Evolution of Cryogenic Engineering

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Abstract. Individual scientists began to address technology problems that would fall in the category of cryogenics in the half century before 1900. Air was liquefied and, in several steps, distillation columns for air separation evolved. The need for pure oxygen and nitrogen motivated industrial improvements and a technology spinoff. Institutions with a department of cryogenic technology emerged in the Netherlands and United Kingdom along with isolated individual scientists. The need for pure oxygen for high altitude aviators in World War II gave local air separation a boost. Military applications provided a strong incentive for cryogenic development until the mid 1950's. After the first Cryogenic Engineering Conference in September 1954, the technology would become the basis for space flight, superconductivity, high energy accelerators and other low temperature research applications. The introduction of multilayer insulation (MLI) in 1957 had a significant impact on all sizes and forms of liquid dewars. Liquid cylinders, field and storage artificial insemination dewars, liquid helium transport dewars and a wide range of technical applications ensued. Cryogenic engineering has matured and has proven equal to the challenges presented.

1. Introduction
Cryogenic technology evolved in two directions. First, technology was invented to solve essential problems. Second, technology went looking for applications as the body of cryogenic engineering knowledge improved. The focus of this paper is a chronological tracing of cryogenic developments from about 1850 to the present with special emphasis on several key events.

2. Pioneering Development
1824: Sadi Carnot – Basic definition of the Carnot Cycle as the measure of ideal process efficiency. [1]
1877: Raoul Pictet and Louis Paul Cailletet, working separately, each developing methods to liquefy oxygen. [2][3]
1892: James Dewar – Invents the vacuum insulated, silver-plated cryogenic glass liquid container – the Dewar vacuum flask. [4]
1895: Carl von Linde – files for patent on the Hampson-Linde cycle for liquefaction of atmospheric air or other gases. [5]
1898: James Dewar – Condenses hydrogen using regenerative cooling and his dewar catch vessel. [6]
1902: Georges Claude – Invents a cycle which uses an expansion engine to do work and produce cooling in a liquefaction or air separation system. [7]

1905: Carl von Linde – Makes pure oxygen and nitrogen using a double distillation column. [8]

1905: Hamilton P. Cady and David F. McFarland (Professors at the University of Kansas, Lawrence) – Discovered that the inert gas in Kansas natural gas wells was helium. This moved helium from the rare gas category to a more readily available commodity. [9]

1908: Heike Kamerlingh Onnes (Leiden) – Assembles a cascade of cryogenic fluids to liquefy air. [10]

1908: Heike Kamerlingh Onnes – Liquefies helium. [10]

1908: Linde – Installs first air separation plant near Buffalo, New York. [11]

1911: H. K. Onnes – Discovers and names superconductivity in metals. [12]

3. Developments 1911 – 1950

This period was marked by expansion of the industrial gas industries and improvement of air separation technology. Many smaller air separation plants were produced to supply high purity oxygen for the pilots and crews of high altitude military aircraft.

Steady technology improvement in this era was accompanied by some noteworthy achievements including the following:

1933: William F. Giauque – Demonstrated the technology of Adiabatic demagnetization refrigeration. This was only one of his many achievements for which he was awarded the Nobel Prize in Chemistry in 1949.

1947: Samuel C. Collins – Invented the Collins Cryostat with which technically limited personnel could safely perform research in liquid helium. The system for liquefying helium was widely used for other applications over a period of many years. The Cryogenic Engineering Conference honored Dr. Collins by presenting the Collins Award to several people since then who have made significant contributions to cryogenic engineering technology.

1948: Harold W. Wooley, Russel B. Scott and F. G. Brickwede (Research Paper #1923) Compilation of Thermal Properties of Hydrogen and its various isotopic and ortho-para modifications. This is the foundation for all hydrogen technology.

An example of the need for competent cryogenic engineering occurred in Cleveland, Ohio on October 20, 1944. A liquid natural gas storage tank, not a dewar, with a gaseous equivalent storage capacity of 89,468 cubic meters started leaking in a crack, presumably in a weld seam. The leaking gas caught fire and then exploded. Another explosion followed and there was burning LNG running down the street and into the underground sewer system. This tragedy led to the loss of 130 lives and burned 79 homes, 2 factories, 217 automobiles, 7 trailers and 1 tractor.

4. Developments 1950 – 1953

1950: On March 10, U.S. President Harry Truman approved a plan to develop a nuclear fusion weapon, the so-called Hydrogen Bomb. At that time, the weapon concept was to use an appropriately sized atomic bomb to provide the heat and pressure necessary to initiate fusion of a volume of liquid deuterium. Significant volumes of liquid hydrogen would be require for
all of the equipment testing to limit the need for expensive deuterium which was obtained from heavy water. Key properties for hydrogen 20.21K boiling point and density of 70 kg/m³. A boiling point of 23.706K and density of 162.4 kg/m³ for deuterium.

The Atomic Energy Commission (AEC) decided that there should be a facility for volume liquefaction of hydrogen and for training personnel in cryogenic engineering. The AEC selected the National Bureau of Standards (NBS) to perform those functions. After a search and negotiations, the city of Boulder, Colorado gave the government an adequate site adjacent to the Boulder city limits. Meanwhile, the NBS technical staff under Russell B. Scott was busy designing a hydrogen liquefier for fabrication in the NBS Washington shop. Actually, four hydrogen liquefier trains were built, with two going to Boulder, one to Eniwetok Atoll for the Operation Ivy “Mike” test, and one was kept for spare parts. This was a signal achievement since none of these people had designed a hydrogen liquefier and few had any design experience. However, there were some stalwarts on the team, including (but not limited to) Bascom Birmingham, William Gifford and Victor Johnson. Program leadership was provided by Dr. E. J. Hamel from Los Alamos.

As the project got under way, NBS contracted with Denver engineering and plant construction firm Stearns Roger Manufacturing Company. Stearns grew out of the mining industry in Colorado and was able to provide a vital reserve of experience for the Boulder facility. Effectively, NBS specified what was needed and Stearns Roger provided everything that was needed to produce a safely operating hydrogen liquefier. Work priority was first for the liquefier, “A” building, then the building housing hydrogen-safe laboratories, business and technical office space and a shop for fabricating research equipment. Logically, the second building was called the “B” building. As the scope of activities shifted from Washington to Boulder, Bascom Birmingham moved to Boulder to provide on the scene coordination with Stearns Roger.

The job was completed in March 1952 in what was a unique educational experience for many people. The “world’s Largest Hydrogen Liquefier” operated safely at a production rate of 340 liters per hour of normal liquid hydrogen. At this point, the NBS focus shifted to cryogenic engineering as the test labs became available.

One immediate task was to identify a catalyst to convert normal hydrogen (75% Ortho and 25% Para) to high concentration Para hydrogen. Studies to identify a workable catalyst were undertaken by NBS Boulder, the University of Colorado Chemical Engineering Department, NBS Washington and Los Alamos. NBS Washington found a workable material and both liquefier trains in Boulder were modified to add catalyst to the liquid receiver. In March 1953, a liquefier containing catalyst produced 240 liters per hour of 95% Parahydrogen to successfully conclude this research effort.

In the same time frame as the NBS program, Los Alamos contracted with the Cambridge Corporation (CAMCO) to design and fabricate equipment for the upcoming Operation Ivy “Mike” test device [CAMCO was a joint venture of Carrier Corporation and Arthur D. Little, Inc which provided industrial capability and a high level of technical “know-how”]. CAMCO designed and built a refrigerated transport dewar (RTD) which consisted of a 2000 liter hydrogen/deuterium dewar with a liquid nitrogen thermal shield. This dewar was mounted on a large flat-bed trailer which housed a diesel powered electric generator and a Collins Cryostat. This system supplied cold helium so that the dewar could hold liquid hydrogen or deuterium without loss. This was particularly important for the valuable and scarce deuterium. CAMCO also developed vacuum-jacketed piping with personnel trained to support the Ivy “Mike” test device.

As the NBS facility began to come on stream in Boulder, CAMCO built a test and operations facility in a fenced area just west of the NBS liquefier building. This made it convenient to get test liquid hydrogen and to facilitate filling RTD dewars to supply liquid to Los Alamos. All of this work came into focus on November 1, 1952. The Operation Ivy “Mike” shot was set up on Elugelab Island in Eniwetok Atoll in the Marshall Islands. The test device was filled with liquid deuterium and fired. A successful 10 megaton explosion resulted. This confirmed the feasibility of the “Hydrogen Bomb” weapon and opened up a new phase of activity requiring substantial cryogenic engineering.
For the new bomb to be effective, there was a need for support equipment and trained personnel to use it. The Operation Castle “Echo” test was planned to confirm the feasibility of a deuterium fusion weapon for use in military applications. Another test in Operation Castle, unknown to the crew of “Echo”, was the “Bravo” shot.

The Air Force cryogenic contracting was extensive and included at least the following:

NBS and its fabrication partner, Stearns Roger Manufacturing Company with work scope:
1. Design and fabricate a 750 liter tactical liquid deuterium dewar.
2. Design and fabricate a 200 liter deuterium storage dewar.
3. Design and fabricate a closed cycle refrigerator to maintain a tactical dewar with no loss. This was a concept of NBS engineer Peter van der Arend.
4. Arrange for on the job training of approximately 50 Air Force officers and enlisted men.
5. Prepare and present an educational lecture series on cryogenics for the Air Force personnel and the field engineers hired by Stearns Roger. A final exam was given and grades were passed on to the Air Force and Stearns Roger.

Herrick L. Johnston, Inc also had an Air Force contract covering at least the following work scope:
1. Design and fabricate a portable hydrogen/deuterium liquefier.
2. Design and fabricate a 750 liter tactical deuterium dewar.
3. Support personnel on Eniwetok for Castle “Echo”.

Cambridge Corporation may have had both Air Force and Los Alamos contracts. During the period from November 1, 1952 through April of 1954, the workforce in Boulder was expanded and the staff on Eniwetok numbered approximately 30 from January through March 1954 for the Castle “Echo” test.

5. Castle “Bravo”
The “Bravo” test of Operation Castle proceeded in parallel to and unknown to the crew of “Echo”. The huge difference between the two was that Castle “Bravo” was based on non-cryogenic lithium-6 (and lithium-7) deuteride. On the morning of March 1, 1954, the observers, including 4 people from NBS, plus Castle “Echo” personnel were directed where to stand with sun glasses in place. Then, Castle “Bravo” was fired in Bikini Atoll for a cataclysmic 15 megaton explosion. Castle “Echo” was now dead and everything involving liquid deuterium was on the way out. For many people, this too was cataclysmic.

6. Post Castle “Bravo”
There was an orderly departure of observers and non-participant technical people back to the U.S. For the most part, equipment specific to Castle “Echo” was left at Eniwetok with general use instrumentation and other small items returned to NBS or elsewhere. With the people it was different. Cambridge Corporation was subjected to an orderly shutdown and all employees were placed in arranged new jobs elsewhere. The 50 or so Air Force personnel assigned to temporary duty in Boulder were reassigned. The Stearns Roger field engineers were terminated except for two or three who were absorbed by the company.

With NBS, the end of the cryogenic bomb was a blackout. The whole purpose of NBS Boulder was to support the bomb and to create relative engineering technology. There was no ready backup funding. Air Force contracts ended on March 30, 1954 and Los Alamos support was uncertain. The Air Force did provide closeout funding for NBS and Stearns Roger to test performance of the tactical dewar and related no-loss refrigerator at Kirtland AFB in Albuquerque, New Mexico. Herrick L. Johnston was funded to provide engineering to operate a portable hydrogen liquefier which the company had designed and built under contract. The liquifier was used to supply liquid hydrogen for the NBS testing.

After April 1954, there was great uncertainty about the future of the NBS Boulder operation. Some people started to leave. Mr. Scott was a long time U.S. government employee and experienced
bureaucrat. His successful career was based on doing good work on government specified and funded projects. He did not go storming off to Washington to demand funded projects for NBS Boulder.

Mr. Scott did not propose having a Cryogenic Engineering Conference, nor did he initially advocate it. The Cryogenic Engineering Conference was proposed and actively advocated by NBS Senior Project Leader M.M. (Mack) Reynolds. His pitch was that a lot of cryogenic engineering had been done on the bomb project and the people who had done it would like to tell about it and that many others would like to know about it. He was right!

Mack Reynolds pushed hard and got the permission of Mr. Scott and Bascom Birmingham to make his pitch to the administrative head of all NBS operations in Boulder, including the Cryogenic Engineering Division. Permission to hold the CEC was granted along with the provision to use the Radio Lab’s newly completed auditorium for the technical sessions. Mack Reynolds got help from other technical people, but he was assigned to be the principal organizer of the first CEC.

Word was sent out about the new CEC with a call for papers. The response from both potential attendees and paper presenters was swift and surprisingly large. People that NBS personnel had never heard of signed up. Of course, all NBS technical workers were encouraged to submit a paper. The meeting was held September 8-10, 1954. There could have been many more papers had there been more time allowed or use of parallel sessions. The list of attendees printed in the CEC Proceedings is still impressive because so many key people in the cryogenics industry were in attendance. Of these, two individuals new to cryogenics were destined to make future contributions. First, Ray “Helium” Brown of the U.S. Navy Bureau of Aeronautics, Helium Division became the father of international shipment of liquid helium. Second, Klaus Timmerhaus of the University of Colorado Chemical Engineering School was introduced at the conference but was not a registered attendee. Professor Timmerhaus came to be the lead planner and organizer of the CEC for many years. With great energy, he made many things happen, in addition to his personal contributions.

Most importantly, the 1954 CEC brought cryogenic engineering into the technical vocabulary. People and government activities began to take notice. The feasibility of volume liquefaction of hydrogen drew the attention of the Air Force for use as an aircraft fuel and for rocket launches. The code term for liquid hydrogen was SF-1 and several liquefiers were built to supply fuel for testing. Vacuum jacketed tanks and piping became more commonplace and the advantages of using mass spectrometer helium leak detectors was soon recognized. New technology and potential markets spawned growth of smaller companies to manufacture equipment and components independent from the larger industrial gas companies. (This was not true of the Linde Division of Union Carbide, which was a product leader.)

7. Post 1954 CEC Cryogenic Activities

- Industry was poised to move ahead after the cryogenic bomb was made obsolete.
- Although many people left, the NBS Boulder Cryogenic Engineering Laboratory did survive. It made notable contributions to evacuated powder insulation, high activity Ortho-Para catalyst, materials testing and ongoing cryogenic physical data research. They also continued to provide technical support to government agencies and the University of California, Berkeley. NBS data publications have continued to be of great value to all users.
- The U.S. Air Force had aspirations for use of hydrogen as an aircraft fuel. Development of external fuel tanks was funded by Beech Aircraft, who established its Boulder, Colorado activity for this purpose. Mr. Paul M. Ordin of NACA (not yet NASA) Lewis (now NASA Glenn) laboratory not only worked on development of an external hydrogen tank, but designed a fuel system for one engine of a twin engine jet bomber. Paul rode in the airplane on its test flight and managed to switch one engine from jet fuel to hydrogen in sustained flight at altitude and back to jet fuel for descent and landing.
- Other, greater and more secretive plans were in the works. Project Suntan envisioned a high altitude, Mach 2.5 reconnaissance airplane fueled with liquid hydrogen. This became the
Lockheed CL-400 aircraft. The CL-400 required large volumes of liquid hydrogen for flights to test aircraft structure and the Pratt Whitney engines. Ultimately, four liquid hydrogen plants were built. One was built by Stearns Roger near Bakersfield, California for use by Lockheed. Another was built near Painesville, Ohio (“Baby Bear”) for use by Pratt Whitney and nearby NACA Lewis Research Center. A larger liquefier was built adjacent to the Pratt Whitney test facility in Florida (“Mama Bear”). When this plant proved inadequate, a still larger plant was built nearby (“Papa Bear”). When the CL-400 Project was canceled in 1958, the output became available to the space program and other NASA activities.

• At the Conference, Mr. Ray Brown of the Navy had asked Mr. Scott if it were feasible to build a dewar to transport liquid helium. Mr. Scott asked NBS engineer Glen McIntosh to make the necessary calculations. After Scott pointed out that the calculations should be based on internal energy, not enthalpy, the calculations were done again and results indicated that it was feasible to build a state of the art helium dewar. This lead to further work, and ultimately the fabrication of an over-the-road helium trailer.

8. Other Developments of the 1950’S

• Intercontinental Ballistic Missiles were being developed. This required rocket motors, mostly burning jet fuel with liquid oxygen. These systems required related cryogenic piping, valves and storage dewars.

• Linde Division of Union Carbide introduced multilayer insulation (superinsulation) based on a Swedish paper. This was a very important innovation. It made liquid cylinders, field and storage artificial insemination dewars practical. Helium transport dewars, space dewars and many physics applications were now possible.

• In 1955 and 1956, NBS cryogenic engineers worked with Dr. Luis Alvarez of the University of California Berkeley to design a 2-meter liquid hydrogen Bubble Chamber. The design called for a hydrogen refrigerator with four independent zones of control. This refrigerator was designed and built in Boulder by Beech Aircraft and then tested at the NBS liquefier compressor facility. The Bubble Chamber was successfully used by Dr. Alvarez in his work which led to the award of the 1966 Nobel Prize in Physics.

• In 1957, John Bardeen, Leon N. Cooper, and Robert Schrieffer invented practical superconductivity. They were awarded the 1972 Nobel Prize in Physics for this work. Superconductivity has many technical applications as well as its use in MRI medical devices. [13]

• In early 1958, the Boulder, Colorado Division of Beech Aircraft responded to a NASA inquiry for small oxygen and hydrogen dewars for space applications. These dewars would be used for breathing oxygen and fuel cell electrical generation. Beech successfully responded with supercritical units with thermodynamic vents. All U.S. manned space vehicles had Beech dewars until 1989 when Ball Aerospace bought the business and continued to manufacture the dewars in Boulder, Colorado.

• At the 1959 CEC in Berkeley California, William Gifford and Howard McMahon introduced the Cryocooler. Gifford had rights to manufacture these coolers and he established CRYOMECH to produce them.

9. Cryogenic Engineering in the 1960’s and Beyond

By 1960, cryogenic engineering technology had matured to some extent. However, there were new challenges to be resolved and new engineers to assimilate the cryogenic basics. Some of the new technology and applications are described in the following:

• International shipping of liquid helium from the U.S. became commonplace. At the start, volume liquid helium was so valuable that it was viable to ship it by air in 1,000 gallon dewars developed for this purpose by Cryenco.
• More powerful collider type particle accelerators were designed and built. The new CERN unit is most noteworthy. Before its funding was withdrawn, the Texas Superconducting Supercollider was destined to be a leader with its 4.5K continuous cooling loop.
• For space flight, subcooling of liquid oxygen and hydrogen prior to launch improves overall efficiency. Oxygen can be subcooled in real time by flowing the LOX through a heat exchanger submerged in atmospheric pressure liquid nitrogen. Cooling the LOX from a supply temperature of 93K to just over 78K is regularly achieved. Less expensive subcooling can be accomplished by continuously subcooling the supply with a relatively small helium refrigerator and an internal heat exchanger. In theory, LOX can be subcooled to near 70K in real time by using a mixture of helium gas and liquid nitrogen. The efficiency of this concept is a function of the mixture heat exchanger and recovery of the helium flow.
• In the 1990's, mechanical cryocoolers were partially replaced by pulse tube coolers which could reach temperatures below 4K. More recently, using a pulse tube cryocooler for the "warm" reservoir for an ADR cooler to reach scientifically significant temperatures below 0.10K. This combination is available as a standard product offered at the 2019 CEC.
• Introduction of MLI was a revolution in cryogenic insulation. Many papers describing test configurations and test results were presented at the CEC over a 35-year period. About 1990, Dr. McIntosh took an analytical approach to MLI which allowed layer by layer density possible. Newly available personal computers and software made calculations of 40 to 50 simultaneous equations possible in a regular office setting. With NASA operating and test support, it was found that variable density MLI could be calculated accurately. The variable density MLI concept is covered by U.S. Patent #5,590,054 (12-31-1996)

10. Fundamental Cryogenic Guidelines
• Aluminum to stainless steel transition joints usually fail.
• Indium sealed joints are reliable especially when fit with Invar washers.
• Indium seals should never be augmented with vacuum grease because the grease will freeze and crack.
• Barstock should not be machined to form a cold barrier between atmospheric pressure or higher on one side and vacuum on the other. These pieces can be absolutely tight when warm and leak profusely when cold.
• 303 stainless steel is not suitable for cryogenics.
• Only oxygen free copper tube or pipe should be welded. Other copper welds crack immediately when cold shocked.
• Piping runs from cold to warm must always slope upward at least one diameter. Level or downward sloping runs must be treated as thermal shorts.
• Pressure build-up calculations can be tricky. Thermodynamics should be based on internal energy, not increase in enthalpy. Think about P * dv with dv = 0.

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