CRYOGENICS FOR SUPERCONDUCTORS: REFRIGERATION, DELIVERY, AND PRESERVATION OF THE COLD

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ABSTRACT

Applications in superconductivity have become widespread, enabled by advancements in cryogenic engineering. In this paper, the history of cryogenic refrigeration, its delivery, its preservation and the important scientific and engineering advancements in these areas in the last 100 years will be reviewed, beginning with small laboratory dewars to very large scale systems. The key technological advancements in these areas that enabled the development of superconducting applications at temperatures from 4 to 77 K are identified. Included are advancements in the components used up to the present state-of-the-art in refrigeration systems design. Viewpoints as both an equipment supplier and the end-user with regard to the equipment design and operations will be presented. Some of the present and future challenges in these areas will be outlined. Most of the materials in this paper are a collection of the historical materials applicable to these areas of interest.

KEYWORDS: helium, superconductivity, refrigerator, cryogenic equipment, thermal insulation, cryogen storage

INTRODUCTION

Discovery of Helium, its Liquefaction and the Reserves

The roots to the discovery of superconductivity originate in the discovery of helium and its liquefaction. The existence of the new element (helium) in Sun came to light while the French astronomer Jules Janssen was observing the solar eclipse (a bright yellow line in the spectra) of August 18, 1868 at Guntur in the state of Andhra Pradesh, India [1, 2]. Lockyer and Frankland confirmed Janssen's results and named the new element as "helium" (after "Helios" – Sun) in October 20, 1868. In 1892 Luigi Palmieri detected the existence of helium on Earth and in 1895 Sir William Ramsay isolated helium on Earth [1].

Helium was first liquefied by Kamerlingh Onnes on July 10, 1908 in Leiden, Holland [3, 4]. The initial attempts to liquefy helium and to confirm the results were extremely difficult, as the apparatus used was very complicated. They used a cascade process with liquid air and liquid hydrogen for pre-cooling the compressed helium followed by a Joule-Thompson expansion process for the final liquid production [4, 5].

In 1905, large reserves of helium were found in the natural gas fields at Dexter, Kansas by Cady and McFarland of the University of Kansas at Lawrence [6]. Additional large sources were found in other natural gas fields of the United States. The special properties of helium for critical applications, particularly for defense, were recognized during World War I by the US Government who subsequently enacted the Helium Conservation Act in 1925. This has since been replaced by the Helium Privatization Act of 1996. Although the US holds 21% of the world's estimated helium reserves, presently it produces 77% of the world's helium [7]. The remaining 79% of the world's reserves remain mostly within untapped large sources in Russia [8], as well as in Algeria and Qatar, who provide helium to the European and Asian markets, respectively. With the US exports substantially rising and US consumption falling (though still the highest in the world at about 45% [9]), it has been estimated that the US helium-rich sources will be depleted in less than 30 years [10]. Global demand for helium is projected to grow from the present 175 to 300 million cubic meters per year by 2030 [8].

DISCOVERY OF SUPERCONDUCTIVITY

After the production of liquid helium, Kamerlingh Onnes conducted experiments to determine the effect of low temperature on the electrical resistance of pure metals. As the temperature of the material approached absolute zero, he wanted to see if one of three results would occur; whether the electrical resistance would (1) reduce to zero, or (2) to a minimum and stay constant, or (3) to a minimum and then increase to a maximum (behaving like an insulator). ***Experiments conducted under Onnes direction by Gilles Holst with mercury resulted in the discovery of superconductivity in 1911, 100 years ago*** [4]. This significant discovery has lead to many other technological developments and associated applications.

In addition to a critical temperature, above which the superconductor will revert from a superconducting to a normal electrical state, superconductors also exhibit a critical magnetic field, above which the same occurs. Superconductors that exhibit a pure Meissner Effect, completely excluding externally applied magnetic fields from its interior, are known as Type I superconductors. These are all pure metals and have transition temperatures ranging up to approximately 9.5 K. Their behavior is described by the phenomenological Ginzburg-Landu theory in 1950 [11] which explained the macroscopic properties of superconductors. This theory has been largely superseded by BCS Theory in 1957 [12, 13] which is a complete microscopic theory of superconductivity and is named after its originators John Bardeen, Leon Cooper, and Robert Schrieffer. In the 1930's the first Type II superconductors were discovered, though not recognized until after the Meissner Effect was discovered in 1933 by Walther Meissner and Robert Ochsenfeld. Type II superconductors have much higher critical magnetic fields and are distinguished from Type I superconductors by exhibiting a mixed state, also called a vortex state, where the external magnetic field is only partially excluded; so that part of the material is superconducting and part is normal. In 1986, high temperature superconductors (HTS) were discovered by Müller and Bednorz, having created a brittle ceramic compound of lanthanum, barium, copper and oxygen that had a critical temperature of 30 K. A year later, teams at the University of Houston and at the University of Alabama-Huntsville replaced the lanthanum with yttrium achieving a critical temperature of 92 K. Many milestones have been since reached. These are ceramic based compounds and are also classified as Type II superconductors with critical temperatures that are above the normal boiling point of liquid nitrogen [14].

Superconducting technology not only brought some new technologies to light which would otherwise not be possible, it also reduced the cost and improved the efficiency, quality, size and weight of other technologies, especially in the area of scientific accelerator technologies.

Applications of Superconductivity

Among the many applications which utilize superconducting technology, a few are listed below:

- Detectors: Josephson junctions, Superconducting Quantum Interference Devices (SQUIDS), fast digital circuits
- Science: Particle accelerators using superconducting magnets and/or SRF cavities, fusion research
- Biomedical applications: Magnetic Resonance Imaging (MRI), Cryosurgery, SQUID devices, EEG, Magneto Encephalography (MEG) to study the neural activity in brain, Magneto Cardiography (MCG)
- Transportation: MagLev (super-conducting magnetic levitation – used for mass transit rail or orbital insertion of payloads)
- Military: Superconducting electric motors for ship propulsion, microwave radar
- Energy: SMES (for energy storage, power quality, pulse power, etc.), low loss power transmission systems
- Other: Cryogenically cooled super computers, communication systems, ore separation, SQUID based Non Destructive Testing (NDT)

Many of these low-temperature superconducting (LTS) and some HTS applications require helium refrigerators in order to operate. The cold must be supplied and continuously maintained by cryogenic fluid or by a cryogenic refrigerator, or a combination of both. To produce and maintain any temperature significantly below ambient is a challenge, but to keep temperatures within a few kelvin of absolute zero is a great challenge that continues today, 100 years after the discovery of superconductivity. In essence, superconductivity is dependent on cryogenics at the present time. The key cryogenic engineering elements include refrigeration, thermal insulation, and delivery of the cold to the heat load. Efficiency, practicality and simplicity are the keys to success in all these elements.

PRODUCTION OF THE COLD: REFRIGERATION

Advancements of Helium Liquefaction

Significant advancements were made by many notable scientists in the cryogenic refrigeration processes that are required for the production and liquefaction of industrial gases, such as air and hydrogen, prior to attempts to liquefy helium [4]. Although helium was first liquefied by Onnes, the initial liquefaction concepts were very difficult and unreliable. The systems developed by Kapitza (1934) used a single expander Claude cycle (note: the Claude cycle was initially developed to liquefy air) with liquid nitrogen as pre-cooling, followed by the JT process to produce liquid helium. Kapitza was the first to apply adiabatic expansion (the Claude approach) to drop the temperature of helium below the inversion point [4].

The National Bureau of Standards (NBS) was established in 1901 and the cryogenics program shortly thereafter, with the funds appropriated in 1904 for the purchase of a two liter per hour (2 L/hr) hydrogen liquefier [15]. The laboratory was originally in Washington, DC and later moved to Boulder, CO in the early 1950's [15]. It has been a leader in the cryogenic research for many years and it was here that the properties of helium were measured, verified, standardized and published for use across the globe.

In 1946, Collins and his colleagues at MIT built a simpler version of a helium liquefier which produced liquid helium without the need for pre-cooling (although liquid nitrogen pre-cooling is used to increase the capacity of the machine) [16]. Most of the previous helium liquefiers used hydrogen pre-cooling to produce liquid helium. Collins' machine made the precious liquid helium available to the scientific masses, previously accessible to only a privileged few, and without the dangers associated with hydrogen. Initially, oil free compressors were used for helium liquefiers due to the fear of oil contamination to the process. However these compressors were unreliable and required frequent maintenance. Collins recognized this weakness and in the 1960's introduced the oil lubricated reciprocating compressors for longevity. After this he made a bold statement [17] (for that time) to his colleagues that, 'We should take advantage of the oil to lubricate and improve the reliability (by keeping the compression process cooler), since whether we use one drop or flood it, the oil removal needs to happen before entering the cold box." This recognition led to the introduction of oil-flooded screw compressors for the helium process in the 1970's. The Collins liquefier was commercialized by A.D. Little, and is known as the 1400-series liquefier. To date there have been more than five hundred 1400 style Collins helium liquefiers operating around the world supporting the low-temperature experiments and applications, thus making it possible for many low-temperature scientists to explore their imaginations.

Present Status of Helium Refrigerators

The overall exergetic efficiency (also referred to as the efficiency as compared to Carnot) of these Collins-type liquefier systems was in the 10% range. However, beginning in the 1970s, the capacity demand for helium liquefiers with improved efficiency and reliability grew exponentially. This led to the development and application of dependable turbo expanders, plate fin heat exchangers, oil-flooded screw compressors and many other components now commonly used for these machines. At present, for refrigerators of greater than 5 kW at 4.5 K, efficiencies are to the 30% level at the design loads with an availability of greater than 99% due to these developments which have improved the component efficiencies and process cycles.

Traditional cryogenic refrigeration and liquefaction process cycles are based on a modified Claude-Brayton cycle (i.e., constant-pressure process) or the Stirling cycle (i.e., constant-volume process) and are typically designed for a specified maximum capacity operating point(s) with a maximum overall exergetic efficiency at that design point.

Capacity margin is normally added to the system design to cover the uncertainty in the estimation of loads, the requirements for high peak low average load operations, short duration peak loads like cool down, uncertainty in the performance prediction of critical components, as well as the inevitable occurrence of underperforming components in the system. This margin, as well as the occasional need to use (adapt) equipment originally designed for some other application results in mismatch between the actual load and the system capacity, resulting in operation of the system at off-design conditions and at substantially lower efficiency in the field.

However, today we are able to design and operate these systems over a wide range of capacities and operating conditions at maximum efficiency using the constant-pressure ratio Floating Pressure Cycle also known as the Ganni Cycle [18]. Two plants presently in production, which are scheduled to be commissioned in the near future, are based on this design approach. These are the 20 K refrigerator for the James Webb Space Telescope testing at NASA Johnson Space Center (JSC) and the CHL-II for the 12 GeV upgrade at JLab. In addition, there are many operating plants at JLab, MSU [19], BNL [20], SNS [21] and NASA [22] that have been converted to this operating philosophy to varying degrees and have demonstrated substantial improvement in efficiency over an increased operating range and with higher availability. As achieved in these previously mentioned converted plants, the CHL-II is designed to operate from 100% to less than 40% of the maximum exergetic capacity without any significant loss in overall exergetic efficiency at *many modes* [23]. Similarly, the NASA-JSC refrigerator system has been designed to handle loads from 15 K to 100 K with a capacity variation from 5 to 100 kW for different modes, without any significant loss in overall exergetic efficiency [24]. This process and method of design minimizes the effect of uncertainties in the load planning and actual component performances which allow the plant to a large extent operate automatically at the maximum efficiency for the capacity needed by the load.

In present helium refrigeration plants approximately 50% [25] of the input power is lost due to warm-compressor inefficiencies. As such, it is clear that the overall system efficiency improvements strongly depend on the compression process improvements. Some work is underway [26] to understand these various loss mechanisms in the warm compression systems.

Cryocoolers

Many applications require only a small amount of refrigeration capacity. The traditional helium refrigerators based on Claude, Brayton and Stirling cycles are often too bulky, complicated and/or expensive. The need to reduce the size, complexity and cost for small loads led to the development of Gifford-McMahon (GM) cycles and J-T cycles. GM refrigerators were initially developed at A. D. Little in Cambridge, MA (as were the Collins' helium liquefiers). These types of refrigerators, in addition to the development of Stirling and pulse tube refrigerators, play a major role in the area of small capacity machines (e.g., [27]). The coupling of cryocooler, heat exchanger, and storage tank technologies has led to the ability to store cryogens nearly indefinitely using "zero boil-off" systems. Efficiency, reliability and vibration still remain to be challenges.

High Temperature Superconductors (HTS)

Although the advancements are taking place in the design of 4 K and 2 K helium refrigeration systems, the reversible (also known as the minimum ideal, or 'Carnot') input power required for each watt of cooling is too high for economical low temperature superconductor applications. Because the reversible input power required for cooling falls rapidly as the temperature increases towards ambient, there is substantial interest in the continued development of HTS. In addition, the material properties like specific heat and thermal conductivity (to support heat transfer) are generally higher at warmer temperatures and are more favorable for the design of the refrigeration equipment. The potential of HTS is estimated to be substantial, especially for electric power applications. There are several ongoing HTS cable demonstration projects world-wide (e.g., [28]). Also prototypes of other HTS power applications such as transformers [29] and fault current limiters are being developed. Similarly technology development for other applications like motors and generators is also progressing. There is also progress in HTS applications for Josephson devices and mobile telecommunications. However, there remains substantial research needed in this area to make these applications economically competitive.

Despite the progress in HTS, superconducting RF cavities and superconducting magnets used for many particle accelerators, as well as, superconducting magnets used for very high field designs, like the 45 Tesla hybrid magnets at FSU (which use both the normal and superconducting magnets [30]) require low temperature superconductors. Although low temperature superconductors impose substantial operating costs and penalties associated with their complexity, they do enjoy a niche for such specialized applications.

Experience Differences: Supplier versus User

It is important to bring out the guiding interests of the equipment supplier and the user in the decision making process. As an equipment supplier the primary interest is to understand the user's needs from their viewpoint. For suppliers, it is not wise to go beyond the basic understanding of the need, since questioning the customer can lead to friction and even cause one to lose the job. Without an end-user need, no refrigerator would be required to be produced; so their need should be carefully considered from all viewpoints. Equipment suppliers aim to design the system utilizing the recognized guiding principles for the location of the critical components, such expansion step placement according to the Carnot step theory and other similar methods [16, 31, 32, 33].

As a user who operates the actual system under innumerable operating conditions, and having an understanding of the actual performance potential of the components in the refrigeration system, one gains much insight on how the fundamental principles of thermodynamics and other sciences can be used more precisely to fulfill the end needs. Such insight led to the development of the constant pressure ratio floating pressure – Ganni Cycle concept [18] to address variations in the required capacity, various modes of operation and actual equipment performance in such a manner that the refrigeration system automatically operates close to its maximum overall exergetic efficiency. This method is also applicable for the common situations of load/capacity mismatch, where the loads are somewhat different from the original estimates, and when using equipment originally designed for an entirely different application.

Other similar insights as a user include, moving the 4-K to 2-K heat exchanger from the central 2-K cold box [34] to the individual cryomodules at SNS [35, 36, 37]. This approach reduced the exergetic load on the refrigerator by moving the distribution system heat in-leak from 2-K to 4-K, decreasing its exergetic effect to less than half and making the system more efficient and stable. Likewise, other insights include modifications to JLab's 2-K cold box (as was provided by industry) to allow it to operate and function in an efficient and robust manner [34, 38], and optimizing the 4 to 2-K pressure profile and cooling curves [39], as well as others [40]. It is understood that some of these optimizations could have been missed because of procurement boundaries, or due to the current understanding of the art, or due to a focus on delivering what the user asked for in a timely manner. These realities point to the need for a close collaboration between the user(s) and suppliers in a manner that allows both to be mutually successful. But no matter how close the user and supplier collaborate in developing the system, there are always unanticipated issues associated with the actual equipment, load(s) being supported, or ancillary systems. Therefore, it is vitally important to gain the experience and understanding of the strengths and limitations of both parties for the progress of the technology.

DELIVERY OF REFRIGERATION

For large systems, the liquid helium required to support the refrigeration or liquefaction load at the final end use is usually supplied through vacuum-jacketed transfer lines with multi-layer insulation (MLI) inside the annular space. The vacuum and MLI combination provides roughly three orders of magnitude lower heat flux compared to conventional thermal insulation systems such as perlite powder, polymeric foams, or fiberglass operating under ambient pressure conditions.

The challenge of providing vacuum jacketing with carefully executed MLI at every point along the distribution system is a main driver of the mechanical design of the overall system. These transfer lines come in many shapes, sizes and forms including small coaxial lines to very large and complex transfer-line distribution systems used at locations such as CERN, RHIC, FERMI Lab and CEBAF. Design aspects and considerations for these complex transfer lines are outlined in many publications (e.g., [41]) including a detailed description of the CEBAF transfer-line designs [42], which are as shown in FIGURE 1. The transfer-line designs at JLab and SNS are based on original FERMI Lab concepts. The interface connections from the refrigerator and loads to the transfer lines are accomplished in many ways. They include permanent connections with isolation valves, joints (sometimes called 'field joints') that involve both the vacuum jacket and process line [43] or couplings (also referred to as 'bayonets'). Some of these bayonet-couplings are designed for easy disconnection at positive pressures and at cryogenic temperatures [42]. Others bayonets-coupling designs can only be disconnected at ambient temperature. In general it is found that the majority of the distribution-system heat input is contributed by components like valves and bayonet-couplings and to a lesser extent by 'straight line' portions of the transfer-line. Although relatively unpublished, it is not uncommon for the heat leak of the distribution system to be underestimated, sometimes significantly. To reduce the effect of heat leak the cold-temperature lines are shielded by lines carrying higher temperature fluid and the valves and bayonets are heat stationed.

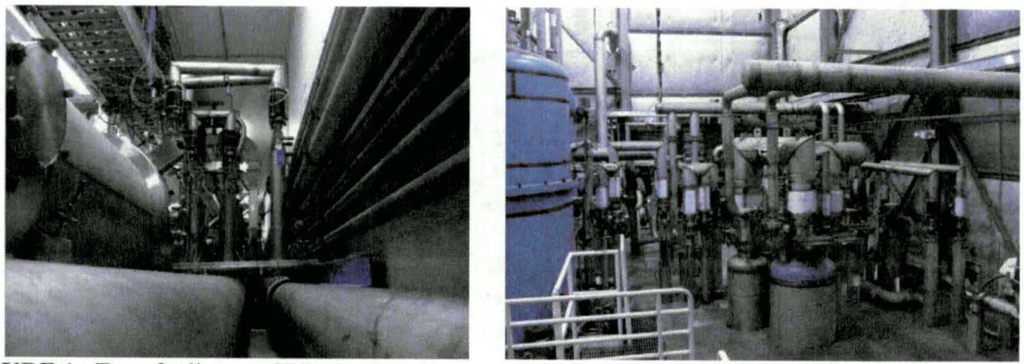

FIGURE 1. Transfer lines at CEBAF connecting the cryo modules to the refrigerator.

Long transfer lines can pose cool down and stability issues on the operations (especially if the vacuums degrade). As MLI requires high vacuum (i.e., below about 10^{-5} mbar) to operate properly, vacuum degradation can be a serious issue for all elements of the distribution system. The technology of bake-out processes, getters, adsorbents, evacuation ports, safety relief devices, and vacuum pumping has been successfully developed in the last 50 years or so to protect against most vacuum failures. However, long-life and high-reliability high-vacuum systems remain a challenge.

PRESERVATION OF THE COLD: THERMAL INSULATION

Cryogenic insulation system development began during the period 1877 to 1908, coinciding with the first liquefaction of key industrial gases. D'Arsonval first demonstrated the vacuum flask in 1887 [44]. This design was significantly improved by Dewar in 1893 by silvering the walls of the flask. The concept of filling the vacuum space with powder was illustrated by Stanley in 1912 [45]. This work led to the development of vacuum flasks (dewars) for the preservation of cryogens, milk, hot or cold beverages, etc. and later for the preservation of biological matter for a wide range of farming and medical purposes.

From the discovery of superconductivity in 1911 to its widespread proliferation by 1961, no significant advancements in superconducting applications were made during this nearly 50-year period. The lull can be attributed in part due to refrigeration processes that were difficult and inefficient, as well as, the lack of high-efficiency insulated storage vessels. With the advent of the Cold War after World War II, especially the beginning of the US space program in the 1960's, there was an explosion in activity with the development of hydrogen and other liquefiers, advancements in high performance thermal insulation systems and new cryogenic tank and vacuum piping designs [46].

An early vacuum-jacketed cryogenic tank insulation system design of multiple radiation shields, shown in FIGURE 2, was advanced by Cornell in 1947 [47]. Multilayer insulation (MLI), which could provide at least an order of magnitude of improvement in the performance as compared to evacuated perlite, was first demonstrated by Peterson in 1951 [48]. MLI systems were well developed by about 1960 through the work of Matsch, Kropschot, Hnilicka, and others [49, 50, 51]. Later innovations improving the performance of dewars included vapor shield cooling, neck tube cooling and low heat leak supports [52].

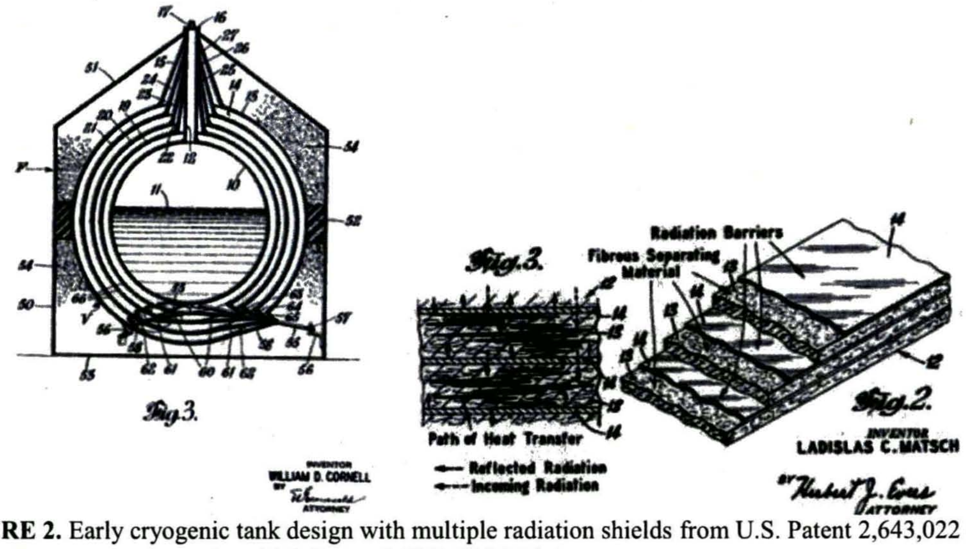

FIGURE 2. Early cryogenic tank design with multiple radiation shields from U.S. Patent 2,643,022 (left) and later MLI system concept from U.S. Patent 3,007, 596 (right).

Of course, there were other supporting technologies that enabled thermal insulation systems for superconducting applications, such as the process of welding of stainless steel (e.g., [53]) and the accidental invention of Teflon [54]. Key innovations in welding were brought about through World War II [55]. Scott's original textbook on cryogenic engineering devotes many pages on "insulation" and most of these involve the discussion of obtaining high vacuum including the use of Teflon; however MLI is never mentioned [56]. These technologies culminated in the production of large size vacuum-jacketed tanks (~50,000 liters liquid nitrogen or liquid hydrogen), high vacuum levels with longer retention life and low heat flux insulation systems (i.e., 10 to 100 times lower than evacuated perlite).

Although the majority of the work after WWII was focused on liquid hydrogen production and storage, the areas of superconductivity and liquid helium were direct beneficiaries. It is important to note that the Collins liquefier was invented and developed during the same timeframe. This simpler refrigeration technology together with the new high efficiency cryogen storage technology enabled the explosion in liquid helium experimentation and research in superconducting applications. Worldwide, the number of labs with liquid helium capability grew from only 15 in 1946 to more than 200 by 1960 [57].

The cryogenic tank technology has improved over the years from storing a cryogen for a few hours to many months. The storage life of cryogens extends to years for space probes thus demonstrating the technology has made improvements by many orders of magnitude. FIGURE 3, illustrates the effect of a given heat flux on the boil-off for various fluids. Originally, the dewars for liquid helium were very small and limited to a few liters. Today tanks are built to large sizes and are mostly limited by shipping restrictions. The largest shipped commercial size is approximately 110,000 liters and there are quite a few of this size currently in operation.

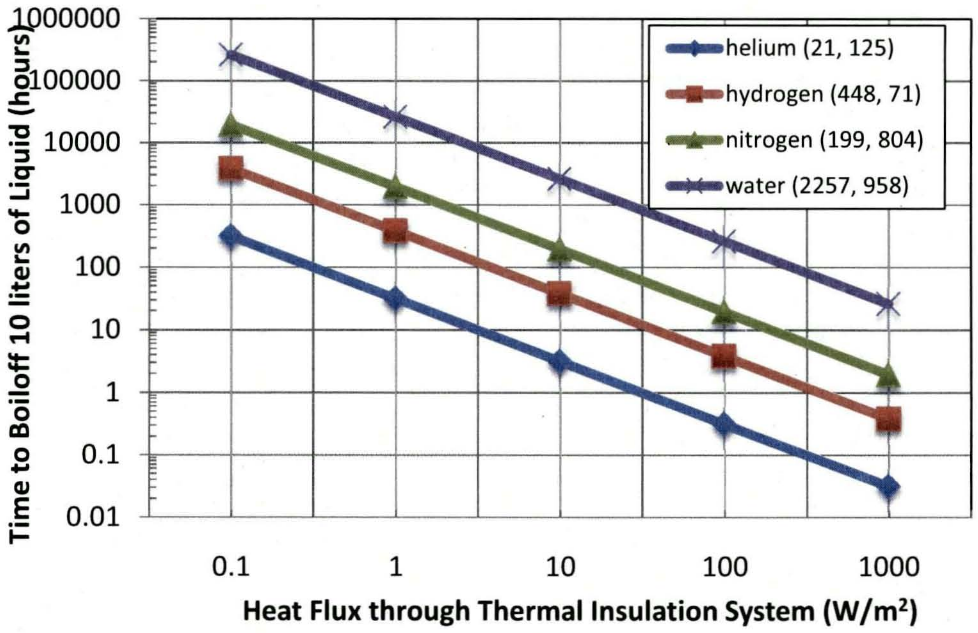

FIGURE 3. Boiloff time for 10 liters of different liquids over a wide range of heat flux values (heat of vaporization in kJ/kg and density in kg/m^3 given in parentheses).

Increasing thermal insulation performance requires optimization of total system heat leakage rate through better thermal data and analysis tools (e.g., [58]), as well as, the implementation of new materials and composites. These may include robust and load-supporting MLI-based systems [59], aerogel blankets [60], aerogel/polymers composites (structures) [61], polyimide foams (structures), glass bubbles [62], and aerogel particles for bulk-fill (terminations & cold boxes) [63]. Developments in improved getters for long-life vacuum and internal vacuum sensors are also needed as vacuum remains a key underlying issue for the operation of very low heat leak systems.

CONCLUSION

The saying that 'necessity is the mother of invention' certainly can be true, and so can its corollary that 'finding applications to the invention is the mother of creation for necessity.' This is akin to the philosophical quandary of 'which came first, the chicken or the egg?' Onnes wrote in Paper 15 of his numerous communications: "The fact that a pure metal can be brought to such condition that its electrical resistance becomes zero is certainly of itself of highest importance [64]." This experimental work was a significant discovery and led to many other developments of applications using this principle of the *disappearance of electrical resistivity.*

For example, the innovations in computers and communications technologies seem to be self-sustaining, doubling the capabilities every eighteen months. However, in the cryogenic refrigeration field, efficiency improvements and miniaturization are comparatively almost listless, thus limiting the advancements to increase potential applications. We must follow the Collins philosophy of observing and learning from nature, apply the known scientific and engineering principles and advance the developments in the support components and processes that improve the efficiency, reliability and simplicity of these systems. By advancing this technology to improve its performance, we can enable the creation of new applications.

In the early 1980s, the innovation of MRI (NMR) came as one of the major superconducting magnet applications. This breakthrough is an example of an exciting innovation for the cryogenic technologies, but was short-lived with no similar applications following it. If cryogenic refrigerators are reduced in size (noting that cryocoolers are making some progress [27]), complexity and cost but improved in efficiency, this could lead to many other innovations; perhaps sensor development and others. Detectors used in physics research today are continuously helping us to understand the nature of this world and such sensors have great potential in many fields such as astrophysics, manufacturing, transportation, medicine, law enforcement and security.

The future use of low-temperature superconductivity applications is strongly tied to efficiency improvements and the reduction in size, complexity and cost of helium refrigeration systems. Likewise, HTS applications strongly depend on refrigerators also and both need these improvements and innovations. Overall, much knowledge has been obtained by pushing on toward absolute zero over the last 100 years.

ACKNOWLEDGEMENTS

The authors would like to express their appreciation and thanks to the TJNAF management for their support. This work was supported by the U.S. Department of Energy under contract no. DE-AC05-06OR23177.

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