
ADVANCES IN CRYOGENIC SORPTION COOLING

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ABSTRACT

Advanced infrared and sub-mm wavelength instruments require the use of detector assemblies with improved sensitivity and background noise performance that can only be achieved at cryogenic temperatures. For many spaceborne missions currently in development, the high cost of achieving improved performance has led to lifetime requirements in excess of ten years; in addition to stringent requirements for vibration, size, weight, power and temperature.

Continued research in sorption cooler technology has resulted in cryocooler designs that offer competitive performance and long life. The substantial progress achieved in the past 3 years is leading to a Space Shuttle flight demonstration of this technology in January 1995.

This paper updates an earlier review of the subject (Wade, 1992a) by describing recent advances in the development status of sorption coolers, materials, and component technologies for spaceborne applications.

INTRODUCTION

This paper primarily reviews the status of sorption cryogenic refrigerator development efforts for spaceborne applications. The extensive commercial heat pump and solar engine developments are not discussed. Typical requirements for spaceborne applications include: multistage cooling of detectors, optics, and baffles; high reliability with lifetimes of up to ten years; high long term temperature stability; negligible microphonics, thermophonics, and cmi; and low system power consumption, mass and volume. Few refrigerators, of any type, have come close to meeting these requirements. The on-orbit performance of the few machines flown has been disappointing.

Because of the present lack of available long-life active

cryocoolers, using a liquid or solid cryogen filled dewar is the only practical method for achieving < 30K temperatures on-orbit (e.g. IRAS, ISO, SIRTf, and Wire). The penalty of utilizing a dewar in terms of mass, complexity and life is substantial. Even with warmer operational temperature, mission duration and mass are severely affected by the dewar. As a result of these impacts, research has continued to develop a closed-cycle cooler capable of meeting these challenging mission requirements. Sorption refrigerators have demonstrated the potential to meet this challenge.

In an attempt to eliminate many of the failure modes associated with mechanical refrigerators, the development of thermally driven sorption refrigerators was initiated. A sorption refrigerator combines a sorption compressor with an expander. Almost any kind of expander (e.g. turbo, Joule-Thomson (J-T), pulse tube, etc.) could be used. However, all but one of the sorption refrigerators developed so far have coupled the sorption compressor to a J-T expansion device (see Figure 1). In this combination, the compressed gas is expanded through the J-T orifice or capillary tube, producing a 2-phase gas/liquid refrigerant mixture without the use of any moving parts. The liquid refrigerant is subsequently separated and retained in a wick material at the cold-tip where it absorbs the detector heat load and is thereby evaporated. A pre-cooling stage is inserted before the J-T expansion to improve cooler efficiency. By resorbing the evaporated/expanded low pressure gas in a cooled sorbent bed, the process can be made into a fully reversible closed-cycle system. Alternations in heating and cooling of the compressor are achieved by heaters (if electrical input power is used) and heat switches. Passive check valves direct gas flows along appropriate paths.

The exact operating temperature of a refrigerator incorporating a Joule-Thomson expansion device is determined by controlling the back pressure above the collected refrigerant. As an example, by using hydrogen as the refrigerant the cold tip can be

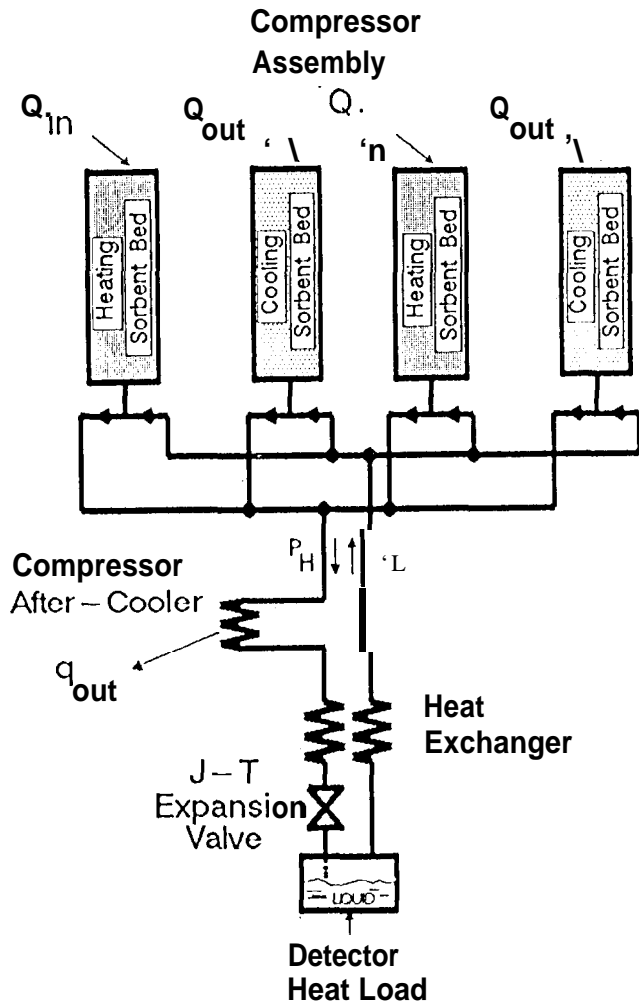


Figure 1. Schematic representation of a single-stage sorption refrigerator

effectively varied between 7.5 and 32K. Pressures above 4 atmospheres lead to temperatures above 26K being provided by liquid hydrogen; whereas for pressures below 50 torr, the hydrogen solidifies, thus providing a sublimating solid cryogen. Very low pressures, near 1 torr, lead to temperatures below 10K.

Sorption compressors are typically combined with J-T's because they are more easily designed to be efficient when producing the high pressure ratios and low flow rates favored by J-T valves. It should be noted that, with continued development, a sorption compressor could be combined effectively with a moderate mass-flow and pressure ratio expander like a pulse tube or a Gifford-McMahon.

Sorption compressors work on the principle that the amount of gas adsorbed, or chemisorbed, by a solid sorbent will decrease markedly with increasing temperature and will conversely increase with decreasing temperature. Thus a sorbent compressor

is able to adsorb quantities of gas at low temperature and pressure and subsequently release the gas at high pressure when heated. Hence compression is achieved thermally rather than mechanically. By nature, the sorption compressor is an intermittent system in which the sorbent bed alternates between gas generation and adsorption stages. However, by combining several sorbent beds in a single compressor and sequencing these sorbent beds out of phase, it is possible to produce a constant flow of gas to the J-T valve and thus achieve continuous closed-cycle refrigeration.

There are two types of sorption processes used by sorption compressors in cryogenic coolers: physical adsorption, and chemical absorption. Physical adsorption, or physisorption, relies on the relatively weak van der Waals interaction between the refrigerant gas and the sorbent material. Activated charcoals, zeolites, and silica gels are some of the materials used in physisorption compressors. During chemical absorption, or chemisorption, a chemical bond, usually covalent, is formed between the sorbent and the sorbate. Chemisorption materials include: hydrides (e.g. ZrNi and LaNi_{4.8}Sn_{0.2}) and praseodymium cerium oxide (PCO).

Efficient refrigeration at temperatures between 0.02 and 200K can be achieved by coupling stages utilizing different sorbent/sorbate combinations. Table 1 shows several options for sorption refrigerator stage sorbent/sorbate combinations.

TABLE 1. TYPICAL SORBATE/SORBENT COMBINATIONS FOR MULTISTAGE SORPTION COOLERS

Stage	1	2	3	3	4	5
Cold Temp. (K)	200-150	150-110	100-70	90-65	60-7	5-0.02
Sorbent	Saran Charcoal	Saran Charcoal	Saran Charcoal	PCO	Hydride	Saran Charcoal
Sorbate	xenon	Krypton	Argon or Nitrogen	Oxygen	Hydrogen	Helium

The absence of motors, and moving displacement devices near the cold end permits sorption cryocoolers to achieve the intrinsic vibration-free operating characteristics that are a strong point of J-T based refrigerant systems. Because the system is hermetically sealed with no lubricants or polymeric components, it is easily vacuum-baked to achieve the necessary system cleanliness and thereby achieve excellent operational reliability. Reliability is also enhanced due to the cooler's relatively simple hardware, and lack of close tolerances. Redundancy is incorporated in the form of extra compressor elements, which dramatically improves reliability without greatly increasing system mass.

In addition to vibration-free operation and high reliability,

sorption refrigerators have a strong efficiency advantage over other cryocoolers when cooling below 40K. Sorption coolers incorporating J-T expanders are inherently split systems and suffer little performance degradation when cooling a number of spatially distributed cryogenic loads. This also permits the compressor/valve assembly to be located in excess of 10 meters from the cryogenic cold tip due to the low mass flow rates and the resultant low pressure drops. Therefore a sorption refrigerator is considerably easier to package within a satellite than a mechanical refrigerator. Scaling of the cooler is nearly linear between loads below one milliwatt and over 5 watts. There appears to be no limit as to how large these machines can be scaled as commercial hydride sorption heat pumps have been developed with capacities in excess of 50 kW. Finally, as the sorption refrigerator is a thermal system rather than a mechanical one, it can operate using heat input from a solar collector or a radioisotope power source rather than electricity. Using heat rather than electricity to operate the refrigerator substantially reduces the overall system mass.

CURRENT AND ANTICIPATED DEVELOPMENT EFFORTS: REFRIGERATORS

Sorption cryogenic refrigerators are being developed for both continuous use and periodic operation applications. The major sorption cooler research efforts in progress are focused on periodic operation coolers using hydride-based sorption compressors (Bard, et al. 1993a).

There are two major kinds of continuous operation coolers: regenerative and nonregenerative. Regenerative coolers utilize some portion of the energy input to heat one compressor element, or sorbent bed, to heat the next in a successive cycle. Analysis demonstrated that over 98 percent of power input to non-regenerative coolers using activated charcoal as the sorbent material went towards heating the compressors. In non-regenerative coolers the energy used to heat the compressors is dumped, in its entirety, to the heat rejection radiator to cool down the elements. Regeneration of the waste heat clearly leads to substantial performance benefits. Regenerative designs are particularly attractive for physisorption systems as the heat of reaction (which for the most part can not be regenerated) is a small portion of the input power. Detailed studies on combined charcoal/PCO refrigerator designs have shown that while regeneration of the waste heat can lead to substantial performance benefits, the weight and complexity of this type of device generally restrict its use to applications requiring one watt or more refrigeration (Bard, et al., 1990).

Continuous Operation Non-Regenerative Cryocoolers

Continuous operation non-regenerative physisorption cryocoolers were first proposed by Vickers (1963). Over the years several coolers based on this principle were fabricated and tested, which used several different sorbents and refrigerants (Hartwig 1978, Bard 1986, Chan 1986, Schember 1989). The

compressor of Schember's physisorption cooler was operated for over 10,000 continuous hours.

Continuous operation, non-regenerative chemisorption cryocoolers were first demonstrated by Van Mal and Mijnheer (1971). This hydride based chemisorption cryocooler delivered approximately 1 watt of refrigeration at 26K. Numerous other demonstrations of continuous operation hydride cryocoolers have followed (Jones 1985, Karperos 1986, Matsubara 1986, Kumano, et al. 1988, and Zhang 1993). A second type of chemisorption cooler using PCO was developed by Bard and Jones (1988). The compressor of this cooler, which achieved cold tip temperatures as low as 72K, is still undergoing life tests with over 35,000 hours (78,000 cycles) of continuous operation accumulated as of 6/93. The compressor has experienced no measurable degradation over the course of these tests.

Very recent studies by the author have indicated that, for the 10 mW load requirements typical of low background, precision-pointing IR instruments, telescopes and interferometers, non-regenerative hydride based coolers can be extremely competitive with mechanical refrigerators. Figure 2 shows a schematic of such a miniaturized cooler. Analysis indicates that a cooler with this design is capable of providing 10 mW of refrigeration at < 10K with a mass of < 7 kg and average input power of only 10 watts. 65K precooling would be provided by a passive radiator in the flight version.

As shown in Figure 2, the proposed cooler consists of two low pressure (ZrNi hydride) sorption compressors coupled to a single high pressure (LaNi_{4.8}Sn_{0.2} hydride) sorbent bed operating on a 12 hour cycle, and J-T expander capable of operating over a temperature < 10K. The low pressure sorbent bed will absorb

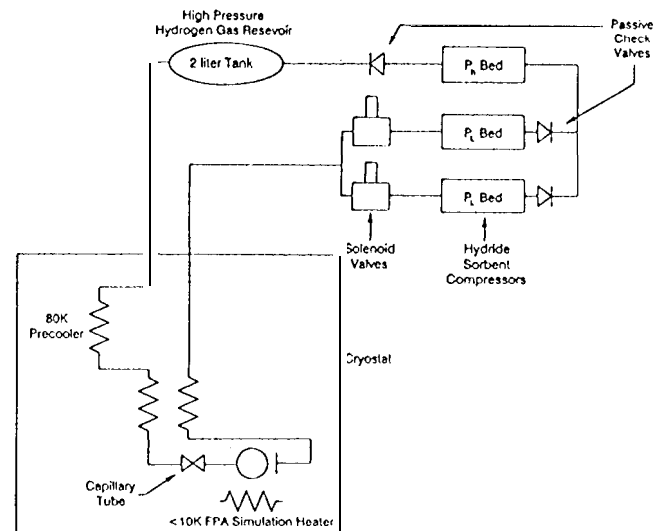


Figure 2. Schematic representation of a continuous 10 mW cooler with two stage compressor.

hydrogen at a pressure below one torr, thereby allowing temperatures below 10 K to be achieved. The hydrogen storage tank serves to supply the J-T expander with refrigerant at approximately 1600 psia (11 MPa). Precooling of the hydrogen to below its inversion temperature is provided by liquid nitrogen (during ground test), which simulates the 60-80K space radiator upper stage. After expansion (and absorbing the detector load) the now low pressure hydrogen is absorbed by the low pressure sorbent bed. A sorbent bed, which is ready for absorption, is selected by activating a solenoid valve. When heated, the low pressure bed will desorb the hydrogen refrigerant at a pressure of 15 psia (0.1 MPa) and a temperature of 575K. The high pressure bed will absorb the hydrogen desorbed by the low pressure bed. Upon heating the high pressure bed will desorb the hydrogen refrigerant at a pressure in excess of 1600 psia (11 MPa) into a hydrogen storage tank. After desorption is complete, the compressors cool by natural convection back to the laboratory ambient temperature (during ground tests) at which time they are ready to begin another absorption cycle. By operating two low pressure sorbent beds 180° out of phase, continuous refrigeration can be achieved. The gas flow during the recharge cycle is directed through passive check valves.

A variant of this concept design which incorporates parallel, two-stage compressors operating 180° out of phase is capable, in a flight configuration, of delivering 100 mW at 20K and 10 mW at < 10K with a mass of only 25 kg and an input power of 2S watts. It is likely that coolers of this type will find application with ground-based, as well as spaceborne, astronomical instruments. In addition, they may prove useful for discrete cooling of commercialized high and low T_c superconducting devices.

Continuous Operation Regenerative Cryocoolers

Regenerative sorption coolers were first proposed for commercial application by Miller (1929). Their application to cryogenic coolers was not proposed until 1989 by Suwulka (Wade, et al., 1990). The first cryogenic regenerative cooler was demonstrated in 1991 (Wade, et al., 1992b) and operated for over two thousand hours.

Two regenerative cryocoolers are now in development. A 10K continuous operation, multistage cryocooler, which uses a mechanical cooler for precooling at 65K is currently under development by Aerojet Electronic Systems Division under contract to the Phillips Laboratory, Kirtland Air Force Base, New Mexico. In addition, a two-stage charcoal physisorption regenerative cryocooler which will operate at 130K is under development at Aerojet with internal research funds (Andrcas, 1993).

Periodic Operation Sorption Cryocoolers

Johnson and Jones (1993) pointed out that periodic sorption cooler technology offers repeatable quick cooldowns, and low average power consumption due to intermittent operation. This concept design provides refrigeration for a relatively short time

with a longer period for recuperation. By spreading the energy requirement over a period of hours a remarkably low power requirement is achieved.

Concept design studies of a periodic operation sorption cooler indicated that a compressor using ZrNi hydride could achieve pressures below 1 torr, thereby enabling a cold tip reservoir of solid hydrogen to be cooled below 10K. A Proof-of-Principle (PoP) cooler was then built to test the feasibility of the periodic operation concept (WU and Bard, 1993). The PoP cooler validated the proposed concept by achieving cooldown to below 10K from an 80K holding temperature in less than two minutes. It then maintained a cold tip temperature below 10K while absorbing a 150 mW simulated detector heat dissipation for 45 minutes. Figure 3 shows cooldown test data for this cooler.

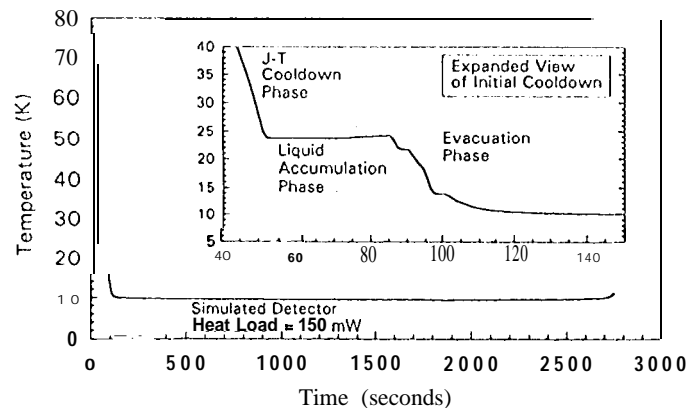


Figure 3. Cooldown performance of the Proof-of-Principle 10K periodic cooler.

To advance the maturity of these concept designs, and mitigate risks associated with this novel cryocooler technology, a near term spaceflight demonstration program, the Brilliant Eyes Ten Kelvin Sorption Cryocooler Experiment, is scheduled for a Space Shuttle Launch in early 1995. A general description of the BETSCE effort, including functional requirements and major component specifications, has been reported by Bard et al. (1993 b). An artist's rendition of the BETSCE instrument is shown in Figure 4. Specific BETSCE objectives include: (1) Demonstrating 10K sorption cooler technology in a microgravity environment; (2) Advancing the enabling technologies and developing integration techniques by developing an automated, space-flightworthy instrument; (3) Characterizing spaceflight performance and developing the needed flight database to aid the future cooler development effort.

More advanced versions of the BETSCE cooler, called the Periodic 10K Sorption Cryocooler Engineering Model and the Protoflight Unit, will be developed as part of the JPL Sorption

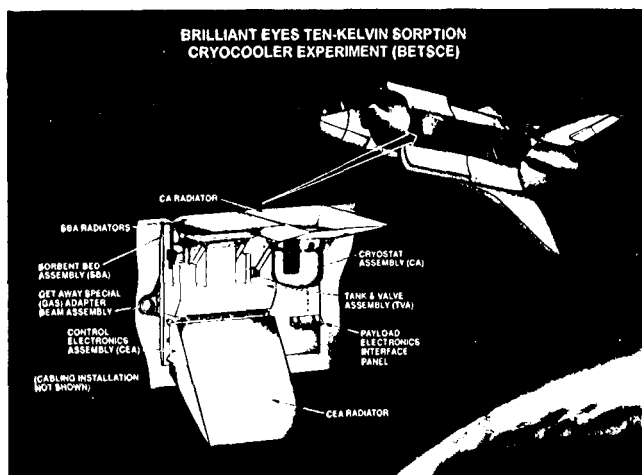


Figure 4. Artist's rendition of the BETSCE instrument mounted on a Get-Away Special (GAS) adapter beam.

Cooler Basic Technology Development Program. This effort will be contracted out to industry and will provide a vehicle for technology transfer; in addition to furthering the maturity of periodic operation sorption devices. As with BETSCE it is intended that these coolers be designed for spaceborne instrument applications. In particular, they are to incorporate advanced container materials and components. It is also intended that these coolers will be used to conduct hardware life demonstrations in excess of ten years duration.

Two very different periodic machines have demonstrated the ability to periodically cool to well below 1 K. Roach and Gray (1988) demonstrated a dilution refrigerator using activated charcoal to pump the helium, which operated at temperatures below 20 mK. Duband et al. (1990) demonstrated a helium 3 refrigerator using activated charcoal, which was used on a sounding rocket launched instrument to examine the background radiation from the big bang. This cooler provided 100 microwatts at 346 mK.

CURRENT AND ANTICIPATED DEVELOPMENT EFFORTS: COMPONENTS AND MATERIALS

The major component and materials development effort currently underway is the JPL Sorption Cooler Development Program, funded by the Ballistic Missiles Defense Organization (BMDO) through the Brilliant Eyes Program. In addition to the cooler development effort mentioned earlier, a substantial component of this program is the reliability physics effort (Wade, et al. 1993a). The goal of the reliability physics effort is to establish the technology base required to design 10 K sorption coolers that can operate reliably in space for 10 years or more. The detailed understanding, gained through this research, of

important failure mechanisms governing hydride refrigerator life is enabling identification of operating constraints, design enhancements, and other means of eliminating failure mechanisms, as well as aiding detailed definition of effective techniques for fabrication, assembly, and handling.

Prototype compressor performance characterization experiments have validated the sorbent compressor designs used in the BETSCE sorbent bed assembly (Wade, et al. 1993b). The performance tests conducted have also permitted validation of the transient analysis models. No compressor tested to date has deviated by more than six percent from the predicted performance. In addition, hydride sorbent bed tests have demonstrated two stage compression from less than 1 torr to over 11 MPa. Hydride compressor long term cycling has been initiated with over 3,500 hours accumulated on one $\text{LaNi}_{4.8}\text{Sn}_{0.2}$ hydride sorbent bed. Ten year equivalent endurance testing of six compressors is planned.

PCO compressor cycling is still underway after over 35,000 hours of endurance testing. These compressors show only a slight improvement in capacity and kinetics. It would appear that long-term degradation does not pose a substantial problem for this material.

Tests have indicated that commercially available flight-qualified valve lifetimes greatly exceed even the ten-year requirement (Futurecraft 1992, Wade, et al. 1992b). This is primarily because most sorption coolers operate with cycle times ranging between 20 minutes and 12 hours. At most, only 263,000 cycles are accumulated over ten years continuous operation. Heater and Inconel compressor life and creep rate tests are consistent with lifetimes in excess of ten years (Wen, et al. 1990). Li, et al. (1993) reported on the hydrogen compatibility of JBK-75 after high temperature hydrogen soaking. This material appears well suited for advanced hydride containers.

The only component which still requires substantial development is the fluid loop circulator used in regenerative sorption coolers. No efforts are currently underway to demonstrate a flight version of an appropriate circulator.

Under the material characterization portion of the JPL Sorption Cooler Development Program reliability physics effort, investigations are underway with candidate hydride materials (e.g. $\text{LaNi}_{5-y}\text{Sn}_y$ and ZrNi). These studies include: (1) characterization of hydride chemical and metallurgical composition, isotherms, kinetics, phase purity, lattice parameters, disproportionation mechanisms, and thermal conductivity; (2) study and definition of hydride manufacturing, annealing and handling procedures; and (3) characterization of long-term hydride stability, degradation, compaction and containment using specially fabricated compressors used to precisely measure the transient pressure response to programmed temperature cycling. This work is a cooperative effort between researchers at JPL, California Institute of Technology, University of Vermont, Ames Laboratory at Iowa State University, Aerojet Electronic Systems Division, etc.

Characterization is well advanced for $\text{LaNi}_{4.8}\text{Sn}_{0.2}$ and ZrNi hydrides and they appear well suited for long-term flight applications. Characterization of the family of $\text{LaNi}_{5-y}\text{Sn}_y$ hydride

tin substitutions for $0 \leq y \leq 0.5$ have been initiated. Isotherm measurements for ZrNi, and the LaNi_{5-y}Sn_y family for $0.1 < y < 0.5$, are reported by Wade, et al. (1993a). The operating (plateau) pressures can be varied nearly two orders of magnitude by varying the level of tin. The ability to shift plateau pressure by varying the level of tin substitution provides a designer with the ability to optimize performance. With a high tin substitution stoichiometry it is possible to build a single-stage hydride compressor, which, given precooling in the 60 to 80K region, will support cooler operation at temperatures down to 15K.

Substantial work has been accomplished in the characterization of physisorption sorbents with various gases. Thanks to the outstanding efforts of Radebaugh (1991) of the National Institute of Standards and Technology (NIST), there is a large database of refrigerant capacities of a variety of activated charcoals, as a function of temperature and pressure. The development of this database has permitted accurate sizing, and design, of compressors using activated charcoals. The database for helium on various charcoals has been extended by Helvensteijn and Kashani (1993) to cover very low temperatures and pressures.

Refrigerant gas properties are available from both NIST and commercial vendors. Of particular interest are the properties of gas mixtures. Al fecv, et al. (1973) was the first to recognize that substantial performance advantages and hardware simplification could result from using soluble gas mixtures as a refrigerant. Jones (1991a) has developed a code for analyzing cryogenic mixed gases. Other studies of gas mixtures for J-T coolers have been performed by Robinson (1993) and Little (1993). In addition, Jones et al. (1991b) have conducted research which validated the feasibility of a physisorption cooler using mixed gases to 65K.

CONCLUSIONS AND COMMERCIAL APPLICATIONS

A near term spaceflight test of a sorption refrigerator is planned for early 1995 (BETSCE). The level of design maturity which has developed as a result of this effort has substantially advanced the state-of-the-art. A major effort is underway to rigorously research the reliability physics aspects required to support mission lifetimes in excess of ten years. As part of this effort basic characterization, performance tests and endurance tests of component and materials are being conducted. The longevity of the prototype compressors and refrigerators tested to date is highly encouraging.

It should be noted that the work reported in this paper has the potential to create substantial commercial spinoffs. The application of miniature sorption coolers to remote ground-based instruments requiring cryogenic cooling, and for discreet component cooling of both high and low T_c superconducting devices appears to offer great promise. The commercialization of high temperature superconductors will not occur without reliable, inexpensive, and user transparent cryocoolers. It appears likely that for applications requiring cooling down to 65K mixed gas sorption coolers are a viable solution. Finally, Jones

(1992, 1993) has developed a regenerative sorption heat pump for home use. This heat pump, which is being developed commercially by the Aerojet Electronic Systems Division, has projected operating costs comparable to vapor compression systems for cooling. In the heating mode, fuel costs are calculated to be about half that of either vapor compression or fossil fuel heating systems.

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