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Construction techniques for adiabatic demagnetization refrigerators using ferric ammonium alum

Grant W. Wilson [a,*], Peter T. Timbie [b]

[a] *University of Chicago, 5640 S. Ellis Avenue, Chicago, IL 60637, USA*
[b] *Univerisity of Wisconsin at Madison, 1150 University Avenue, Madison, WI 53706, USA*

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Abstract

We describe techniques used to fabricate the cold stage of an adiabatic demagnetization refrigerator that uses the paramagnetic salt ferric ammonium alum. We discuss the design of a leak-tight housing for the salt as well as a technique for growing ferric ammonium alum crystals that results in a housing filled with > 98% refrigerant. These techniques have proven to be reliable in creating robust single-stage refrigerators. Similar techniques can be used for the second stage of a dual-stage adiabatic demagnetization refrigerator. © 1999 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Adiabatic Demagnetization Refrigerators (ADRs) have been used as reliable tools for cooling to temperatures below 100 mK for over 60 years [1]. In the 1970s, however, ADRs took a back seat in the typical low temperature laboratory environment to the dilution refrigerator, which offers higher cooling power (although at the expense of additional complication). The last several years have witnessed the development of two-stage ADRs [2] which, when combined with a low-temperature mechanical cryocooler, will achieve temperatures below 100 mK without cryogens.

In an ADR, a paramagnetic material is suspended in a magnetic field, typically provided by a superconducting magnet. The refrigerant is thermally isolated from higher temperature stages of the cryostat and thermally connected to the parts that are to be cooled. Many suitable paramagnetic materials are salts of hydration, and the salt is housed in a hermetically sealed container (the "salt pill") to prevent dehydration in the cryostat's vacuum. In this article we discuss robust and reliable fabrication techniques for a salt pill made with ferric ammonium alum (FAA or $Fe(NH_4)_2(SO_4)_2 12H_2O$), a paramagnetic salt often used in ADRs designed to achieve temperatures below 100 mK. These techniques were developed to fabricate several pills for the ADR flown on the Medium Scale Anisotropy Measurement (MSAM II), a balloon-borne measurement of the Cosmic Microwave Background Radiation [3]. We begin with a description of the physical properties of FAA, highlighting those properties which must be addressed in ADR fabrication. See Hagmann et al. [4] for a complete thermal description of FAA ADRs. We then describe our salt pill housing (SPH) which is a variant on that described in [5]. We round out the design with a description of our FAA growth technique, which has yielded > 98% filling in five completed salt pills.

2. FAA and the SPH design

There are a number of constraints on the SPH which must be satisfied to ensure durability, a short internal thermal time constant, and low parasitic heat loads for the ADR. Fig. 1 shows the SPH which was used in the MSAM II radiometer and diagrams our solutions to the issues raised below.

* Corresponding author. Tel.: + 1-773-702-4314; fax: + 1-773-834-1891; email: wilson@flight.uchicago.edu

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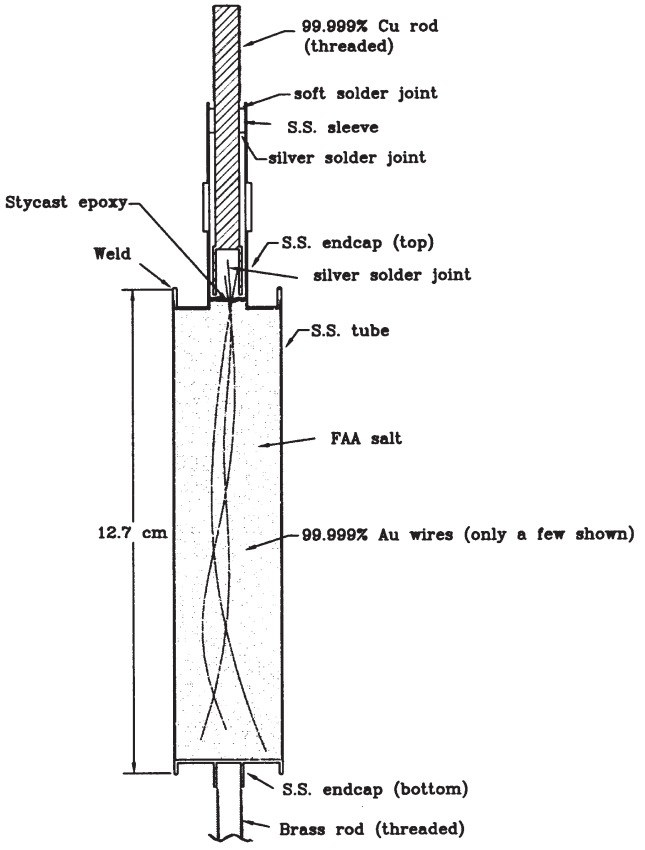

Fig. 1. Salt pill housing for our ADRs. The FAA is contained by a thin-walled stainless steel can which is welded at the two ends. Heat is extracted from the sample by a Cu braid (not shown), drawn down the Cu rod through the Au wires and into the cool salt. An epoxy barrier protects the Cu rod from the corrosive FAA.

2.1. FAA properties

Ferric ammonium alum (FAA) has a spin ion density of 1.17×10^{24} kg^{-1}, a total spin angular momentum, $J = 5/2$, and an internal magnetic field of approximately 50 mT [4]. Its magnetic properties, combined with its high solubility in water, make FAA a practical working substance and nearly ideal paramagnetic material for 100 mK operation from a pumped LHe bath. Table 1 summarizes some of the relevant physical properties of FAA.

Unfortunately, FAA has a few properties which complicate its use. FAA is corrosive to copper-based alloys. This effect is not subtle. We found that gold plating the inner walls of a brass tube was not enough to prevent the salt from eating through the 500 μm tube wall within a few days. Consequently, no copper alloys used in the SPH can be allowed to come into contact with the FAA. We also attempted to grow FAA pills in MACOR (a glass-ceramic made by Corning) and Plexiglass housings. In both cases the housings cracked during salt growth; the FAA expands slightly when crystals form. Currently, we use type 304 stainless steel as the housing material. 304 stainless is non-reactive with FAA, welds easily, and is strong enough to support the FAA/cryostat vacuum transition with a 250 μm wall thickness. The biggest drawbacks of stainless steel are its large heat capacity and poor thermal conductivity, properties which are manifested by an increase in the time constant of the salt pill.

Table 1
Some physical properties of FAA

Chemical symbol	$Fe(NH_4)_2(SO_4)_2 12H_2O$
20°C solubility	1.2 g/cc
35°C solubility	4.0 g/cc
Crystal density	1.35 g/cc
Spin angular moment J	5/2
Spin ion density	1.17×10^{24} kg^{-1}
Internal field B_{int}	50 mT
Ordering temperature	35 mK

FAA undergoes a chemical transition when heated above 40°C which permanently destroys its paramagnetism. While still in solution, the transition is obvious by a change in color of the solution from brown to nearly black. FAA crystals which are heated above 40°C change their crystalline structure and lose their structural integrity (i.e. they become mush). We have experimentally determined that an FAA pill which has been heated above 40°C is no longer a useful refrigerant.

Finally, FAA dehydrates when exposed to a vacuum. The resulting powdery substance does not act as an active paramagnet at low temperatures. Consequently, the SPH must be a vacuum-tight container. Several techniques have been developed to construct this container without heating the FAA above 40°C. A common method for sealing the ends of the SPH is to glue them closed using an epoxy which is thermally well matched to the SPH material (such as Stycast 2850GT). However, to improve on the reliability of this joint we chose to TIG-weld the endcaps of our pill into place with a microwelder. We have found that careful welding of the joints while attaching the body of the SPH to a heat sink or wrapping with a wet cloth results in a reliable and robust leak-tight SPH with negligible loss of refrigerant due to over-heating.

2.2. Thermal link and eddy current heating

Establishing a good thermal link between the FAA and the rest of the cold stage is one of the most difficult parts of the SPH construction. Since the salt itself is a very poor conductor, a good conductor must occupy a substantial volume of the SPH to keep the time constant of the system short. Conversely, the larger the effective cross-section of the thermal link, the larger the eddy current heating during times when the magnetic field is changing. We use a nest of ~ 200–250 μm diameter 99.999% Au wires (Suprepure Chemicals Inc., Florham

Park, NJ, tel: (201) 377-4081) which is suspended in the salt, and silver soldered with non-superconducting cadmium-free silver solder at one end to a high purity 99.999% Cu rod (Johnson & Matthew West, Chester, PA, tel: (610) 648-8070). The Cu rod and silver-solder joint are isolated from the FAA by a layer of Stycast 2850GT epoxy (Emerson & Cuming Inc., Canton, MA, tel: (617) 821-0737). These wires could be arranged uniformly; we have not done so in the salt pill described here.

The total amount of eddy current heating in the SPH can be calculated [1]. The power dissipated in a metallic rod of radius r, volume V, and resistivity ρ is

$$P = \frac{1}{8}\rho^{-1}r^2VB^2 \text{ W} \quad (1)$$

where B is the rate of change of the magnetic field in units of T/s. If we model the gold wires as independent rods aligned with the magnetic field, the eddy current heating for each wire is

$$P_{\text{goldwires}} = 4.3 \times 10^{-7}B^2 \text{ W} \quad (2)$$

and for the stainless can (the outer shell)

$$P_{\text{stainlesscan}} = 0.12B^2 \text{ W} \quad (3)$$

Using our specific system as an example of the eddy current heating, we consider a 12.7 cm long stainless SPH with an OD of 3.2 cm containing 200–250 μm Au wires. This SPH will hold roughly 120 g of FAA. In our case we ramp from a full field of 3 T to a controlling field of 100 mT (roughly 100 mK) in aproximately 10 min. During this ramp eddy current heating in the 200 gold wires contributes 0.89 μJ while the stainless can adds 1.8 mJ—both insignificant amounts compared to the 134 mJ of enthalpy of the salt. During a balloon flight, the deviations in field while controlling at 100 mK are small compared to the ramp down (typicaly $B = 1.2 \times 10^{-6}$ T/s). Thus, eddy current heating in the pill is not a concern.

3. Salt pill growth

The FAA crystals are grown from a heated saturated solution which is slowly cooled to room temperature. As mentioned above, FAA undergoes a chemical transition when heated above 40°C which destroys its paramagnetism permanently. Consequently, the growth of the pill requires a careful balance between keeping the FAA solution as close to 40°C as possible, to maximize the FAA in solution, without exceeding the critical temperature. We have developed a simple method for reliably filling our SPH with FAA crystals using a home-made oven and standard laboratory chemical equipment.

The "oven", shown in Fig. 2 is composed of two concentric Plexiglass tubes. The inner tube has the same outer diameter as the SPH and is wound tightly with Cu magnet wire which acts as the heater. The SPH is attached to this tube by a vacuum-greased semi-flexible rubber hose. The outer tube and several Styrofoam spacers act as baffles to reduce convective cooling to the heating element. The goal is to keep a column of FAA solution near 38°C for one week. The required heat input is calibrated by filling the inner tube and SPH with water and measuring the temperature as a function of height along the tube length. We found that 7.07 W of input power over 91 cm of tube resulted in the temperature ranging from 36°C at the bottom of the tube to 38°C at the top. The temperature of the solution could be regulated with a feedback loop, but we found this complication to be unnecessary.

The process for growth is straightforward. We mix the FAA solution in a beaker on a hot plate and then pour the solution into the inner tube and SPH with the heater

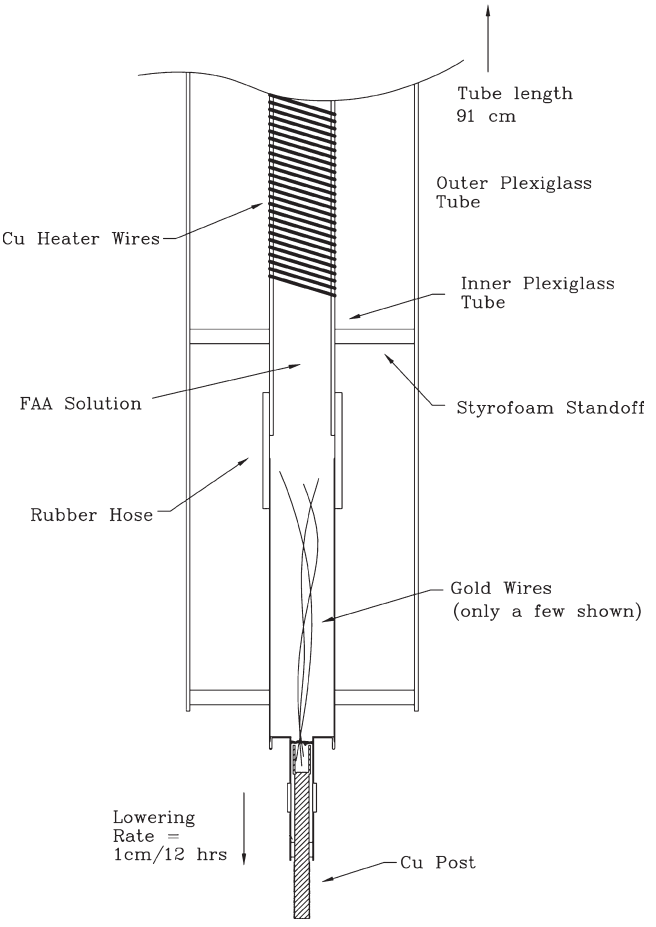

Fig. 2. Oven for salt pill growth. A tightly wrapped Cu wire heats the inner tube to keep the FAA solution near 38°C. The SPH is slowly lowered into the cool lab air over the course of many days to force the growth of salt crystals from the bottom of the SPH upwards.

turned on. The growing assembly is lowered out of the outer tube in steps of 1 cm every 12 h. Lowering the SPH into the 20°C ambient lab air at this rate helps ensure that salt crystals will grow from the bottom to the top. If crystals do begin to form near the top, it is possible for them to seal off the lower volume, starving it of fresh solute from the reservoir above. Note also that because the gold wires are thermally anchored to the Cu post, which is always near room temperature, crystals will begin to form on the gold wires. The slow cooling rate also allows relatively large crystals to form. Presumably, the thermal conductivity from the wires into large salt crystals is better than into many smaller crystals.

When the SPH is fully retracted from the outer tube, the current through the copper heater wires is stepped down by 1.75 W each 12 h until zeroed. Twelve hours after this the SPH is detached from the oven and excess salt is removed to make room for the second endcap to be welded in place.

We have used this growth technique to fabricate 5 salt pills. In each case density measurements showed that the SPH was filled with salt at the 99 $\pm$ 1% level. These measurements were confirmed by cutting open one of the pills and inspecting for voids. None were found. Furthermore, inspection revealed that the nest of gold wires was encased in FAA crystals which were a few millimeters in size.

4. Conclusion

We have described the vacuum-tight housing and growth technique for fabricating an FAA paramagnetic salt pill for an adiabatic demagnetization refrigerator. We have used these techniques to build a number of FAA ADRs for use in the laboratory and measurements from a scientific balloon. The salt pill housings have proven to be robust and reliable, with the oldest still running after 4 years of use. The growth technique has given consistent and pleasing results. With simple laboratory chemical supplies it is possible to grow large-crystal FAA pills with a > 98% filling factor.

While an FAA ADR can be used on its own to reach temperatures below 100 mK, the future of ADR cryogenics lies in dual-stage devices. The techniques described here are suited for mass construction of FAA ADRs as a second stage in these refrigerators.

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