



Investigation of the corrosive properties of phase change materials in contact with metals and plastic



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ABSTRACT

Phase change material (PCM) is used as a means of storing thermal energy. The method of PCM containment in thermal energy storage (TES) systems is important as the material is in direct contact with metal piping, plates and housing units which can be damaged if the material is corrosive. The compatibility of PCM with various materials for use in TES applications has been investigated using the immersion corrosion method and suggestions to minimise their corrosive behaviour or to mitigate it by using an alternative PCM have been presented and are described in the following paper. The metals and plastics tested were copper, brass, aluminium, stainless steel, mild steel and Perspex. These were immersed in five different PCM, three fatty acids, a salt hydrate and Micronal[®] for a period of 722 days. Stainless steel was determined to be the most suitable of all the PCM investigated. Aluminium can be used with all fatty acids, however caution is advised as the maximum corrosion rate was found to be 12.4 m g/cm²/year. Copper and brass can be encapsulated using fatty acid and again caution is advised as corrosion rates were found to be 22.15 g/cm²/year and 1.67 g/cm²/year, respectively.

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1. Introduction

Phase change materials (PCM) absorb thermal energy as latent heat at a constant phase change temperature. PCM with a suitable phase transition temperature can be used to as a means of storing thermal energy for later use [1–3] or as a method of temperature regulation of PV [4–7]. There has been considerable research on types of PCM, their applications and thermophysical properties [8–12].

PCM encapsulation can take the form of micro or macro encapsulation [13–15]. With regard to macro-encapsulation, it is important the encapsulation material in the TES system is resistant to corrosion by the PCM to ensure the system remains fully functional for its lifetime.

The availability of data on the corrosive nature of PCM was very limited until an investigation into the corrosion resistance of metallic alloys to hydrated salts was carried out where stainless steel appeared to be the most corrosion resistant and aluminium the least [16]. Further investigation into the compatibility of high phase change temperature (160 °C - 420 °C) salt eutectics with

mild, boiler and austenitic steels and the influence of water and other impurities was carried out and found LiCl-LiNO₃-NaNO₃ salt mixtures lead to surface corrosion in all steels, NaCl-NaNO₃ is compatible with all steels and KCl-LiCl are much more aggressive [17].

Cabeza et al. carried out a series of short- [18], medium- [19] and long- [20] term corrosion tests. The process involved immersing the metal sample in melted PCM in a test tube which remained in a water bath for the duration of the experiment to ensure the PCM remained in a liquid state. The short-term investigation involved leaving the metal samples immersed in the following salt hydrates; zinc nitrate hexahydrate, sodium hydrogen phosphate dodecahydrate and calcium chloride hexahydrate with melting temperatures of 36 °C, 35 °C and 32 °C respectively for a period of two weeks [18]. The zinc nitrate hexahydrate emerged as a very corrosive material as aluminium and steel were highly corroded. Calcium chloride hexahydrate produced a pitting effect in aluminium and stainless steel. The samples continued to be immersed for 75 days [19].

The metals previously mentioned were immersed in two PCMs TH29 and a chemical composition of MgCl₂·6H₂O for a period of 500 days in the long term corrosion tests [20]. As graphite can be used as a method to increase the thermal conductivity of PCM, the study immersed both the graphite and metal pieces in PCM. Aluminium and steel with either PCM are not recommended, brass

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and copper are recommended with both PCM and stainless steel is recommended for containment of PCM with caution.

Investigations into the compatibility of commercially available PlusICE E17 and Climsel C18, two salt hydrates with melting temperatures of 17 °C and 18 °C respectively, with an aluminium alloy and copper separately and aluminium in the presence of copper while immersed in an electrolyte [21]. The samples were immersed in liquid PCM for 17 h. It was found copper samples experienced greatest mass loss varying between 0.8 and 0.145 g/(m²h) for ClimSel C18 and PlusICE E17, respectively. Aluminium showed pitting corrosion at a mass loss of 7×10^{-5} g/(m²h) while in contact with ClimSel C18 with negligible mass loss when immersed in PlusICE E17. For combined samples of aluminium and copper, no mass loss occurred on the copper due to the cathodic protection but the corrosion rate increased by a factor of 8 in Climsel 18 compared to aluminium alone [21]. García-Romero, Delgado, Urresti, Martín and Sala [22] presented the corrosion behaviour of four aluminium alloys (EN AW 2024, 3003, 6063 and 1050) in the commercial salt hydrate Glaubers salt (Na₂SO₄ · 10 H₂O) which has a melting point of 32 °C. Samples were fully and partially immersed in the PCM and experiments were conducted for a period of up to 90 days. It was found the aluminium alloy 2024 was not compatible with this specific salt hydrate, however, 3003, 6063 and 1050 were shown to be fully compatible.

An investigation into the corrosion resistance of metal samples, stainless steel, carbon steel, aluminium and copper to fatty acids stearic, palmitic, myristic and lauric acid with melting temperatures between 40 °C - 63 °C [23]. Following 910 thermal cycles of being fully immersed in each PCM, the metal samples were removed and a gravimetric analysis carried out. The highest corrosion rates calculated were 3.4×10^{-2} and 4.9×10^{-2} mg/cm² for carbon steel in contact with myristic acid and copper in contact with stearic acid. Stainless steel with a chromium oxide layer and aluminium with an aluminium oxide layer were found to be compatible with fatty acids.

An investigation into the compatibility of four organic and one salt hydrate PCM (namely a molecular alloy C₁₆-C₁₈, RT20, RT26, RT25 and TH24) and plastics (high density polyethylene, low density polyethylene, polyethylene terephthalate and polypropylene) was carried out [24]. A furnace was used to cycle the plastic bottles to increase the rate of phase change. LSPE was discarded as an encapsulation material due to the mass variations and large deformations caused by the PCM. Polyethylene terephthalate and polypropylene were shown to be the best encapsulation material of the organic PCM and high density polyethylene for inorganic PCM. Polypropylene, polyethylene terephthalate and low density polyethylene are not recommended for using with TH24.

Further immersion corrosion tests have been carried out for a period of 84 days on aluminium, stainless steel, carbon steel and with salt hydrates, salt hydrate eutectics and fatty acid eutectics [25–27]. The results suggest that stainless is recommended for use with any of the PCM studied and all other metals would either be recommended with caution or not be recommended at all. Fatty acid eutectic of capric:palmitic (75.2:24.8%) show that copper is the only material tested which is not suitable for containment of the PCM. However, these tests were carried out for a short period of time and further analysis should be carried out to draw accurate conclusions. Oró, Barreneche, Farid and Cabeza [28] has also investigated the compatibility of metal and polymers with PCM with lower phase change temperatures (–22 °C to –16 °C).

There has been no research carried out on eutectic fatty acids, particularly capric, palmitic and lauric acid which have often been used in thermal energy storage systems. The phase change process has been accelerated using furnaces and thermal cycling however, due to the reduced time immersed in PCM may potentially affect

the corrosion rate. Finally, there has been no investigation into the effect of PCM on Perspex a commonly used material which can be easily manipulated. The corrosive nature of PCM in TES systems is an important factor to be considered during system design as it will impact on the lifetime of the system and thus, the economic advantages of the system.

The methodology presented involves immersing five materials, including metals and a plastic, in five different PCMs for an extended period of time. The investigation records the rate of mass loss of the metals and plastic at specified intervals over the course of the experiment. In addition to this, a series of Scanning Electron Microscopy (SEM) images were taken to assess the corrosion on a microscopic level.

As the study presented is carried out under real time conditions, it presents a more detailed account of the natural corrosion process of material samples immersed in PCM compared to accelerated methods. The paper shows the calculation of the rate of corrosion of each sample by the individual PCM. This provides an insight into the extent of the corrosive nature of PCM and can be used to ensure the optimum encapsulation material is chosen during system design. Furthermore, SEM imaging can be used for a greater insight into the initial stages of corrosion which are not initially visible to the naked eye.

2. Materials and methodology

This section describes the material samples assessed for corrosion resistance, in addition to the PCM investigated. The methodology of the experiment is presented to show the approach undertaken. Finally, a brief description of scanning electron microscopy is given as images of the samples were taken on specific days throughout the experiment to assess the level of corrosion at an increased magnification.

2.1. Material samples

Five materials that are commonly used in TES system designs and construction [5,29–31] were tested in the experiment. The metals were aluminium, brass, copper, stainless steel, steel and the plastic used was Perspex. The chemical compositions of the materials tested are presented in Table 1 using a Tescan Mira Scanning Electron Microscope. The samples were cut from larger pieces of materials and each had dimensions of 50 mm × 14 mm and a depth of 2 mm.

2.2. Phase change material (PCM) samples

Fatty acids are an organic PCM and have many superior properties including high heat capacity, congruent melting, little or no supercooling, non-toxic, good thermal and chemical stability, low cost, non-flammable and small volume change [32–34]. They are known to have high melting temperatures which are outside the range of thermal energy storage applications [35]. To overcome this, a eutectic mixture of fatty acids is two or more fatty acids combined to form a binary fatty acid with a lower phase change temperature and feature the same stability of a single component. The advantage of fatty acid eutectics is the ability to gain a lower melting temperature with all the material advantages of a fatty acid by varying the mass ratio of the mixed fatty acids [35,36]. Capric acid and two fatty acid eutectics, capric:palmitic and capric:lauric acid have been investigated. Capric:palmitic acid is a mixture of 75% capric and 25% palmitic by weight and capric:lauric acid is a mixture of 45% capric and 55% lauric acid.

Inorganic salt hydrates consist of salt and water which combine in a crystalline matrix when the material solidifies. Different salts

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