Filtration Materials for Separating Isotopes

Certain porous materials or layers have been used for filtering substances since the first human tried to strain muddy drinking water. Thin filter media, such as filter cloth, filter screens, or laboratory filter paper, have mesh sizes smaller than the particles to be trapped. "En masse" filter beds, such as a thick layer of sand, or a coke bed, porous ceramics, or porous metals operate on the principle that, though the individual pores in the filter media are much larger than the particulates to be removed, the tortuous path ensures that particulates will be trapped by attaching to the medium before they can emerge on the other side of the filter.

A third separation technique uses the thermal motions of molecules through the tangled "maze" of filtration media. Lighter molecules will diffuse through the material faster than heavier molecules; with several such filtration stages, the fraction of lighter molecules can be increased with each pass through the media. Perhaps the most extraordinary use of this filtration technique was for separating uranium isotopes in the 1940s. Doing so required the development of exceptional new membrane materials and extremely high quality-control standards.

Natural uranium contains about 99.3% of the isotope with an atomic weight of 238, and 0.7% of the 235 isotope. Separation of the isotopes was not possible using chemical methods because the uranium chemistry remains the same for both isotopes. The only way to separate them *had* to be physical.

Isotope separation had already been achieved in 1932 by Gustav Hertz in Germany. Hertz isolated isotopes of neon by cycling the gas through 12 mercury diffusion pumps in a cascade, allowing the neon to diffuse through a stream of mercury vapor. By 1938, this technique had been used to enrich C-13, N-15, and O-18 ten-to twentyfold. However, the mass difference between isotopes of lighter elements is about 5%, while for uranium isotopes U-238 and U-235 it is closer to 1%. A different separation scheme was required.

In the early 1940s, researchers attempted various physical methods, such as electromagnetic separation using the new cyclotrons at Berkeley and thermal diffusion using a heated tube inside a cooled concentric tube. Thermal diffusion caused the heavier fraction to concentrate at the cooler interface, and the lighter isotope to concentrate at the hotter interface; convection then allowed a greater fraction of the lighter isotope to rise toward the top.

The "gaseous diffusion" method using filtration media, however, required the development of a new kind of material with pores so fine that they would preferentially allow one isotope of uranium to pass through more rapidly than another. Since the mean molecular velocity in a gas is inversely proportional to the square root of its molecular weight, a light isotope will diffuse through filtration media slightly faster than a heavier isotope.

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Separating uranium this way offered yet another complication: The gaseous form of uranium to be used, uranium hexafluoride, was extremely corrosive and destroyed most typical materials, including steel piping and any organic-fiber filtration media. In addition to developing a suitable barrier material, the gaseous diffusion process also required new designs for piping, pumps, and backup systems that could withstand the corrosive gas. Uranium hexafluoride is also a solid at room temperature and pressure, so it needed to be heated before it could be of any use.

Early in 1942, John Dunning, Eugene Booth, and Astrid Grosse at Columbia University designed a workable cascade system of diffusion barriers that would, in theory, enrich the concentration of U-235 at the top end and leave the more plentiful U-238 toward the bottom—given an appropriate filtration material for the barrier membranes. However, no one had yet come up with a filter media that could perform the job. Dunning and his group continued their work at Columbia, narrowing down the candidate materials—porous metal meshes, fiberglass, and ceramics all failed because of the corrosive feed gas.

The characteristics of the uranium separation process required that the mesh be free of holes much larger than 0.01 μm, but with billions of smaller holes—none of which could be allowed to enlarge or plug up as a result of corrosion or dust; *and* the mesh had to withstand a pressure differential of one atmosphere.

Edward Adler, a 26-year-old chemistry teacher at City College in New York, joined the Columbia group and tried his own ideas. He started by using metal alloys in which he etched out one component of the alloy. He removed the zinc from brass alloy sheets, leaving only the remaining copper. This showed some promise, but still couldn't match the extreme requirements for putting the process into industrial practice. He tried etching a thin sheet of silver-zinc alloy with hydrochloric acid to create a porous barrier. Adler also attempted to add impurities to copper, electroplate it onto a sheet of base material, and then chemically remove the impurities. He tried other ideas through the winter and spring of 1942, without success.

Then John Dunning asked Adler to meet with Henry Norris to discuss a new idea. Henry Norris was an interior decorator, had had only two years of high school, and knew very little about science—but he had invented a new kind of metal mesh that looked extremely promising.

Norris's incentive to create such a fine mesh was his dislike for the then-available spray-painting guns he used for interior decorating. In 1934 he used an electrodeposition process to make a fine metal screen for a new type of spray gun that operated on centrifugal force rather than compressed air. The C.O. Jelliff company, which made woven-wire products, bought Norris's fine screen and hired him to manufacture them.

British scientist Franz Simon, who was also working on the problem of gaseous diffusion of uranium saw Norris's commercially available screens. Simon contacted Norris to ask if Norris could manufacture an even finer metal screen, half an inch square with half a million holes in it. When Norris succeeded, Franz Simon brought him to meet Dunning at Columbia University. By the end of 1942, working in his own home workshop, Norris succeeded in creating two samples that appeared more promising than anything else the Columbia group had come up with.

In 1943 Norris joined Edward Adler in his laboratory. Both men got along well and pursued any ideas that came to them, though neither the chemistry teacher nor the ex-interior decorator proved to be meticulous or tidy workers. They managed to eliminate most known metals and alloys from consideration, as well as glass. Only nickel looked promising.

The chance always remained that a few too many large holes in any one segment of the membrane material would ruin the barrier. Before the barrier could be tested in the laboratory, such large defects had to be mended by hand. Adler himself spent many hours staring under a microscope and painstakingly filing holes using a sewing needle.

Adler and Norris managed to create a nickel screen with the appropriate porosity, with holes of the correct fineness. At first they thought they had solved the problem, but upon larger scale testing they found their new material proved too fragile to withstand any gas pressure and too brittle to be assembled in large barriers for an industrial process. The Norris-Adler barrier was also quite non-uniform and varied widely from sample to sample. The fragility and non-uniformity of the Norris-Adler barrier material caused some of its detractors to call it the "lace curtain." But it had come closer than any other prospect, so they continued their work.

Mass-production techniques for the new barriers were investigated, including a four-month study that used lithography to lay down the filter pattern. Adler's investigations took place in secret on a special printing press commandeered from the American Chicle company, manufacturers of chewing gum, but still Adler and Norris could make no major improvement in the material.

Meanwhile, at Bell Telephone Laboratories in New York, Foster C. Nix was working on the same problem, trying to develop an isotope-separation barrier using compressed nickel powder. Nix's and Dunning's groups worked independently, though Nix did get along well with his competitors. His initial tests with the compressed-nickel powder showed that it had superior structural properties to the Norris-Adler barrier, though Nix's pinholes were not as fine. Nix didn't yet know how well it might separate the uranium isotopes, and his special in-house method of preparation for the filter media would not lend itself to large-scale production...and if the process were to be used for a full-scale gaseous diffusion plant, it would require thousands of stages and acres upon acres of the filter media.

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The mass-production of Nix's alternative material proved to be one of its greatest difficulties, but a new way to make the pressed nickel powder was eventually developed by a technician named Frazier Groff, brought to Bell Laboratories from Union Carbide to tackle the problem.

Another scientist, Clarence Johnson, working at the Jersey City Laboratory for Kellex (a wartime industrial company), spoke with Frazier Groff about the possibilities of combining the two processes, the electrodeposition technique and the pressed-nickel-powder technique. Johnson and an assistant worked together, trying variation after variation until finally they achieved a sample barrier that appeared to have the strength and flexibility of Nix's pressed-nickel-powder barriers, and after testing proved to have the appropriate porosity and pinhole-size as the Norris-Adler work.

Johnson tried several more times, and the subsequent two samples seemed to be as good as the first—but then the next three all failed. They couldn't understand this sudden failure, until they realized that in the excitement of succeeding after years of work, they had forgotten one of the preparation steps! Unfortunately, the details of this process remain classified after 50 years, so it is impossible to determine exactly which step they forgot or how the two approaches were combined.

The first small samples of the Johnson-Kellex barrier material (about the size of airmail envelopes) passed all the required quality-control tests. Kellex began to ramp up for production work, creating larger batches of the new barrier. They brought in Hugh Taylor, a British-born chemist who had been teaching at Princeton, to inspect the new filtration material. To begin his testing, Taylor took a single square foot of the Johnson-Kellex material and cut it into 144 separate square-inch pieces, gave them to his technicians, and asked them to test every single swatch. The quality ranged from poor to excellent across the first sheets of material, and with Taylor's suggestions and improvements, the Johnson-Kellex barrier became the material of choice for uranium isotope separation.

Millions of square feet of the material were eventually used in the K-25 gaseous diffusion plant at Oak Ridge, Tennessee, where uranium hexafluoride gas passed through a thousand stages of the filtration material, to increase the enrichment of uranium-235.

The gaseous-diffusion process was only one technique among several used for separating the isotopes to obtain pure uranium-235. The output from the gaseous diffusion cells—still not pure—was then added as feed material to the electromagnetic separation plants and thermal diffusion cells. Altogether, after an enormous effort, the two isotopes could be sorted even in spite of the vanishingly small differences between them.

Other techniques for separating the isotopes of uranium eventually proved successful, such as laser isotope separation at the Lawrence Livermore National Laboratory, but the Oak Ridge K-25 plant is still being used for gaseous diffusion today.

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