10). When Dr. Deduve retired from the Rockefeller University, we collected all his used laboratory furniture for our first lab. Centrifugation for an industrial isotope production, however, is truly a messy tech. Prof. Manson Benedict, then the head of the Nuclear Engineering Department at MIT, an expert in the diffusion process, told me that in these large scale centrifugations, if the electricity is removed from the instrument, the spinning cylinder would continue to rotate for another year. Technology developed to the level with such sophistication is completely beyond my simple tech comprehension.

The fourth method is distillation, made famous by the deuterium enrichment from water in the Canadian program of nuclear reactors using the unenriched Uranium. In a liquid, heavy molecules have a slightly higher vapor pressure because of higher momentum from the same thermal energy so that by passing through thousands of stages of repeated distillation and condensation, the heavy and light components can be separated. This process is particularly efficient for light molecules with high vapor pressure at ambient temperature such as water. Candu is the Canadian effort for the enrichment of heavy water where deuterium is enriched from its natural abundance of 150ppm to nearly 100%. Candu can produce it at under $10 per liter. Heavy water enables the moderation of nuclear chain reactions with natural uranium where ^{235}U has 0.711%. Otherwise, ^{235}U must be enriched to approximately 3% with regular light water for moderation.

Since ^{235}U decays over geological time and reduced its fraction to the current 0.711%, it did pass through the 3% stage and did start the nuclear chain reactions moderated by the light water surrounding Uranium ore. That is, natural ore started the chain reactions long before the Manhattan Project.

The fifth method is the use of laser excitation. Promoted by President Ford, Lawrence Livermore National Lab and Exxon used a two-stage laser excitation, with the first stage to excite all fluids in a raised energy state and the second stage selectively ionizing the targeted molecules. Ionization, however, limited the throughput as the fluid has repulsive charges, like those in the Calutron, that the product stream cannot easily be bundled for a large fluid flow. More recently the Australian group applied a low temperature approach to the fluid. By using the selective excitation of laser energy for a gaseous phase transition from the liquid without ionizing it, the fluid flow can be arranged for a much higher throughput.

The sixth method uses a supersonic fluid impinging on a bent wedge. By forcing the fluid to bend over a sharp corner, the centrifugal force would cause the fluid to have an outer layer with a more heavy component and an inner layer with a more light component. A slit is cut in the curved wedge so that the outer component can be collected. It is a "poor man's centrifuge". Compared to diffusion, centrifugation can improve the thermodynamic efficiency by a factor of 20, but at twice the capital cost. Centrifugation is generally favored over diffusion if the cost of capital is not an issue. But using the wedge, as promoted by the Brazilian government, it pushed the centrifugation into a more messy tech, too messy to be useful.

My Ventilated Turbine Blades and Other Schemes

The seventh method is the use of a ventilated turbine blade. The most difficult issue of centrifugation is the management of input and output of fluids under a very large force. The bent-wedge made the situation worse. The ventilated blade, on the other hand, greatly simplified the situation and made it a simple tech. In a turbine blade, the fluid is sloshed back and forth between rotators and stators at approximately 500,000g (g is one gravitational acceleration on Earth's surface), a typical number for centrifugation. If the rotating blade is ventilated, the heavy component would be forced to enter into the blade surface over a very stable boundary layer because the ventilation also sucks away the turbulent energy. Having entered the blade, the fluid can exit at the tip of the rotating blade and be part of a separated product of waste stream. The processing

surface of a moderately large turbine can be measured in hundreds of meters, while the separative boundary layer where the fluid must penetrate through is only under 10 microns. This is a large reduction from a meter-long centrifuge tube. This blade method, changing from emitting gas at the blade surface for turbine thermal management to deleting gas at the blade surface for enrichment and stream separation, greatly simplifies the fluid management; nevertheless, it remains to be a somewhat messy tech.

My Solar Wind Method

The eighth method is the solar wind time-of-flight. In a solar flare, the photons would need 500 seconds to reach the Earth's orbit, with electrons 30 hours later, followed by protons in another 30-50 hours. They all start from the same spot in the rotating chromospheres, travel with different garden-hose trajectories and are completely separated at Earth's obit because of their difference in the time-of-flight in entirely different trajectories as observed by our solar wind satellites. The mean-free-path of the solar wind is longer than the distance to Earth, therefore there is almost no scattering, and no increase of entropy. The thermodynamic efficiency of separation is nearly 100%, or $2x10^8$ fold higher than the diffusion process.

George Uhlenbeck of the Rockefeller University

At the insistence of Gus Kinzel, who said that he told many of his friends about me, but had almost no scientific articles about my inventions to show. I

told him that I was tired of fighting silly battles, but if he really insisted, I would do some patents etc. After that, I wrote an article on this solar wind method for PNAS (Proceedings of the National Academy of Sciences) that was reviewed by George Uhlenbeck. The fluid mechanics of the continuum (highly collisional) is rather simple, and the fluid mechanics of the rarified flow (collisionless) is also relatively simple. But dealing with the transition region, I had no mathematical tool. I went to the volumes of the Annual Review of Fluid Mechanics to look for some guidance and saw a volume of that year in honor of George Uhlenbeck of the Rockefeller University.

I told him that while I had no mathematical tool nor large computers to do numerical simulations of scattering, I use entropy to link the two regions, assuming that the fluid flow from a collisional region to evolve into a collisionless region would maintain the same entropy. Under this assumption, the analysis could proceed reasonably straightforwardly. He complemented me by saying that it was as good as any one could do, because there is no mathematical tool that can effectively deal with the transitional region except for numerical simulations. The analysis indicated that the separative power, if a machine can be constructed according to the solar wind/supersonic nozzle principle, would be proportional to the eighth power of the Mach number, with Mach one being the speed of sound. That is, for light molecules capable of high supersonic flow, the method would yield an exceedingly powerful separation. But for heavy molecules that could not be pushed to high Mach flow, the method would be virtually useless. Note that Parker described his solar wind model from the mathematics of a supersonic nozzle. As the nozzle confines the fluid molecules from three degrees of freedom to a one dimensional vector flow, all collisions at the nozzle throat would feed to energize the vector direction and thereby accelerate the nozzle flow to supersonic speed. The solar gravitational pull also reduces the fluid motion from three-dimensional degrees of freedom to the one-dimensional radial flow into the vacuum, while absorbing all those non-radial flows back to the chromosphere under gravity. In other words, there is a filtering selection here in addition to the "nozzle confinement"; as a result, the solar wind would accelerate to high Machs against the solar gravitational pull, as predicted by Parker and it was verified by our solar wind satellites.

Parker used the nozzle analysis to describe the rotational (the chromosphere rotates at 25 days per revolution) garden hose trajectories. I simply used the measured solar wind parameters to go back to rotating supersonic nozzles for separation without centrifugation. Note that the accelerations of solar wind can be applied to the galactic flows as well as to the expansion of the early universe

where the non-radial flows are filtered back into the source, promoting the radial accelerations against the gravitational pull. Such a filtering process invokes no "dark energy", which renders the measured acceleration of the big bang expansion to be rather mysterious.

We cannot, of course, build a separator the size of the Earth's orbit. If we do poor engineering and harvest only 2% efficiency with a one-meter chamber, we could already gain a huge improvement over diffusion at 2 parts in 10^8. The chamber must have a rotating nozzle at the center to deliver the fluid radially to a co-rotating wall designed to collect (empty) the fluid. The leading collector would receive the light component and the trailing collector would receive the heavy component. In a meter-sized chamber, a single stage had experimentally altered the input of N_2/O_2 ratio at 80/20 to 96/4 ratio as received by the collectors without centrifugation or ionizing the fluid.

A Seminar at Lawrence Livermore National Laboratories

Edwin Huddleson

Edwin Huddleson, my mentor lawyer friend from Cooley Godward of San Francisco, once arranged me to give a seminar at Laurence Livemore National Lab on my ventilated turbine blade method mentioned above. I gave out some technical data and reprints a few days earlier to prepare the audience as the laboratory at that time was also doing isotope enrichment using a two-stage laser excitation, which was very different from my approach. But I mistakenly thought that the seminar topic was the solar wind model and started to talk solar wind astronomy and completely lost the audience. After 10 minutes into the seminar, realizing that I was delivering the wrong topic, I apologized and went back to the original topic and finished the seminar without a second thought. Two decades later, I met someone visiting New York interested in isotopes and was astonished that he knew a great deal of my solar wind method. I did have an article published in PNAS decades ago, but it is not an easy article to digest and the visitor was not a solar wind physicist. Finally he told me that he was in my audience at Livermore. Even though I talked on the subject for only a few minutes, the audience was much intrigued and assigned him, being the lowest ranking member, to "check out", so to speak, my solar wind story. The audience probably would not have paid much attention if I gave a proper seminar on solar wind physics, but I stopped after a few minutes, and therefore it must be interesting. As a research scientist, one must routinely give seminars all the time. But to give a wrong seminar and get checked out is something else.

The ninth method is a further simplification of the fluid manipulation under a supersonic nozzle. When I first talked to Ed Huddleson of Cooley Godward about the isotopes, he sent me to Luis Alvarez, an experimental physicist. It was before he and Walter, his son, discovered the cause of the dinosaur disappearance 65 million years ago. Luis was an extremely versatile physicist with a Nobel in experimental particle physics. Upon hearing my methods on turbine blades and solar wind, he fully appreciated that they have sound scientific base. But he told Ed to tell me to stop working on them because "He will never make a dime" in this field, regardless of whether they are simple tech or messy tech. He was almost correct, and Kinzel too would not touch them. Nevertheless, these three enrichment methods were some of my better inventions and could serve some useful biomedical applications, which currently cannot be properly produced.

Some Applications in Everyday Use

There is a great variety of applications in stable isotopes, particularly if their cost of production can be reduced drastically. By far the largest use could be the light isotopes for Functional MRI, which will be described in the chapter 7. Two

applications for industrial use will be described. Take Mercury (Hg) for fluorescent lighting for example. Hg has seven stable isotopes; ^{196}Hg (0.15%), ^{198}Hg (9.98%), ^{199}Hg (16.87%), ^{200}Hg (23.10%), ^{201}Hg (13.18%), ^{202}Hg (29.86%), and ^{204}Hg (6.87%), but only six of them participate with fluorescent activities. If the light ^{196}Hg fraction can be increased by an order of magnitude to more than one per cent, all seven would participate and thereby enhance fluorescent efficiency by up to 15%, although some experiments claimed to have gained only 5%. Note that besides mercury lighting, other uses such as the mercury battery, thermometers, switches, etc. are not dependent on the ^{196}Hg and would not have a negative impact if their portion of ^{196}Hg has been deleted at a facility near the Mercury one and applied for lighting applications.

Silicon has three stable isotopes; ^{28}Si (92.2%), ^{29}Si (4.7%), and ^{30}Si (3.1%). If the heavy isotopes of silicon can be removed, the pure ^{28}Si at six-nines may conduct heat more effectively than copper on silver provided that the highly enriched Si-28 does not have substantial impurity phonon scattering to reduce the thermal conductivity.

Not All Attempted Methods Are Successful

I had a failed attempt to enrich oxygen-18 isotope. ^{18}O has been used as a cyclotron target to create excited fluorine-18 for positron-emission-tomography (PET) for brain evaluation using fluoro-deoxy-glucose, a rat poison that engages neutrons extremely effectively. Water with ^{18}O costs approximately $100 a gram and the demand for ^{18}O, which is produced in nuclear reactors, outstrips the supply. Water with ^{18}O is heavier than normal water by more that 10%, and with the natural abundance of ^{18}O at 0.2%, its supply is unlimited. Biologists have been separating cellular components with centrifugation with a weight difference far below 10%, so I thought it should be a fairly easy task to deliver ^{18}O. I spun the water at 100,000g, the limit of most ultra-centrifuges, without any enrichment.

I believed that the Brownian motions must have neutralized the separation, so gel was added to the solution, as usually practiced by biologists, and still obtained no enrichment, without realizing at that time that the Van de Waals force of the polar molecule H_2O is really very strong. H_2O molecule has basically a tetrahedral form, with oxygen in the middle, two of the 4 tetrahedral corners are hydrogen with slight positive charges and the other two corners orthogonal to the line linking the hydrogen atoms, are negatively charged electron clouds. Electron clouds would attract the protons of neighboring H_2O

molecules while the protons would attract the electron clouds of neighboring H_2O molecules. These hydrogen-based attractions are the electrostatic Van de Waals Forces. That is, instead of freezing the sample under gel in a refrigerated centrifuge to prevent Brownian motions, the samples should instead be heated to near boiling temperature where the thermal energy is high enough to neutralize the Van de Waals force to become vapor. Alternatively, instead of the polar H_2O, the proper target molecule should be non-polar light molecules having one oxygen. The stickiness of the H_2O molecule, on the other hand, explains the relatively high viscosity of the liquid.

Speaking of viscosity, which gives a limiting speed of boat transport, particularly that of racing speed boats. At high speed, a large amount of water would be carried by the moving boat surface creating a huge screen of splashing liquid. If the bottom of the lifting surface, such as a hydrofoil, is made movable and rotational, like a snow mobile, then the moving surface engaging the stationary water will be more like a rotating wheel engaging the ground. Once the viscosity of the liquid can be eliminated from the speeding surface, the boat transport could function more like an airplane but still benefit from the lifting power of the water.

CHAPTER THREE REFERENCES

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