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Chemical Compatibility and Wettability of various Materials with various Working Fluid for Two-Phase and Heat Pump Systems

ESR: Executive Summary Report

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1. INTRODUCTION

This document summarises the activities of the contract work investigating the chemical compatibility and wettability of various combinations of metals and working fluids applicable to two-phase and heat pump systems. In order to maintain nominal operation of electronic devices used in spacecraft, thermal control systems are used to transfer the generated waste heat to radiators where it is dissipated to cold space. Increasing onboard power consumption budget requirements means that cooling by liquid pumping alone can no longer remove adequate amounts of heat to maintain safe operation conditions. Two-phase and heat pump cooling solutions can overcome these limitations whereby the heat is removed during evaporation of a coolant fluid. They can be very effective solutions offering the advantages of reduced size and weight, as well as reduced power consumption through passive pumping methods.

Chemical compatibility is a particularly important consideration for two-phase systems as they can only be manufactured from materials which are considered inert with the selected coolant fluid. Performance and system degradation can occur across the whole spacecraft lifetime as a result of chemical reaction or decomposition of the fluid and corrosion of the container. Any gases generated during chemical reactions can accumulate in the condenser section, eventually leading to blockages or device failure.

In addition to this, wettability, which is a property of a solid material that allows liquid to adhere to its surface, is important for correct two-phase device operation as it generates the required pressure to circulate the fluid by capillary pumping. Wettability is described by the contact angle formed between a solid material and an interacting gas-liquid interface.

The recent emergence of Additive Layer Manufacturing (ALM), otherwise known as 3d-printing, as an important commercial technology has the potential to create highly complex and innovative components for two-phase cooling systems. However, the chemical compatibly between ALM metallic materials and coolant fluids is currently unknown. Much of the data regarding chemical compatibility between different fluid-metal combinations dates from the 1960's-1980's and requires updating due to the latest advancements in materials and manufacturing processes.

The objectives of this study were therefore to investigate these two aspects, by firstly defining a suitable list of interesting fluid-metal combinations, and then secondly undertaking the manufacture of the required test samples for detailed experiment characterisation. The sole contractor for this work was the University of Limerick (UL), Ireland, which undertook the design, development and testing required to meet these objectives.

2. MATERIALS & METHODS

2.1. Chemical compatibility

The selection of a coolant working fluid for two-phase systems is generally based on its capability to effectively transport heat as it changes phase. Transformation from a gas to a liquid occurs when it absorbs heat, and then after transport to another location, releases it again as it changes back from a gas into a liquid. To aid in the selection of fluids to study from the wide range of available possibilities, an analysis was used to predict their thermal performance in a number of two-phase devices. These included heat pipes, loop heat pipes and heat pumps.

Ammonia is the most common fluid used in spacecraft two-phase equipment as its heat transfer performance is greater than other fluids within the environmental operating temperature range found in space. For terrestrial applications, water has by far the best predicted performance, as well as other advantages such as low safety risks and wide availability, often make it the cooling fluid of choice. However its higher melting point compared to ammonia and know corrosion issues with aluminium means it is less useful for space applications. It has found potential usefulness for high-temperature applications so it is included here for study. As well as these, other fluids with high predicted performances were also selected and are namely acetone, methanol, toluene, propylene, and ethylene glycol.

Materials used for the manufacture of pipes, wicks, valves and other components of two-phase systems are generally selected based on their capability to maintain the internal pressure of the working fluid, good thermal conductivity for heat transfer and good strength-to-weight ratio, as well as historical chemical compatibility and wettability information. As the focus of this activity relates to ALM, different commonly used and commercially available metal alloys where selected for study. These include aluminium AlSi10Mg, 316L grade stainless-steel and titanium Ti6Al4V. In addition, aluminium AlSi7Mg has been identified as an important alloy for aerospace applications, and ALM Invar has many potential applications for spacecraft thermal control of sensitive optical equipment because of its very low coefficient of thermal expansion. Conventionally manufactured materials, such as aluminium Al6061 and 316L grade stainless-steel, were also included to act as a direct comparison to the ALM metals. Bimetallic couplings, which allow for connection of components made from dissimilar materials were also investigated as chemical compatibility data is scarce. Finally, shape memory alloy couplings produced by SCouP srl which can achieve leak tight sealing for pipes in areas inaccessible for welding were tested with ALM materials.

The standard method for experimental life-testing of two-phase devices is known as the Gas Plug Test and was employed in this study. As the main interest is in understanding the chemical compatibility between the fluid-metal combinations, the simplest two-phase device in terms of design will be used, namely a thermosyphon. This device consists of a long narrow tube of circular cross section, sealed at one end. Dimensions used were 12.7 mm outer diameter, 0.9 mm wall thickness, and lengths varying from 180 – 400 mm depending on material thermal conductivity. The open end is used to firstly evacuate any air inside before being back-filled with a given working fluid and then hermetically sealed. Compared to the base metal alone, their effective thermal conductivity can be greater by several orders of magnitude. The thermosyphons are set up in the Gas Plug Test in a vertical orientation, heated at the lower evaporator end by applying a constant temperature, and then monitored over an extended period of time for any change in the condenser temperature at the top end of the device. An increase in the temperature difference along the device is indicative of a potential internal build-up of unwanted non-condensable gas (NCG).

In order to confirm the capability of the ALM process to manufacture leak proof devices, extensive materials testing was firstly undertaken. This involved micrometry measurements to ensure correct replication of the specified sample diameter and wall thickness geometries, porosity measurements to ensure no voids were present in the solid materials, surface roughness measurements, and cross-sectioning for examination using scanning electron microscope and spectroscopy techniques. An example of some of the ALM thermosyphon samples after metal 3d-printing are shown in Figure 1. All samples were subjected to proof pressure testing at 1.5 times there maximum design pressure and helium leak testing to ensure safe operation.

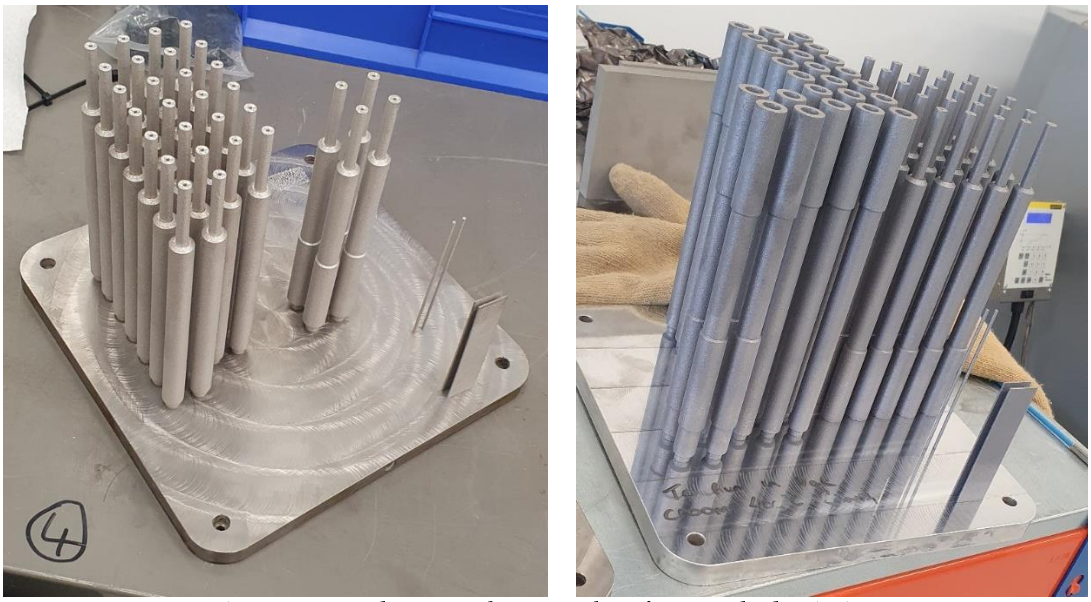

Figure 1: ALM thermosyphon samples after metal 3d-printing.
Left 316L stainless steel Right: aluminium AlSi10Mg

Filling of the working fluids was achieved through the design and development of specific test benches capable of charging devices with fluids which are either a liquid or gas at standard temperature and pressure. The test benches were used to fully evacuate the samples using a vacuum pump and degassing of any NCG dissolved in the working fluids prior to filling. Liquid dispensing was achieved with a graduated burette integrated into the test bench. For gases, such as ammonia and propylene, a technique known as vapour transfer, or distillation, was used to accurately transfer the fluid from a supply tank firstly into a calibrated intermediate tank, before then being transferred again into the thermosyphon sample. Filling volumes were in the order of 1.5-4.0 ml. A significant amount of time during the manufacturing phase was also dedicated to developing a methodology for producing the hermetical seal using a hydraulic crimping process which deforms and seals the open end of the tube.

Figure 2: Gas working fluid filling test bench

Cleaning of samples is of critical importance at all stages of two-phase device manufacture to avoid compatibility issues. Different cleaning steps included removal of any loose ALM particles, ultrasonic cleaning in solutions of organic and alkaline solvents, acid cleaning treatments and flushing with the corresponding working fluid before final filling.

In total, 33 different fluid-metal combinations were selected for study. For each combination, 3 identical samples were manufactured for redundancy purposes, as well as to allow selected samples to be removed from the gas plug test for additional characterisation testing as required. Gas plug testing took place inside environmental chambers to allow for constant ambient conditions throughout testing. Total test duration was up to 9000 hours.

Figure 3: Environmental Chambers. Left: Exterior with controller and data logging equipment. Right: chamber interior with samples mounted in heater blocks

2.2. Wettability

Due to the nature of some of the working fluids at standard temperature and pressure, these experiments were performed inside a pressure vessel where they existed in a state of two-phase equilibrium. The experimental procedure involved observing the meniscus produced by the different ALM test coupons when they were in contact with the working fluid. Dynamic contact angle measurements were performed whereby the liquid was either ascending or receding along the solid material. This was achieving by causing the liquid level inside the pressure vessel to rise or fall by imposed pressure differentials. A macro lens and camera allowed for recording of images through a glass viewport with the aid of a backlight source. The ALM coupons were 3d-printed along with the thermosyphon samples and are visible as the smaller rectangular and circular shaped samples in Figure 1.

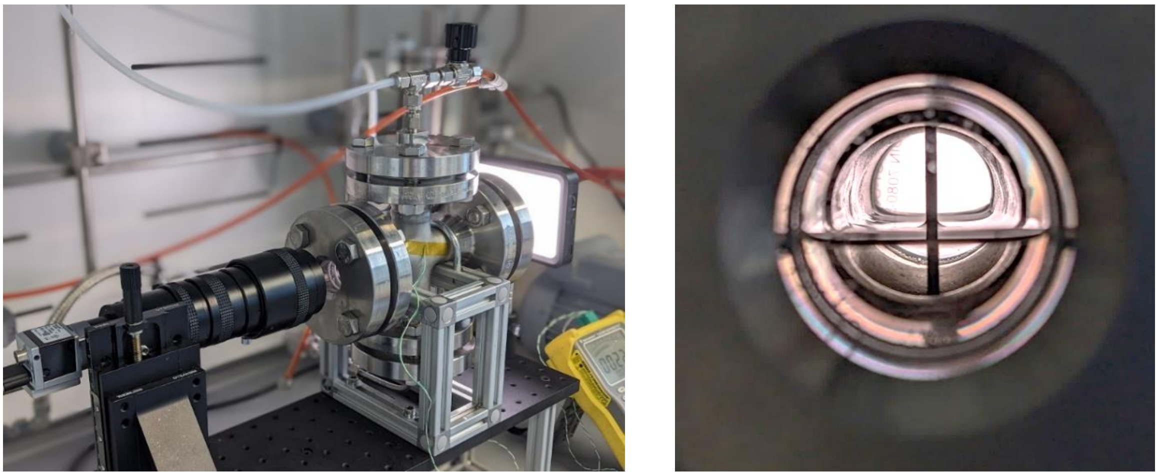

Figure 4: Wettability experiment setup. Left: image acquisition in progress. Right: sample partially submerged in high pressure liquid ammonia visible through viewport.

A post-processing methodology was developed to extract the quantitative contact angle data from the resulting images. This involved detecting the locations of the sample and meniscus in each image frame, and then using theoretical equations to determine the meniscus shape and resulting point of intersection with the coupon, as shown in Figure 5.

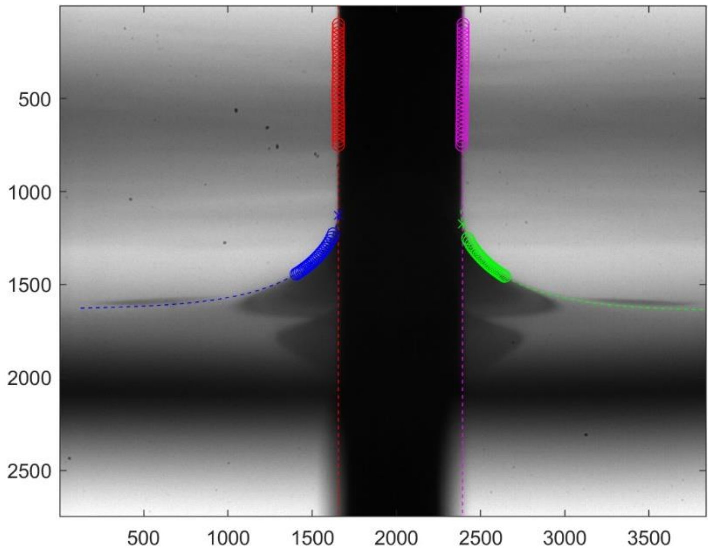

Figure 5: Sample wettability image showing vertical metal coupon and liquid-vapour meniscus profile. Coloured regions highlight results of detection methodology,

3. RESULTS AND DISCUSSION

3.1. Chemical compatibility

The findings of chemical compatibility testing between the different fluid-metal combinations are listed in Table 1. They are coloured as compatible (green), incompatible (red) and inconclusive (yellow). The majority of cases were found to be compatible, with a number of notable exceptions. These results were determined through a combination of the gas plug test, and where generation of NCG was suspected to have taken place, additional characterisation was performed. Where possible, the gas was identified by gas chromatography analysis, then sectioning of samples took place to locate any areas of corrosion (some examples are shown in Figure 6), and X-ray Photoelectron Spectroscopy (XPS) was used to determine the surface chemical condition.

Table 1: Results of chemical compatibility testing for metal-fluid combinations

Metal / Fluid	Conv.		ALM					Bimetal	SCouP ALM		
	Al6061	SS316L	SS316L	Ti6Al4V	Invar	AlSi7Mg	AlSi10Mg	Al6061-SS316L	SS316L	AlSi7Mg	AlSi10Mg
Acetone											
Ammonia											
Ethylene glycol											
Methanol											
Propylene											
Toluene											
Water											

In regards to spacecraft heat transfer, where the most common fluids are either ammonia or propylene, all the test metal combinations listed in Table 1 were found to be chemically compatible with little to no NCG generation. Samples run at very low temperatures showed a very stable temperature difference between the top and bottom sections of the device. Gas chromatography analysis detected small quantities of nitrogen gas present in the samples which was believed to have been inadvertently introduced during filling. Ammonia was also found to be compatible with the different coupling configurations tested.

For the low pressure solvents of acetone, methanol and toluene, most materials were also found to be compatible except in two cases. For methanol-invar, rapid decomposition of the fluid occurred due to the nickel in the alloy acting as a catalyst to the chemical reaction. For the bimetallic stainless steel/aluminium-toluene case, galvanic corrosion at the interface between the metals was observed, as well as the generation of NCG. These two cases are therefore deemed incompatible.

Figure 6: Cross - sectioned samples. Top right: SS316L ALM-water. Top left: SS316L-ammonia. Bottom left: AlSi7Mg-ammonia, Bottom right: Bimetal-toluene

Ethylene glycol is not recommended in any case as comprehensive testing showed that this fluid underwent thermal decomposition/degradation at temperatures above 120°C at which its predicted thermal performance was expected to be the most attractive for two-phase heat transfer applications. Device failure occurred irrespective of the metal used.

Testing of water with ALM SS316L and Ti6Al4V were inclusive at the end of the experimental campaign as a continuous slow increase in temperature difference was observed which indicated ongoing NCG generation over time. Even after purging of this NCG, generation continued again at the same rate. Additional focused testing of these combinations, along with samples using conventional stainless steel and titanium materials, is recommended to fully understand the compatibility behaviour.

3.2. Wettability

In all cases, excellent wettability between the different fluids and ALM metals was observed, with contact angles measured to be in the range of 1°-38° Angles less than 90° are generally classified as having good wetting characteristics, with 0° indicating complete wetting.

Surfaces which have a large surface roughness are known to produce smaller contact angles compared to smooth surfaces. This is caused by the rougher peaks along the surface pinning the liquid in place and generating a taller meniscus. This was the case for the ALM samples in this study as they had measured surface roughness's 1-2 orders of magnitudes greater than those of the conventional materials.

4. CONCLUSIONS AND RECOMMENDATIONS

A range of materials, with a particular focus on those used in metal ALM, were investigated in terms of their chemical compatibility and wettability with various working fluids for use in two-phase and heat pump systems. This has lead to an update of the current state-of-the-art for recommended fluid-metal combinations for future implementation in two-phase thermal management systems.

The majority of fluid-metal combinations tested were shown to have excellent chemical compatibility, with a number of small exceptions. These included invar-methanol, bimetallic stainless steel/aluminium-toluene and all cases of ethylene glycol. Regarding spacecraft thermal control using ammonia, all tested combinations and coupling configurations were found to be compatible meaning therefore they can be implemented in future innovation designs which wish to make use of the benefits offered by ALM. Additional focused study using water as the working fluid is recommended for its compatibility with stainless steel and titanium to be fully understood for potential high temperature applications.

In addition, very small values of dynamic contact angle were observed for all ALM combinations indicating that the material surfaces exhibit the excellent wetting characteristics making them suitable for two-phase capillary pumping applications.

For both aspects of testing, the surface roughness of the ALM materials was found to play a significant role on in all areas of sample manufacture and characterisation. This included welding, sealing, crimping, cleaning and wettability. Use of ALM materials should take this into consideration when designing components where roughness may have detrimental effects on manufacturing or overall performance.

The information generated during these activities is intended to be fully disseminated in order that it is freely available to those who wish to develop two-phase heat transfer equipment using ALM. This has been achieved through presentation of all the work completed to ESA and relevant industry at the final presentation as well as during European Space Thermal Engineering Workshops (ESTEW). To further achieve this objective, different aspects of the developed methodologies and results are also intended to be published as journal articles in scientific literature.