

An Early History of Heavy Water

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Abstract

Since 1945 Canada has had a nuclear power industry based on reactor design which uses natural uranium and heavy water. The tortuous and improbable sequence of events which led to this situation is examined.

1 Introduction

Pure heavy water, D_2O , is the oxide of the heavy stable isotope of hydrogen, deuterium, denoted by the symbols 2H or D . It is physically and chemically almost identical to ordinary “light” water, H_2O , except in that its density is 10% higher, hence its name. The answer to the question everyone asks is “Yes, you can drink it”; the Material Safety and Data Sheet states that dizziness and nausea can occur if you ingest 10% of your body mass of heavy water, and LD_{50} is 30% of body mass. Hence it is safer than ethanol.

Most of the deuterium (heavy hydrogen) was formed about 10 minutes after the Big Bang, along with other very light isotopes presently found in the universe. More recently, 2.5 billion years ago, most of the deuterium atoms on the earth were incorporated into water molecules. As a small isotopic fraction of natural hydrogen (0.015%), deuterium existed then, as now, mostly in the form of HDO molecules. And this is the way things remained until deuterium, and thus heavy water was discovered in 1931. However, before we examine how deuterium was discovered, let us examine the events which led to the manufacture of heavy water, for these seeds were sown before those of discovery.

2 Birkeland and Norsk Hydro

At the turn of this century Professor Kristian Birkeland[1] of the University of Kristiania (soon to be renamed Oslo) was experimenting with a device called the Terella. This was a laboratory model of the earth, complete with magnetic field, placed in an evacuated vessel into which ions could be injected at high voltage. By observing the discharge glow, Birkeland was able to understand the three-dimensional structure of the Aurora Borealis. However, the equipment was expensive and money, then as now, was hard to come by. Kelvin suggested to Birkeland that research into armaments might prove lucrative and allow him to continue with the Terella. This was not the first time this idea had occurred to a physicist, Birkeland with his expertise in electromagnetism, set about producing an *elektriske kanon*, a rail-gun.

On Feb. 6th, 1902 Birkeland's *kanon* short-circuited and exploded during a test, but the disappointment he felt was muted by the observation of a disc-shaped arc, spread by the magnetic field, and the smell of nitrogen oxides. The reason this was intriguing was that the world was then gripped by a fear greater even than that of the looming conflict in Central Europe - that of world famine. Chilean nitrate deposits, on which the world depended as a fertilizer, were on the brink of running out, and chemists were scurrying to find an economical way of fixing atmospheric nitrogen. Clearly the acrid brown stench of nitrogen oxides was the smell of nitrogen being fixed, as it was a short hop skip and a jump to nitric acid and any nitrate you please.

Birkeland was not the first to fix nitrogen by electric arc - Crookes in Manchester already had a pilot plant producing calcium nitrate by this means - but the disc-shaped arc promised a high yield[2]. However, a week after the explosion he met a man who was already working on the means which would make economical production possible. That man was Sam Eyde, a civil engineer who

was fascinated by the enormous potential of Norway's mountains and rainfall for the production of hydro-electricity. After that things moved with breath-taking speed. A week after meeting, Eyde and Birkeland submitted a patent for artificial fertilizer. They obtained money from the Swedish financiers, the Wallenbergs, and a mere three years later a hydroelectric plant had been built out of the wilderness at Notodden and a Birkeland-Eyde arc furnace was producing the first Norgespeter - Norwegian Saltpeter, i.e. calcium nitrate.

In the same year as the first Norgespeter was produced, Fritz Haber discovered a much better way of producing nitrates via ammonia made by what is now known as the Haber process. Indeed, the Norwegians soon abandoned the arc furnaces and adopted Haber's idea. However, Birkeland's discovery had started something irreversible: the large scale development of hydro-electric power in Norway by the company he and Eyde had started: Norsk Hydro.

Now we will examine the other chain of events, that of discovery. First, however I should note the strange and sad circumstances of Birkeland's death in 1917 - too soon for him to know anything about heavy water. He had been spending some time in Egypt, where it was hoped that the warm weather would improve his health. He wished to return home to Norway, but found the direct route barred by the First World War. During the ensuing tortuous detour, he died, in Tokyo.

3 Discovery

The path which led to the discovery of heavy hydrogen and heavy water was every bit as tortuous as Birkeland's attempt to get home from Egypt. In 1913 Arthur Lamb and Richard Lee at New York University were trying to improve measurements of the density of water; this was a very important quantity to know accurately because of its importance as a standard[3]. They were attempting an accuracy of 200 ppb, but were not able to get agreement between samples taken from different geographical locations to better than 800ppb. They did not grasp the significance of this discrepancy - the abundance of deuterium varies due to rates of evaporation and condensation - despite the fact that the concept of isotopes as developed by Frederick Soddy and Kasimir Fajans was a significant piece of scientific news that year.

In 1929 William Giauque and Herrick Johnston discovered ^{17}O and ^{18}O . Two years later Harold Urey at Columbia constructed chart of missing and known isotopes and from gaps in the pattern mused on the possible existence of ^2H , ^3H and ^5He . In the same year Raymond T. Birge at Berkeley and Donald H. Menzel at Lick noted[4] that all spectrograph measurements were based on $^{16}\text{O}=16$, but chemical methods based on natural oxygen=16. However the chemical value for the mass of hydrogen was 1.00777 ± 0.00002 on this scale which compared very closely to Aston's spectroscopic value of 1.00778 ± 0.00015 . Hence, the existence of heavy isotopes of oxygen must be balanced by one or more heavy isotopes of hydrogen; if this was all ^2H , then its abundance compared to ^1H must be around 1:4500.

Urey and Murphy set about looking for ^2H in the spectrum of natural hydrogen; the expected shift was very small, $m_e/2m_p$, or 1.8 Angströms on 6565 Angström Balmer alpha line. Seeing only

tantalizing hints of a second line in natural hydrogen they proceeded to distill liquid hydrogen, hoping to leave the heavy isotope preferentially behind. Victor LaMer, a Columbia electrochemist had argued that electrolysis of water would have no effect on the isotopic fraction; it never occurred to them that the huge fractional mass difference between H and D would make electrolysis and excellent separator! In fact it was worse than that because Brickwedde from NBS who prepared the samples chose nice and pure but deuterium-depleted electrolytic hydrogen for the distillation process. Eventually the deuterium concentration in the electrolyte increased with use and the line appeared. After the errors were discovered an isotopic ratio of 1 in 6500 was obtained, very close to the 1 in 7000 value accepted today. Soon afterwards Edward Washburn and Urey[5] used the electrolytic method to prepare the first isotopically enriched sample of heavy water.

Urey had trouble getting travel funds to present his work at December 1931 APS meeting in Tulane; an appeal had to be made to the President of Columbia to provide his travel costs. Two years later he was awarded the Nobel Prize for Chemistry.

As a footnote to Urey's discovery, in 1935 Aston found error in his measurement of the mass of hydrogen[6]. Now he got 1.00813 on physical scale or 1.0078 on chemical scale which implied if anything a lighter isotope of hydrogen. In fact the existence or otherwise of rare hydrogen isotopes was well buried in the experimental errors. But it was too late: deuterium had been discovered.

4 Production

The first concentration of deuterium in water was achieved by using electrolysis; the light isotope of hydrogen was preferentially evolved, leaving behind water enriched in deuterium (see appendix). Separation factors of about six can easily be obtained and double this is possible. The economics are not great, however. It is necessary to electrolyze 2700 litres of natural water to obtain one litre of water enriched in deuterium by 10process of electrolysis, pure heavy water can be produced; this is a conceptually simple method but it is enormously expensive.

Scientific interest in heavy water started to emerge soon after its first production by Lewis and MacDonald[7] as its analytical potential in chemistry and biology, and its possibilities in cancer therapy, as well as more direct application in nuclear physics, became apparent. Small amounts (grams) for scientific use were available in the United States by 1933.

5 Norsk Hydro Enters the Field

Jomar Brun, the Head of Hydrogen Research at Norsk Hydro, and Leif Tronstad, a physicist from Trondheim realized that the conditions for large-scale production (kg) of heavy water existed at Norsk Hydro's plant in Rjukan, where large amounts of water were already being electrolyzed as part of the Haber-Bosch process for producing ammonia for nitrogen fertilizer. They drew up a plan, with some involvement from Karl-Friedrich Bonhöffer, a German physical chemist at Leipzig,

for the industrial production of heavy water. It was in many ways an astonishing venture as a large amount of equipment had to be built - hundreds of combined electrolysis, combustion and condensation cells - and the market must have been uncertain. However, Norsk Hydro went ahead, and built a plant by the generator building at Vemork, just outside Rjukan (fig.1).

A glance at the first order book (fig.3) shows impressive and rapid progress. The first order of one litre of 0.43% heavy water was shipped to Birkbeck College, London, on August 9th, 1934, and by mid-November 95% was achieved (and sent, not unreasonably, to Tronstad's lab in Trondheim). By January 1935, the first production of more than 99% pure heavy water was available at 10Kr (about 50 cents) per gram, a tenth of the American price. Tronstad and Brun themselves did important standards work on deuterium and heavy water[8].

About this time, the idea of producing heavy water at Trail, British Columbia, was first discussed in correspondence between the Canadian National Research Council and the owners of the Trail hydroelectric plant, Cominco[9]. This plant was the largest producer of electrolytic hydrogen in North America; it was used, as was Norsk Hydro's, for making ammonia for the fertilizer and explosives industry.

6 Loss of Innocence

For a few years life with deuterium and heavy water was pleasant and productive. As a target for nuclear physicists to play with deuterium provided an excellent source of neutrons, then a primary tool in fundamental subatomic physics. Paul Harteck (whom we shall meet again), a young German working with Rutherford and Marc Oliphant at the Cavendish Laboratory used it as a stepping stone to discover an even heavier hydrogen isotope, tritium[10]. And although heavy water never panned out as a cure for cancer (it was not possible to utilize the different cell growth rates in heavy and "light" water to this effect), biologists made hay with the material; for example George Hevesy in a classic set of experiments used it to establish the fraction of water in mammalian biology[11].

In 1937, Hans von Halban and Otto Frisch, working at Bohr's Copenhagen laboratory, noticed that heavy water had very low neutron absorption compared to light water. Protons and neutrons like to be paired off; the cross section for neutron capture on a proton to produce a deuteron is much greater than that for neutron capture on the already paired deuteron. This latter event does occasionally happen, producing ^3He , and this in turn loves to capture neutrons to produce the doubly-paired ^4He .

Around Christmas of 1938 the world changed when Otto Hahn and Fritz Strassman at the Kaiser Wilhelm Institute in Berlin discovered that low energy (thermal) neutrons could split uranium nuclei, releasing an enormous amount of energy. Using the liquid-drop model, Hahn's erstwhile colleague Lise Meitner and her nephew Otto Frisch were able to understand the basics of this fission process and deduced the energy release to be about 200 MeV, or about 10 million times larger than the energy change in a typical chemical reaction[12]. In January 1939 Hahn and Strassman[13] suggested that thermal neutron-induced fission of uranium could release secondary neutrons. These

could go on to produce a chain reaction provided they were slowed down (moderated) to thermal energies to increase their chance of causing further fission (see Appendix).

By April, Frédéric Joliot and his colleagues Hans von Halban and Lew Kowarski from the Collège de France in Paris had observed these secondary neutrons, and they measured the number produced in each fission[14]. By August they found that blocks of uranium oxide showed increased fission activity when immersed in ordinary water[15]. However, absorption of neutrons on hydrogen prevented a self-sustaining chain reaction.

Across the Atlantic, Fermi and Szilard at Columbia University examined alternative moderators and quickly decided on ultra-pure graphite. Work on carbon was also begun by George Laurence at the Canadian National Research Council in Ottawa (later joined by Bern W. Sargent from Queen's University)[16]. Szilard started to persuade all physicists working on fission to cease publishing; he sent a cable to Joliot on April 6th requesting a delay in further publications “in view of possible misuse in Europe”[17].

Sometime in early summer, the Paris team alighted on the idea of using heavy water[15] as a moderator. Deuterium was known to have a much lower absorption cross section for neutrons than ordinary hydrogen[18] and its low mass makes it an almost ideal moderator. Use of heavy water would thus make a self-sustaining chain reaction more accessible. Halban and Kowarski did some simple modelling of neutron moderation and this was enough to suggest D₂O as the best candidate[15]. The putative moderating qualities of deuterium were then widely appreciated; as early as the beginning of February 1939 Oppenheimer had a crude drawing on his blackboard of an atomic bomb made of uranium deuteride[19].

At the end of October Halban, Joliot and Kowarski deposited a sealed envelope with the Academy of Science. This was opened in 1949 and the paper inside “Sur la possibilité de produire dans un milieu uranifère des réactions nucléaires en chaîne illimitée” was published in Comptes Rendus[20]. The paper shows the group had a very firm theoretical grasp of reactor physics and includes what we now know as the Fermi four-factor formula. When much later Patrick Blackett wrote Joliot's obituary he said “There is little doubt that, had the war not intervened, the world's first self-sustaining chain reaction would have been achieved in France”[21].

We now know a natural uranium reactor is possible with only three practical moderators: heavy water, ultra-pure graphite, and beryllium. Heavy water is by far the best, but in 1939 it was not available in very large quantities. Graphite was more convenient, being very common, but it had to be extremely pure. The purity of a graphite moderator is crucial as natural impurities tend to be highly efficient neutron absorbers. Early measurements of absorption cross-sections made by Joliot and independently by Walter Bothe and Peter Jensen[22] in Heidelberg were much too large for this reason. Thus carbon was rejected in Europe and this precipitated a fight for the Norwegian heavy water.

After moving to Chicago, Enrico Fermi used 40 tons of uranium and uranium oxide and 385 tons of ultra-pure graphite to achieve the first self-sustaining chain reaction, on 2nd December 1942.

7 La Bataille de L'Eau Lourde[23]

As the commercial and military potential of heavy water sank in, French military intelligence (the Deuxième Bureau) learned that there was considerable German interest in not only obtaining existing Norwegian stocks, but in a contract for large and regular supplies[24, 25]. In March 1940, Lieutenant Allier of the Deuxième Bureau left Paris for Oslo to negotiate with Norsk Hydro. The resulting agreement ensured that France was to have not only the 185kg of heavy water then at Rjukan, immediately, but also a priority claim to the plant's entire output. Allier suspected he was a target for German agents, and took the precaution of double-booking himself and his cargo on both a flight to Scotland, and on one to Amsterdam. It seems his fears were justified, as Luftwaffe aircraft forced the Amsterdam flight to land in Hamburg, where it was thoroughly examined. Allier and his 26 cans of heavy water landed safely in Scotland; then he travelled to the French Military Mission in London, and eventually across the Channel. The heavy water was installed in a special air raid shelter in the Collège de France.

In the summer of 1940, as France faced defeat, Dautry, the French Armaments Minister, ordered Joliot to ensure that his cans of heavy water did not fall into enemy hands. Hans von Halban, his colleague, first took the cargo to Mont-Dore, the spa in central France. He put his wife and one-year-old daughter in the front of the car, one gram of Marie Curie's radium in the back, and, to minimize any possible danger from radiation, the cans of heavy water in between. Upon arrival, Halban was allowed to lodge the cans in the safety of the town's women's prison. The following morning, after they had been moved for safety to the condemned cell of the prison in nearby Riom, Halban began to set up a new laboratory in a small villa. Shortly afterwards, the evacuation order came. A number of prisoners serving life sentences were ordered to move the world's total stock of heavy water from the condemned cell to a waiting vehicle.

The evacuation was to be through the port of Bordeaux. Here Halban found the *Broompark*, a British coaler (fig.4). They were met by the Earl of Suffolk, liaison officer in France for the British Department of Scientific and Industrial Research, who was charged with rescuing rare machine tools, \$10M worth of industrial diamonds, fifty French scientists, and the heavy water. The cargo was strapped to pallets on deck which would float free in the event of the ship being sunk, and thus make rescue a possibility. Joliot chose to remain in France and began a difficult period in charge of the Collège de France cyclotron. This was the only one available to German Scientists in Occupied Europe, for although there was one in occupied Copenhagen, Neils Bohr simply forbade its use by Axis personnel; an act which only Bohr could conceivably have gotten away with[26]. Later in the war Joliot went underground and became a leader of the Resistance.

Meanwhile in Bordeaux, the harbour was bombed, and the *Broompark* sailed down the Gironde estuary amid chaos. The ship next to her was sunk by a mine. (When questioned about the heavy water, Joliot said it was on this ship). Eventually the heavy water reached London, where it was deposited in Wormwood Scrubs prison. It was later moved to the Cavendish Laboratory in Cambridge, where the Collège de France team were setting up to continue their experiments.

8 World War II: The Western Allies

A team coalesced in Cambridge around Hans von Halban and Lew Kowarski. By 1941 their experiments with uranium oxide and the 185kg of heavy water had shown sufficient increase in neutron and fission activity to predict that with 3-6 tons of heavy water, a self-sustaining chain reaction could be achieved[27]. A plan for industrial production of heavy water in Britain by I.C.I. was shelved in favour of an approach to the United States for supplies. However, once the United States joined hostilities and the centre of gravity of fission research moved inexorably westwards, it was decided that the Cambridge team should move across the Atlantic. Chicago was the first choice as that was the site of Compton's group, but this was not possible due to American concerns about citizenship (some of the Cambridge team were citizens of Axis nations) and ties with I.C.I. (a rival chemical conglomerate to Du Pont, chief industrial partner to the American effort)15.

Canada was a workable alternative and had the advantage that some fission work had already been initiated there by Laurence and Sargent. Montreal was chosen and in early 1943, the team, families, equipment, uranium and heavy water made the journey while the Battle of the Atlantic was raging. Once in Montreal, they were to interact freely, at least in theory, with Met Lab team in Chicago. Detailed theoretical work on a prototype heavy water/uranium reactor was to have begun immediately, following the work of Fermi and Wigner on a carbon moderated reactor[28], but was delayed by worsening relations with the Americans. When it was finally started, after a series of visits to Chicago by George Volkoff and George Placek in January 1944, the American design effort was well underway.

The Americans had noted the British request for a heavy water supply in 1941, and Urey and Hugh Taylor of Princeton had suggested the Consolidated Mining and Smelting Company of Trail as a likely source9. The U.S. National Defense Research Committee offered the company \$10 a pound for high grade heavy water; at the time the American product cost \$1,130 a pound[27].

In July 1942 it was clear the cost of Project No.9, as it was known, was going to be enormous, but the U.S. government agreed to pay for it, with the acquiescence of C. D. Howe, the Canadian Minister for Munitions and Supply. The Canadian Government only officially learnt of the project in August 1942 [16]. The final cost was \$2.8M, and the annual operating costs about \$700K. The company produced the heavy water for the American nuclear effort at cost, and did so from 1944 until 1956 when the plant became uneconomical compared to the large U.S. plants which were by then in production[29].

Cominco had produced electrolytic hydrogen since 1930 in a \$10M plant consisting of 3,215 cells consuming 75MW of hydro-electric power. In addition a tower was built for initial concentration (0.015% to 2.3%) by D/H exchange on a catalyst; Hugh Taylor developed a platinum on carbon catalyst for the first three stages and Urey developed a nickel-chromia one for the fourth stage tower. The "P-9 Tower" building housing this plant still stands (figs.5,6). To the electrolysis plant were added secondary electrolysis cells used to increase the deuterium concentration in the water from the exchange process from 2.3% to 99.8% (fig.7, see appendix).

Development work had been done at Columbia and Princeton in 1941. It was found that a catalyst

was necessary and Hugh Taylor established nickel as a likely candidate[30]. The flow chart is shown in fig.13 Many stages of enrichment can be achieved before the necessity of power-consuming electrolysis[31], and thus the Trail operation was as efficient as was possible with the techniques of the day.

In November 1942, C. J. Mackenzie of the NRC requested that the Montreal group receive the first year's output from Trail, about 6 tons. Then followed the rift between the two groups, and in the late summer of 1943, the Chicago group were given permission to build their own heavy water pile at Argonne. Trail heavy water production started in January 1944 and was sufficient for the Argonne pile - the first heavy water/natural uranium reactor - to go critical on 15 May of that year. The project was directed by Walter Zinn, a native of Kitchener, Ontario. In fact, the Americans had decided on a full-scale program to produce plutonium with a heavy water reactor, and DuPont eventually built three additional plants on their own soil, in West Virginia, Indiana and Alabama. These used the fractional distillation technique; this method is very simple but very heavy on power (see Appendix).

In April 1944 the rift between the Montreal group and the Americans was patched up. American assistance, partly in the form of Trail heavy water, was assured for the construction of a Canadian pile. The chosen site was Chalk River. The first director of the new laboratory was to be John Cockroft, although he was replaced before the first pile went critical by W. B. Lewis, who went on to dominate the Canadian nuclear scene for decades. Both men had a pre-war background of nuclear physics and wartime experience of radar research and development in Britain. The pilot pile was known as the NRX, and the 10 MW design was completed in July 1944. However, it was not to be the first Canadian reactor. Cockroft proposed building a smaller zero-energy pile making use of American experience at Argonne[17]. Thus, ZEEP (Zero-energy experimental pile) was built, by Section 6 of Chalk River's Nuclear Physics Division; Kowarski was Section Head and B. W. Sargent was Acting Division Head [16]. Criticality was achieved in September 1945. NRX was completed and went critical in July 1947.

In France in September 1945 Allier obtained a promise from Norway that France would receive the first five tons of heavy water produced once the Norsk Hydro plant got started again. The original 185kg, on loan since 1940 and still in use in Canada, was finally paid for. After ZEEP, Kowarski returned to France and built that country's first pile, ZOE (*Zero Oxide Eau Lourde*) in the old prison at Châtillon. Criticality was achieved on 15th December 1948[32].

As a footnote to history of the 185kg of heavy water, the Norsk Industriarbeidermuseum in Vemork would like to track down one Joliot's original containers for its exhibition. However, after extensive enquiries, it seems likely that these were disposed of when the Montreal lab closed and was moved to Chalk River in 1947.

9 World War II: Germany

In contrast to the Allied effort, heavy water was central to Heisenberg's rather ambiguous fission program in Germany[33]. A large-scale industrial effort like that mounted in the United States for isotope separation was impossible, and the entire program centred on a small amount of unenriched uranium and Norwegian heavy water.

In February 1940 Werner Heisenberg, in Berlin, reviewed the published literature, particularly the work of Bohr and Wheeler which pointed out that it was the likely the rare isotope of uranium ^{235}U which was being fissioned. He decided that to build a reactor from natural uranium required good moderator - D_2O or carbon. If H_2O was to be used as a moderator, then the uranium would have to be enriched in ^{235}U [34]. By summer Walter Bothe, in Heidelberg, had tried the purest form of carbon he could obtain - electrographite from Siemens - but found the absorption too high for natural uranium. They recognized that this was probably due to residual impurities, and worked out further refining steps, but Army Ordnance, their paymasters, ruled out trials due to cost. Robert Döpel in Leipzig had shown experimentally that D_2O was a good moderator; indeed it looked like the only possibility.

There was much experience with D_2O in Germany; Paul Harteck, now at Hamburg, had studied it with Rutherford, Munich physical chemist Klaus Clusius had worked with it, and Bonhöffer had worked with Norsk Hydro from the start. In a discussion as to the best method of producing the material, Bonhöffer favoured electrolysis, while Harteck thought catalysis more cost effective. They contacted the chemical conglomerate I. G. Farben whom they hoped could handle production. External events took a hand in April 1940, when German forces invaded Norway. After the situation was stabilized - the Telemark region was the last part of Norway to be subdued - I. G. Farben was given control of Norsk Hydro. Soon production with the existing plant was increased from 20 litres per year to 1 ton per year.

As experimental work in Leipzig progressed ever more accurate estimates were made of the amount of heavy water required by an uranbrenner (uranium burner, i.e. reactor). This was 4-5 tons, and Harteck and Suess developed a catalytic exchange method (dual temperature $\text{H}/\text{H}_2\text{O}$, essentially the "Trail Method") for raising production to this level in one reactor charge per year. In 1941 the Norwegians were forced to install catalytic plant at their own cost, as their contribution to the Axis war effort. At the end of 1941, possibly in anticipation of the trouble to come, Harteck and Clusius proposed a new plant in Germany. He surveyed the possibilities: the two used in Rjukan, and repeated distillation of hydrogen. However, Army Ordnance preferred Norway as the cheapest option.

Meanwhile, experimental and theoretical work on reactor design was proceeding apace. An alternating layer design was produced by Heisenberg and Weizsäcker's graduate students Karl-Heinz Höcker and Paul Müller. In addition to Döpel working with uranium metal and heavy water in Leipzig, Karl Wirtz at Berlin-Dahlem did experiments with uranium oxide and paraffin. It looked increasingly like D_2O would work. In addition, Weizsäcker started exploring the explosive possibilities of element 94, which would inevitably be produced in the uranbrenner. Despite this, in early 1942, Army Ordnance decided fission was irrelevant for the war effort and proposed suspending

research.

By the end of 1942, the picture was mixed. The first neutron increase (13%) in a heavy water/uranium pile, the L-IV, had been observed by Dpel and Heisenberg in Leipzig. However, the catalytic plant in Rjukan was still under construction so heavy water was still in short supply. In addition there had been an attempted commando raid on Rjukan; it had been a disastrous failure and the fate of the commandos makes unpleasant reading[35], but it was a sign of things to come.

In a second commando operation, on 28th February 1943, Norwegian saboteurs guided by Tronstad (in London) and Brun (locally) destroyed the Rjukan electrolysis plant, with the loss of 500kg of heavy water. This highly economical and clinical operation has long been celebrated in Norway, particularly in the Rjukan area, where residents retrace each year on July 1 (note not in February!) the steps of the commandos down from the Hardanger Plateau where they gathered prior to the attack. In a war that killed maybe 50 million people, this courageous action, which resulted in no injuries and, mercifully, no reprisals, is well worth remembering.

The plant was, however, quickly repaired and General Groves, fulminating against British control of Norwegian special operations, ordered the job to be done properly, by massive air bombardment. This followed in 16th November 1943 and resulted in considerable loss of civilian life and minimal damage to the electrolysis building. Clearly the Allies thought nuclear fission worth pursuing, even if the German Army didn't. It was decided to ship all the heavy water and its production to Germany, mostly because of the fear that further raids might endanger explosives production at Vemork. On 20th February 1944, the last shipment of heavy water from Rjukan to Germany was lost when the ferry carrying it across Lake Tinnsj was also sabotaged. This was the last time Rjukan people died in what Abraham Esau described as the "Norwegian National Sport" of destroying heavy water plant. Norsk Hydro received no payment for the heavy water produced, and had suffered 16 million kroner of damage to the plant.

The effort to build a plant in Germany dragged on for the next year, mired in bitter price and patent disputes with I. G. Farben. The end of heavy water production caused severe difficulties for the experimental groups, and air dilution was downgrading existing stocks. Attempts were made to build a polishing plant using equipment looted from Norway. Air attacks on Hamburg and Berlin forced the evacuation of research labs to the countryside.

Heisenberg and his team moved to Haigerloch, near the Swiss Border, where they hoped they could work in relative peace. The laboratory was constructed in the side of a cliff beneath an old church. They were still trying to achieve criticality with the B-VIII reactor when first contact was made with American forces: the fact-finding "Alsos" group[36]. With lingering worries about the German nuclear threat, and always mindful of post-war commercial competition and the fact that Haigerloch was to be in the French zone, an American army team dynamited the laboratory before handing the region over to their allies, who were just then entering in the town.

Why did they fail? There are many reasons as the reader will have gathered, but perhaps the most pivotal is that there were never more than 70 scientists involved, and only 40 worked more than half time on the uranbrenner. (This is in stark contrast with the \$2 billion Manhattan Project). Only a handful of these scientists were of the first rank; Germany had lost its best to emigration,

and of those that remained the most brilliant worked in aerodynamics and rocketry.

Although never used in Germany, Karl-Hermann Geib in Leuna in 1943 developed what we now regard as the most cost-effective process for producing heavy water: the dual temperature exchange sulphide process (see appendix and fig. 10). Contemporaneously, the process was also developed by J. S. Spevack at Columbia University[37], and his process became the basis of the post-war North American plants under the name of the Girdler Process, named after the company which first exploited it. North American scientists were not aware of Geib's work for many years after the war; Maloney et al. in their book "The Production of Heavy Water" (1955)[38] complain that relevant German wartime work was still classified.

Unfortunately Geib was not able to benefit from his work; in 1945 he was taken to the USSR, along with many others, who were given a 10-year contract to work on fission and aerodynamics. Many German scientists found this very congenial and some even went as far as to describe these 10 years as the time of their lives. However, Geib was not so happy and he made the mistake of applying for asylum in Canada, giving the name of Professor E. W. R. Steacie as a reference. Officials at the Canadian Embassy in Moscow did not know what to do with him and told him to come back the next day and that was the last time he was seen. His wife in Germany received his effects in the mail[33].

10 World War II: The Soviet Union

In October 1939 Zel'dovich and Khariton reviewed Joliot's and Fermi's results, and concluded, correctly, that heavy water and carbon were the only feasible moderators for a natural uranium reactor[39].

In August of 1940 Kurchatov, Khariton, Flerov and Rusinov submitted a plan to the academy "On the utilization of the energy from U fission in a chain reaction". Khariton calculated 2.5t of uranium oxide and 15t of heavy water were needed for a reactor. The shortage of uranium was the main problem for experimenters; ironically the Soviet Union, in spite of its huge size, had no uranium mines. Kurchatov sent young colleagues to Leningrad photographic shops to buy uranium nitrate.

In 1941, German forces invaded the Soviet Union, and Flerov found himself in the Red Air Force near Voronezh. The previous year he and Petzhak had discovered the spontaneous fission of uranium and he was interested to see if there had been any reaction to this event from his colleagues abroad. Looking in the local university library he found that all the scientists he knew to be working on fission had disappeared from the literature. This was the first clue Soviet scientists had of what was going on in the west. Flerov sent a letter to Stalin, warning him. Soon a concerted Soviet fission effort was initiated, with Kurchatov in charge.

By March 1943 Kurchatov was receiving Fuchs' reports and was abreast of work in the West. At this time there was only 2-3kg of heavy water in country, and partly because of this, and because

General Groves had restricted heavy water exports from the United States, they chose graphite for their first reactor. More uranium was needed than with heavy water but electrolytic graphite was available in the country. A heavy water plant had been planned before the war at a nitrogen plant in Chirchik, Tajikistan, but it had not been completed. Alikhanov was given the more distant task of building a heavy water pile, as he would not work under Kurchatov and this was a convenient way of removing him from the main group. Construction began after the war.

In late 1943 the Soviet purchasing commission in the U.S. obtained 1kg of heavy water a further 100 g in February 1945. Given the munificence of these gifts, it seems as if the early decision to concentrate on graphite was a good one! On December 25th, 1946 the graphite reactor F-1 went critical.

Work on the Chirchik Nitrogen Combine plant started in 1944; the Institute of Physical Chemistry in Moscow started research on the physical chemistry of electrolytic cells at the same time. After hostilities ceased in Europe, the NKVD assembled German heavy water experts at the Leuna works in Merseberg, where they designed a new heavy water plant. In October 1946 they moved to Institute of Physical Chemistry in Moscow. The plant producing large quantities by 1948. Alikhanov's reactor at the Thermotechnical Institute in Moscow went critical in April 1949.

11 Postscript

Since World War II only Canada amongst the industrialized nations has pursued the commercial possibilities of heavy water/natural uranium reactors. A large number of CANDU (Canadian-Deuterium-Uranium) reactors are now operating in Eastern Canada, and the reactor and heavy water technology has been exported to several other countries. A downturn in the nuclear industry in the 1980s led to one thousand tonnes of heavy water being made available for use as a neutrino detector in the Sudbury Neutrino Observatory, which is now operational in Northern Ontario. I am a member of this effort, which is how I became interested in the subject of this paper.

Acknowledgments

The author would like to thank the Cominco personnel Richard Fish and Brent Cross for their assistance regarding the history of Project 9 and a fascinating tour of the derelict Trail heavy water tower in 1995. One of the photographs is reproduced with permission from the Cominco Magazine. I thank Norsk Hydro chemist and historian Per Pynten for a marvellous day at Vemork and Rjukan in 1996. I am indebted to Professor Maurice Pryce for pointing me toward several valuable references, and to the late George Volkoff for reading an early version of this manuscript.

Many thanks to Don Cody of California, Captain Paulsen's grandson, for information about, and the photograph of, the Broompark.

Afterword

This article was written between 1992 and 1998. In 1999 Per Dahl's book "Heavy Water and the Wartime Race for Nuclear Energy" was published. Dahl covers many aspects of this story in great detail and the book is highly recommended to interested readers.

12 Appendix

12.1 Separation of Deuterium and Heavy Water

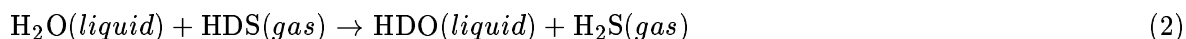
There are four basic methods of producing heavy water, they have different economics depending on the initial and final concentrations, and so they tend to be used in combination.

1. Distillation of water or hydrogen. Heavy water boils at 101.4C, only slightly higher than light water, so the process requires many stages and a large amount of thermal energy. The separation effect is bigger for liquid hydrogen, but this is less convenient to work with than water.
2. Electrolysis of water. H^+ ions are evolved preferentially over D^+ . As above, multi-stage and heavy on electrical energy, now used as final "polish".
3. Hydrogen/water catalytic exchange, the "Trail Method", depends on the fact that, when isotopic equilibrium is established between hydrogen gas and water, the water contains 3 or 4 times as much deuterium as does the gas. See figure 13.



This method can also be used in dual-temperature mode (see point 4 below)

4. The Dual-Temperature Sulphide Process relies on the following reaction:



Deuterium enrichment in the water increases as the temperature decreases. This method has two big advantages over the Trail method. The use of hydrogen sulphide eliminates the need for a catalyst, and the dual temperature aspect means that the hydrogen sulphide is maintained in a closed loop (see figure 10) and no electrolytic regeneration of hydrogen is required.

Modern Practice (Bruce Plant, Ontario)

* Three-stage dual temperature sulphide process (0.015% - 25%)

* Distillation with waste reactor heat (25% - 99.85% reactor grade)

12.2 Neutron-Induced Fission

A slow neutron is absorbed by a ^{235}U nucleus, which splits into two medium-mass nuclei, emitting 200 MeV of energy and, on average, 2.5 fast neutrons. For example:



To promote a chain reaction, the outgoing fast neutrons have to be slowed by collision with nuclei in a moderator, without absorption.

12.3 Moderators

The best moderator would be hydrogen, whose nucleus has a mass equal that of the neutron; all the neutron's energy can be lost in one collision. However, hydrogen absorbs neutrons, producing a deuterium nucleus (D; or ^2H) and a gamma-ray:



In reality the best moderator is deuterium, which cannot easily absorb a second neutron. The most convenient form of deuterium is Heavy Water (D_2O).

Carbon also makes a good moderator, but tends to have neutron-absorbing impurities; it therefore has to be very pure (Electrolytic Graphite).

13 Figures

Fig 1: Author at hydro-electric generator building at Vemork, just outside Rjukan, now the Norsk Industriarbeidermuseum (Industrial Workers Museum). The heavy water plant was in a (rather unattractive) building, now demolished, in front of this one. (Author photo).

Fig 2: Part of the electrolysis plant from Rjukan, now reassembled in the Resistance Museum, Oslo. The longer steel cylinders are for electrolysis and the shorter ones are the combustion chambers. The brown tubes above are condensers. (Author photo).

Fig 3: Norsk Hydro heavy water order book, pages 1 and 3. (Per Pynten).

Fig 4: The Broompark sailing out of Le Verdon Roads in 1940 with the supply of heavy water, under the command of Captain Olaf Paulsen. The Broompark was built in 1939 and torpedoed and sunk in 1942. The Denholm Shipping Lines office in Glasgow have a glass-encased six-foot model of her. (Don Cody)

Fig 5: The Trail Heavy Water tower in the 1940s. (B.C. Archives and Records Service Cat. No. NW669 C733 V.30-32/1).

Fig 6: The Trail Heavy Water tower in 1995. (Author photo).

Fig 7: A few of the secondary electrolytic cells at the Trail Plant (B.C. Archives and Records Service Cat. No. NW669 C733 V.30-32/1).

Fig 8: One of the containers used for shipping Trail heavy water, now in the museum at Rossland, B.C. (Author photo).

Fig 9: Trail method for D₂O enrichment.

Fig 10: Sulphide method for D₂O enrichment.

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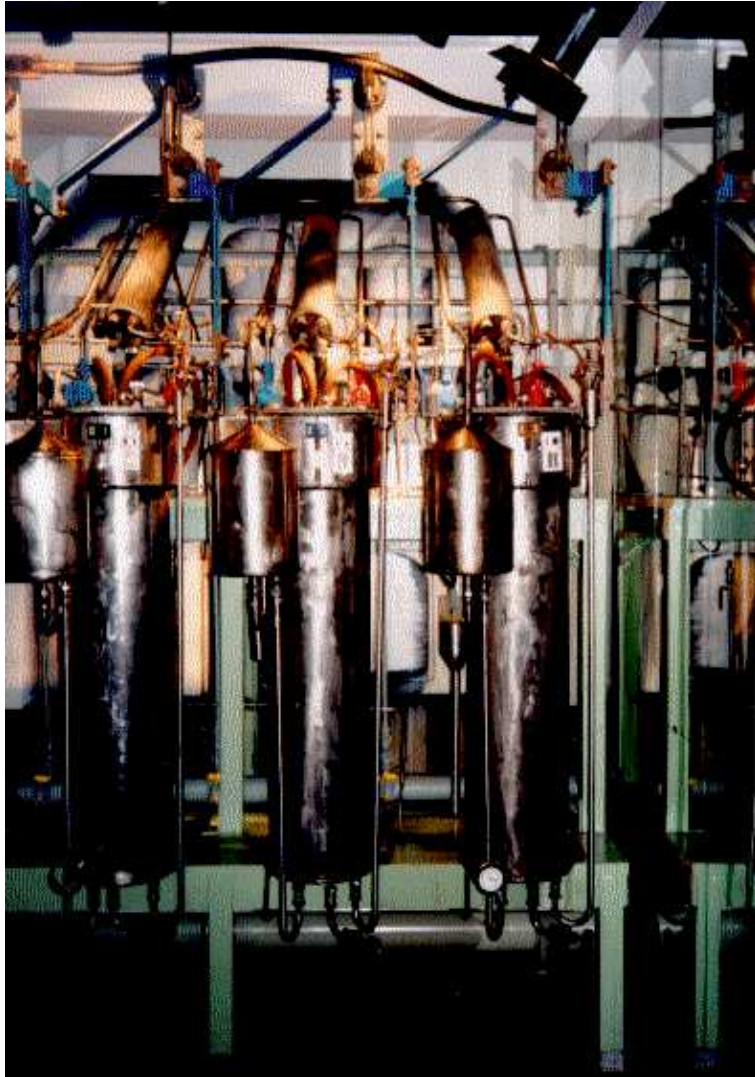


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1937							1937						
Date	Sol.	address	%	litre	amount		Date	Sol.	address	%	litre	amount	
Aug 9	B.T.O. Am. U.S.C. Lab.	% Standard Storage University of London Bancroft B.P. England	0.45	1.0	9% 34	0.070	10/11	Uranium 235 miste kobaldrum	Getinsid. of B Folshovum W.	10.2	200 gr	14/11-31	1098
Aug 9	Staff. G. Aveline	May. Vera Höger- lund typiska Insti- tution. Stockholm	1.73	1.0	9% 34	0.159	11/11	H. H. and P.	Oslo	85.2	10 g	14/11-31	---
8	Lord Taylor & S.	Leeds. 7 Oct	0.40	10.0	9% 34	0.179	16/11	Prof. E. K. Baford	University of London, Dept. of Chemistry Riverside Lodge W. B. I.	2	2000 g	14/11-31	1168
Sept 25	D. M. P. Hobbey	Phys. Clinic, Bristol Bristol Bristol on - West England	10.0	2	23% 24	6/10	"	"	"	20.8	100 g	---	---
Sept 28	North. Kaye and S.	Leeds. 7	10%	1000	23% 24	---	17/11	D. L. Kærsted	H. H. Fordleim	95.2	200 g	14/11-31	1168
1	Metabolisk & Kemi	Carlsbergsgatan 11 Stockholm	10%	20 gr	9% 34	7.738	20/11	Dr. H. J. A. F.	Trin. P. S. S. S. S. S.	50.2	1 g	14/11-31	1189
6	Norsk Hydro and S.	Leeds. 7	20%	1000	9% 34	---	"	Dr. H. J. A. F.	Trin. P. S. S. S. S.	50.2	1 g	14/11-31	1189
15	and P.	Leeds. 7	20%	1000 Prove	14/11-31	---	"	Dr. H. J. A. F.	Trin. P. S. S. S. S.	50.2	1 g	14/11-31	1189

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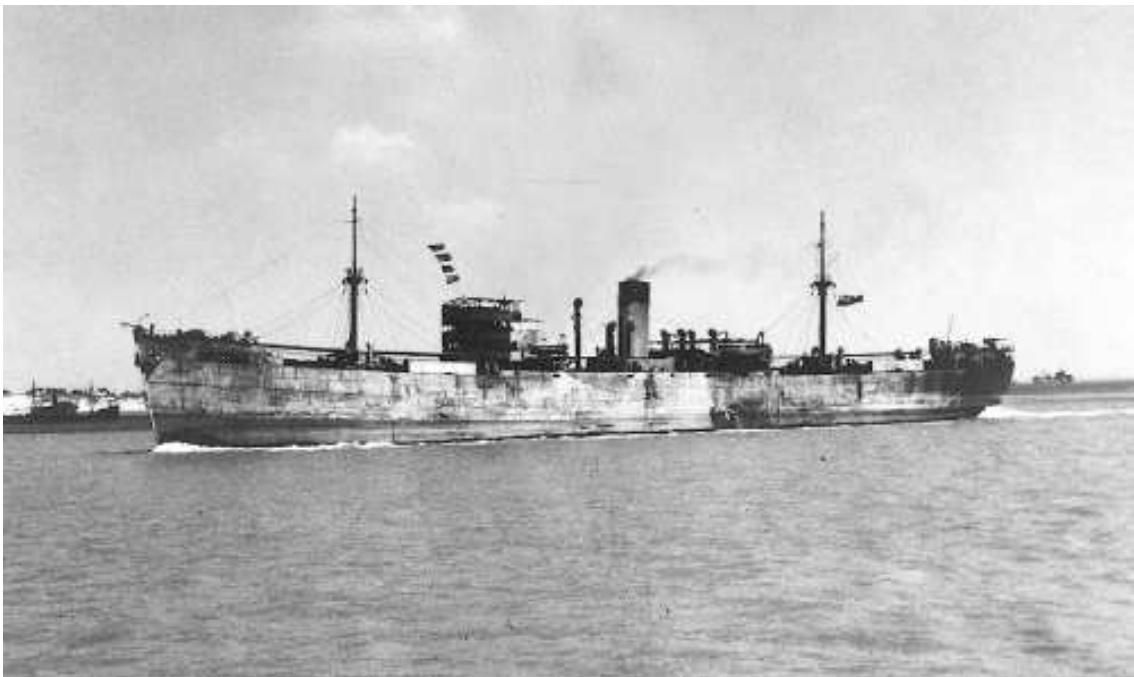


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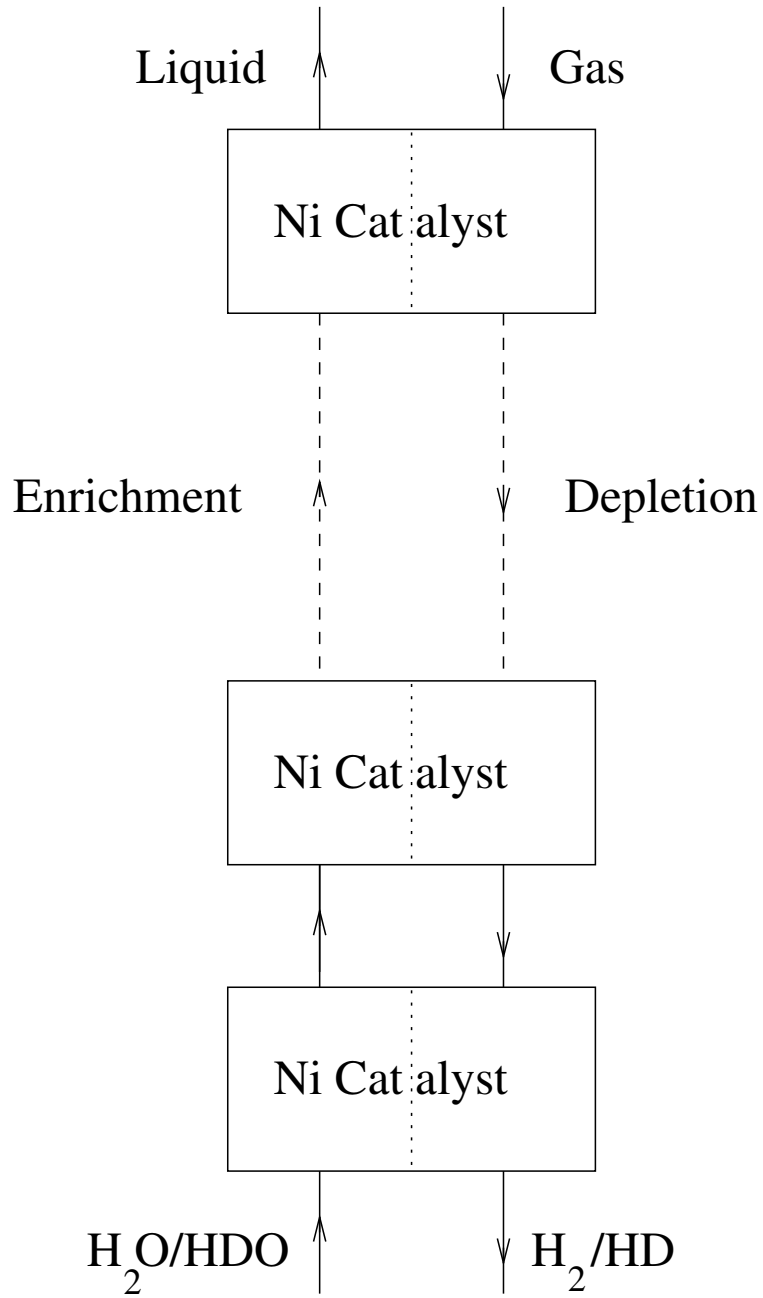


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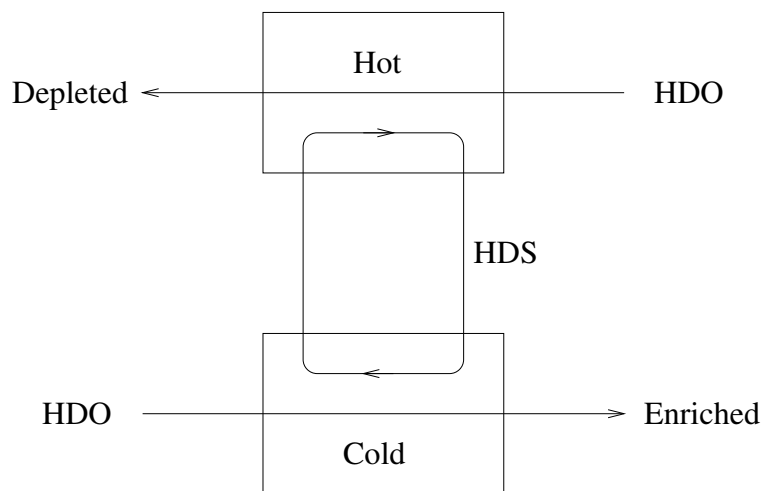


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