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The future with cryogenic fluid dynamics

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This paper is an edited text of an invited plenary presentation at ICEC25/ICMC2014 by Professor Scurlock on the occasion of his being presented with the ICEC Mendelssohn Award for his many contributions to Cryogenics.

As long ago as 1992, he first proposed in his "History and Origins of Cryogenics" that the temperature range for Cryogenics should be extended up to the ice-point at 273K. This paper expands on this proposal with the implicit assumption that Cryogenic Fluid Dynamics can provide a universal basis for modelling heat transfer and convective fluid behaviour of all fluids, at all temperatures, below the ice-point at 273K; or below 250K if you wish to exclude refrigeration engineering."

Abstract

The applications of cryogenic systems have expanded over the past 50 years into many areas of our lives. During this time, the impact of the common features of Cryogenic Fluid Dynamics, CryoFD, on the economic design of these cryogenic systems, has grown out of a long series of experimental studies carried out by teams of postgraduate students at Southampton University. These studies have sought to understand the heat transfer and convective behavior of cryogenic liquids and vapors, but they have only skimmed over the many findings made, on the strong convective motions of fluids at low temperatures. The convection takes place in temperature gradients up to 10,000 K per meter, and density gradients of 1000% per meter and more, with rapid temperature and spatially dependent changes in physical properties like viscosity and surface tension, making software development and empirical correlations almost impossible to achieve. These temperature and density gradients are far larger than those met in other convecting systems at ambient temperatures, and there is little similarity. The paper will discuss the likely impact of CryoFD on future cryogenic systems, and hopefully inspire further research to support and expand the use of existing findings, and to improve the economy of present-day systems even more effectively. Particular examples to be mentioned include the following. Doubling the cooling power of cryo-coolers by a simple use of CryoFD. Reducing the boil-off rate of liquid helium stored at the South Pole, such that liquid helium availability is now all-the-year-round. Helping to develop the 15 kA

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current leads for the LHC superconducting magnets at CERN, with much reduced refrigeration loads. Improving the heat transfer capability of boiling heat transfer surfaces by 10 to 100 fold.

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1. The origins of cryogenic fluid Dynamics, CryoFD

Cryogenic Fluid Dynamics, CryoFD, is the technology, based on a long series of experimental studies, whereby the extraordinary fluid properties of cryogenic liquids and vapours at low temperatures may be used collectively and advantageously to provide efficient low loss cryogenic systems. The future of cryogenic applications is going to be determined by rising energy costs and increasing shortage and cost of helium gas, which can be met by increasing the use of CryoFD in the design and operation of cryogenic systems.

The dynamics at decreasing temperatures of natural convective motion, and associated heat transfer and mass flow, are strongly enhanced by the large changes in basic properties such as increasing density and decreasing viscosity. As a result, the natural convective fluid dynamics of vapours and liquids, both single and multi-component in composition, are not the same as fluids at ambient temperatures, being driven by powerful density gradients up to 1000%/m and local temperature gradients up to 10,000 K/m.

This behaviour needs to be recognised and incorporated into the design culture for cryogenic systems, requiring the lowest refrigeration electric power and miniscule boil-off rates as standard practice.

The first advances in CryoFD were carried out by Sir James Dewar at the Royal Institution, London in the 1880's. Dewar realised that he could not study the low temperature properties of gases and liquids without suitable insulated containers. He therefore spent his first 15 years at the R.I. in developing a suitable design. He used glass, with its low thermal conductivity, and a double-walled vessel to employ vacuum insulation. This was not good enough however, even for the small quantities of liquid air he was making. He spent many years putting various fillers in the vacuum space, even several layers of aluminised cigarette paper, but no progress was being made.

Then, by chance, he noticed that mercury droplets from his mercury vacuum pump, in the vacuum space of his double walled container, evaporated and condensed on the inner glass wall to make a shiny film when liquid air was placed inside. He noted that as the film grew with time, the evaporation rate of the liquid air diminished rapidly. He soon found that introducing a silver coating on the inner walls was more effective and reliable; and the double walled, silvered glass, vacuum vessel, the dewar, was born.

Dewar's dewar was an excellent application of early CryoFD, and enabled him to liquefy hydrogen in 1898; and Kamerlingh Onnes to liquefy helium in his Cryogenic Laboratory in Leiden, in 1908. This piece of technology was then the standard cryogenic liquid vessel for the next 70 years until the 1950's, and no further development took place in this time period.

The first systematic research on the low temperature fluid behaviour was started in 1952 by Russell Scott and his early team of research staff at the newly formed National Bureau of Standards Cryogenic Laboratories, Boulder, Colorado, USA, with the initial aim of developing liquid hydrogen technology. This included research firstly into the containment of liquid hydrogen in portable transport vessels, and secondly on improving the available insulations.

In 1954, NBS expanded its research into declassified industrial cryogenic applications, with industrial funding to replace the reduction in federal government funding. Thereafter it became the global research centre for liquid hydrogen, oxygen and helium applications, notably in supporting the growing requirements of NASA's space rocket programme.

In 1959, Scott published his book "Cryogenic Engineering" (Scott, 1959), as a record of the research findings and thinking that he and his colleagues had achieved in cryogenics, in the first five years of unclassified operation at the NBS. In his chapter on 'Storing and Transporting Liquefied Gases', he considered the concept of the vapour-cooled vent tube. He observed that the cold available in a cryogenic vapour for absorbing heat inflows can be many times

larger than the latent heat of evaporation of the cryogenic liquid. He further stated that “in those cases in which heat conduction through the supports or vent tubes is a substantial part of the total, it may be worthwhile to use the escaping vapour to intercept part of the heat”. He was indeed sowing the seeds for developing CryoFD as a major support for cryogenic technology.

His words provided the inspiration for the expansion of CryoFD, in parallel with the work at NBS, at the newly formed post-graduate training centre in Cryogenics and its Applications, and the subsequent Institute of Cryogenics, at Southampton University, UK, for the next 50 years.

2. Summary of CryoFD Findings

The research findings at Southampton (Scurlock, 2006) include the following, some of which will be introduced in particular examples later in this paper.

2.1. Under isobaric storage and use, efficient containers have heat in-fluxes (W/m^2) to the stored liquid which are well below the minimum heat flux required for nucleate boiling (by a factor of 10 – 100 times too small.). This means that evaporation to absorb the total heat influx is by surface evaporation only – there is no nucleate boiling. However, the term ‘boil-off’ is commonly used to mean the evaporation loss, and we shall continue to do so.

2.2. Surface evaporation can only take place from superheated liquid in a surface layer. The evaporation mechanism, whereby the heat is absorbed by the latent heat of liquid molecules leaving the surface, is contained in a surface layer a few mm, only, in thickness. The heat energy is transmitted by micro-convection cells and then by thermal conduction across a 1 – 2 μm thick layer to a thin surface layer controlling the flux of molecules evaporating from liquid to vapour phase. The details of this mechanism are not fully understood because the molecular evaporation flux is not uniform spatially across the surface, and varies rapidly with time with observed variations in magnitude up to 10% of the average boil-off rate.

We have also found that the surface evaporation mechanism is sensitive to impurities in the surface and to mechanical disturbance of the surface, leading to boil-off instabilities and vapour explosions.

2.3. The available “cold” in the vapour, i.e. the enthalpy increase between boiling point and ambient temperature at the boil-off pressure, is much larger than the latent heat of evaporation of the liquid, for helium (x 75), and hydrogen (x 7.9); and the same magnitude for oxygen (x 0.9), nitrogen (x 1.2), and methane (x 0.8). Heat inflows into the stored liquid can be divided into ‘A’ heat inflows, absorbed only by the latent heat of the evaporation loss, and ‘B’ heat inflows which can be absorbed by the available cold in the boil-off vapour. Thus, to design for minimum boil-off, all the various A heat inflows into the liquid need to be identified and reduced to zero as far as possible, or converted into B heat inflows.

2.4. Heat transfer between cold vapour and neck wall is extremely good. Moreover, heat transfer from a heater element mounted in the cold vapour is shared across the whole vapour column by a spontaneous process of horizontal convection, via the density stratification in the vapour. Measurements have indicated that the effective heat transfer coefficient is enhanced by up to 10 fold.

Thus, it has been found that horizontal vapour cooled radiation shields in the neck can absorb infra-red radiation heat flows down the neck very efficiently. With optimum positioning, they can totally absorb the radiation heat flow, with almost zero radiation heat flow into the liquid.

2.5. In the vapour space above the liquid, there is closed loop, thermo-syphon type vapour recirculation, with a finite, recirculating, mass flow $R(\text{vapour})$ kg/sec which may well exceed the boil-off mass flow. The recirculating flow mixes with the upward boundary layer flow adjacent to the neck wall, with a total mass flow of R plus the boil-off mass flow, while the centre of the vapour column moves downwards with a reducing mass flow. The recirculation is driven by conducted heat flow absorbed from the neck wall by the boundary layer flow. The A heat inflow from the central, downward vapour plume appears to provide the residual value for minimum boil-off

performance. Further research is needed to understand how to reduce the recirculating vapour mass flow at the liquid surface, and the associated residual heat inflow.

2.6. In the liquid volume, there is also closed loop, thermo-syphon type recirculation with a different total mass flow $R(\text{liquid})$ kg/s. Again, the recirculation consists of an upward boundary layer flow adjacent to the neck wall, with a total mass flow of $R(\text{liquid})$ plus the boil-off mass flow, while the centre of the liquid has a total downward mass flow R concentrated into a central jet. Evaporation of the liquid only takes place at the surface as the boundary layer flow turns over and flows radially inwards along the surface to the central downward jet. Further studies on large scale storage systems are needed to measure the scale of the recirculations and the associated build-up of stored thermal energy in the liquid.

2.7. Vapour cooling of the neck wall happens very efficiently via the boundary layer flow at the neck wall. Scott's analysis of the vapour cooled vent tube can be turned round to give the minimum length of neck required for a given boil-off mass flow, when the conducted neck heat flow is reduced to zero by the Scott vapour cooling effect. Any longer neck will result in over cooling of the neck wall and a waste of the available vapour cooling.

2.8. In addition to cooling the neck tube, the excellent heat transfer capability and cooling capacity of the vapour can be used to absorb many of the heat influxes as B fluxes, which would otherwise enter the liquid as A heat fluxes. This includes the cooling of multi-shields in the insulation surrounding the liquid.

2.9. In general, A heat fluxes are absorbed by the boundary layer flow at the wall of the liquid containment, thereby superheating the boundary layer and providing the buoyancy from the density difference to drive the natural convective flow at the wall and the liquid recirculation. The superheated liquid is carried up to the surface where it turns over and streams radially inwards along the surface, during which time evaporation takes place to provide the boil-off mass flow. At the centre, the radial inflow joins the downward jet flow, evaporation ceases, and any remaining superheat is carried into the bulk of the liquid. The point to note is that the surface where the heat is absorbed and carried away by the boil-off vapour is a remote distance from the entry areas of the A heat fluxes.

2.10. The enhanced heat transfer capability by natural convection in a vapour column in a neck tube cannot be improved by inserting vertical tubes or high conductivity metals into the vapour to change the convective recirculation. This has been tried many times and results in an increase in boil-off on every occasion. For example, trying to force the vapour into closer contact with the neck wall, or with a superconducting current lead, via annular inserts will only result in a significant increase in boil-off.

2.11. Development of multi-layer insulations have enabled a significant improvement in the performance of MLIs to be achieved by incorporating getter materials such as activated carbon in the spacer material, and avoiding all use of plastic material. Using aluminum foil and carbon loaded glass micro-fiber paper, thermal conductivities below $10 \mu\text{W/mK}$ can be obtained by vacuum baking at 200 – 300 degrees C of all the MLI components before assembly. Baking at 100 degrees C will only remove adsorbed water, but not adsorbed gases like hydrogen or helium.

2.12. Cryogenic liquid mixtures have additional properties not experienced by single component liquids. While density stratification in pure liquids is mainly temperature dependent, stratification in a multi-component liquid is both temperature and composition dependent. In isobaric storage, this presents the problem of rollover between two stratified layers with different densities and hence different temperatures and composition. If the density difference is greater than about 1%, the heat influx to the lower layer becomes trapped within the lower layer, the temperature rises and the density decreases with time. Eventually the two densities approach the same value, when an extraordinary self-mixing process commences. All the excess heat trapped in the lower layer, together with the heat of mixing, is released as a vapour spike, 10 to 100 times greater than the normal boil-off rate. This rollover is a problem for the LNG industry to avoid (Scurlock, 2014).

2.13. On the other hand, when a cryogenic liquid mixture is stored isochorically, under pressure at constant volume, so as to obtain zero boil-off loss of the liquid, the accompanying stratification in the pressure vessel appears not to lead to rollover. This arises because the density difference between the two layers remains small, less than 1%, and there is no trapping of heat in the lower layer.

2.14. Because mixing is an irreversible thermodynamic event, the increase in entropy ΔS is accompanied by a large heat of mixing $\Delta Q \sim T \Delta S$. Moreover, because an irreversible process is path dependent, so is the additional heat of mixing also path dependent. Thus mixing two liquids by adding cold to hot will create twice as much vapour flash as that when adding hot to cold.

2.15. Another unexpected property of cryogenic mixtures is that while conventional $T - x$ data is obtained from freely boiling mixtures, the $T - x$ data relevant to storage conditions from surface evaporating mixtures is quite different and varies with the evaporation rate. Thus the methane composition, of vapour evaporating from LNG in storage, is much higher than predicted by published $T - x$ data.

2.16. CryoFD was very fruitful in improving boiling heat transfer in the condenser/reboiler of Air Separation Units. Apart from considerable advances achieved with porous surfaces and to restricting the depth of the LOX to 1m in the reboiler, the discovery of the falling film reboiling surface enabled a 10 to 100 fold increase in boiling heat transfer to be achieved, with incredible simplicity.

3. CryoFD applications

3.1. On 10th July 1962, the space satellite, Telstar 1, was launched into low earth orbit for the first trials of direct trans-Atlantic transmissions of TV signals between the USA and UK. The analogue noise problems were such that a low noise, liquid helium cooled, first stage amplifier was mounted on the receiver aerial at Goonhilly, Cornwall, UK. Because the aerial had to scan through 90 degrees, from vertically upwards through to the horizon, the helium cryostat was mounted so that it tilted at 45 degrees when the aerial pointed to the horizon, and tilted 45 degrees the other way when the aerial pointed vertically upwards. The boil-off from the tilted cryostat was phenomenal, and consumed the total supply of liquid helium for the UK, during the 1962 summer months.

Then, a visit to the GPO Dollis Hill research centre revealed the problem. The cryostat contained a large number of transverse, vapour cooled, copper radiation shields, which, on tilting to 45 degrees to receive signals from Telstar 1 near the horizon, resulted in a thermal short circuit of the Scott vapour cooling effect. Replacement with low thermal conductivity Perspex shields removed the problem, the boil-off dropped rapidly and the UK supply of liquid helium was suddenly available for everyone again. This success was among the first significant and inspirational uses of CryoFD.

3.2. A Cryomech PT410 two stage, pulse tube cryocooler, being used as a helium condenser, was modified by adding several horizontal fins, cooled by vapour around the upper and lower stage regenerator and pulse tubes. The result was an almost double increase in helium liquefaction rate from 12.8 to 21.4 L/day. These modifications by Wang and Scurlock in 2008 (Wang, 2008) were the first attempt to increase the condensing rate of a cryocooler by using CryoFD to increase the use of distributed cooling along the length of the cryocooler; not just the cold tip.

Further work could lead to more improvements in cryocooler performance, without changing the mechanical design or compressor power.

3.3. The thermal efficiency of the 15 kA current leads for the many LHC superconducting magnets at CERN round the 26 km ring was significantly improved. The final Southampton design of current lead provided a heat leak of less than 1 W at 4.2 K, when carrying the full current of 15,000 A. This design greatly reduced the total refrigeration power required, and the associated major running cost, to an acceptable practical level.

3.4. Isochoric storage of LNG in pressure tanks up to 25 bar will enable LNG to be used in road vehicles and other transport as a green fuel. The problem of rollover following stratification in the pressure tanks appears to be

non-existent. The stable buildup of a heated upper layer at zero boil-off is enabled by the small density difference maintained between layers as the pressure and temperature rises.

3.5. The storage dewars for liquid helium which were installed at The Amundsen-Scott South Pole research station can only be refilled once a year. However, the first dewars were found to have a higher boil-off than expected, so that the availability of liquid helium ran out after six months. By changing the neck inserts for each dewar in June 2001 to a CryoFD design, of a simple stack of horizontal vapour cooled radiation shields in place of vertical annular tubing, and retrofitting the new inserts, it proved possible to reduce the total boil-off time to over a year. This enabled all-the-year-round liquid helium cooled experiments to be operated without interruption, long before pulse tube cryocooler/condensers were introduced in 2007.

3.6. Full scale experiments with 2.5 m height heat transfer sections of ASU reboiler-condensers revealed the limitations presented by subcooling of the isothermal boiling liquid. Below a depth of about 1 m, the subcooling produced by the liquid head prevented high heat transfer boiling from taking place. Below this depth, the sections were redundant.

Using a falling film of boiling liquid removed the subcooling effect and enabled a 10 to 100 fold improvement in heat transfer rate per unit area to be achieved.

3.7. The concept of a heat engine employing a cryogenic fluid as a “cold” store is being developed rapidly for energy storage in several ways.

The increase in thermodynamic efficiency by exhausting an expansion engine at temperatures down to 100 K, together with the use of isochoric compression from the liquid phase, are viable techniques for energy storage. Following a successful prototype, the first industrial 5 MW / 15 MWh cryogenic liquid energy storage plant is being constructed in the UK.

Meanwhile, the replacement of water injection in an IC engine with liquid nitrogen injection in the Dearman engine is being successfully pioneered as an alternative power source to IC engines.

4. Conclusion

A great number of cryogenic systems can be improved by the simple use of Cryogenic Fluid Dynamics to reduce boil-off and heat losses. The Southampton findings are very incomplete and much more research and development is needed to test and improve these findings, particularly at the larger scales of operation today.

My experience is that many users are unaware of the benefits which can be gained from simple changes in design. As helium sources become more expensive in the future, so the need for improved low-loss cryogenic systems will grow.

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