

REVIEW PAPER

Phase Change Materials: Technology Status and Potential Defence Applications

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ABSTRACT

Phase change materials (PCM) are being utilised world over for energy storage and temperature smoothing applications. Defence Laboratory Jodhpur (DLJ) has initiated a R&D programme to apply PCM in solving many heat related problems being faced by Indian forces during desert operations specially failure of mission-critical components. Under the programme, special organic PCM (Patent application no. 2258/DEL/2007 and low melting metal alloys have been developed well tuned to desert diurnal cycle. The PCM panels, when applied as an internal lining in buildings, structures and vehicles can moderate the extreme temperature within human tolerable range (below 40 °C) without the use of any external power for cooling. The panels can also act as power saver in air conditioned buildings. A cool vest has also been developed with chargeable PCM packs to provide comfortable microenvironment to a soldier on field duty (below 30 °C) for 2-3 hrs. To improve reliability of mission critical electronic instruments during desert operation, technology of absorptive PCM heat sinks is under development at DLJ. The special heat sink will absorb heat generated by component for critical mission (up to 1 hr) independent of environment temperature and thus ensure smooth functioning of critical components even in extreme hot conditions. In present paper status of PCM technology world over has been reviewed along with the brief account of research on PCM at DLJ.

Keywords: Phase change materials, PCM, microencapsulation, energy storage

1. INTRODUCTION

Phase change materials (PCM) are a group of materials which exchange large amount of heat as latent heat within a narrow temperature range of phase transformation. Since the first application of PCM by NASA in aerospace field¹, thousands of single materials and mixtures of two or more materials have been investigated for their use as PCM in areas like solar energy/waste heat storage, load shifting, and power saving², textiles³, passive-cooled shelters⁴, energy-efficient buildings^{5,6}, cooling technology for electronics⁷, transport containers for food and medicines, human comfort⁸, and energy conservation through energy storage⁹, etc.

Indian forces are working in very harsh environment in hot deserts at western border and cold deserts at high altitudes. Performance of man, equipment, and weapon system especially sensors and electronics, get adversely effected in harsh environment of desert, many times, leading to failure of critical equipments. Use of PCM can provide practical solutions to many of these problems. Defence Laboratory, Jodhpur (DLJ) has taken up a R&D programme to develop PCM-based products to meet requirements of Armed Forces. Present paper describes status of science and technology of PCM along with materials and products developed at DLJ.

2. WORKING OF PHASE CHANGE MATERIALS

Working of PCM is schematically depicted in Fig.1. When heat is supplied to PCM from outside, it absorbs a large amount

of latent heat at phase change temperature in breaking internal chemical bonds. In reverse cooling cycle, for phase reversal to start, temperature of PCM has to go down below phase change temperature (known as sub-cooling or under-cooling) to overcome the energy barrier required for nucleation of second phase. Once phase reversal starts, temperature of PCM rises (due to release of latent heat) and subsequent phase reversal takes place at phase change temperature by releasing back the latent heat to environment. Requirement of sub-cooling or under-cooling for phase reversal is an important property of PCM governing its applicability in particular applications.

Latent heat of PCM is many orders higher than the specific heat of materials. Therefore PCM can store 2-3 times more heat or cold per volume or per mass as can be stored as sensible heat

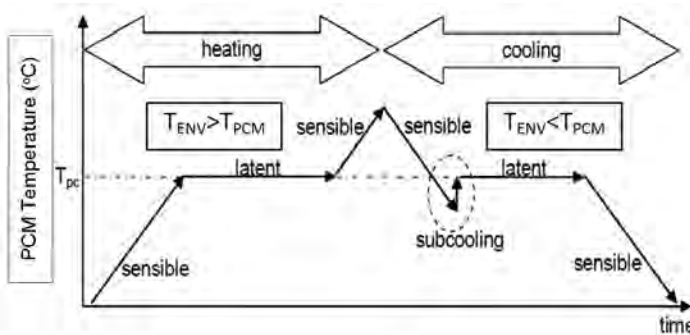


Figure 1. Schematic diagram, showing working of PCM.

in water¹⁰ in a temperature interval of 20 °C. As heat exchange takes place in narrow band of temperature the phenomenon can be used for temperature smoothening also.

Important properties of PCMs other than latent heat, phase transformation temperature and sub-cooling requirement are thermal conductivity, cyclic stability, congruent melting of PCM, and little volume change during phase change, low vapour pressure, chemical stability and compatibility of PCM with other materials like plastics¹¹. In addition to these technical requirements, safety concerns, low cost, easy availability, and good recyclability are important criteria in selecting a suitable PCM for a particular application¹²⁻¹⁴. It is difficult to get a PCM ideal for a particular application. Certain amount of tradeoff between different properties is therefore essential. Selection of a suitable PCM, determination of its quantity requirements based on heat-balance calculations, packaging of PCM, design of heat exchanging and heat distribution surfaces are other important steps in developing an effective PCM-based device or product⁸.

3. PHASE CHANGE MATERIALS

Phase change materials can be divided into two broad categories: organic and inorganic. Salt hydrates, eutectic salt mixtures, and metal eutectics are covered in the inorganic PCMs, while fatty acids, paraffins, sugars, alcohols, etc. are commonly used organic PCMs. Properties of some of the commonly used PCM are summarised in Table 1.

For applications around 0 °C, the best PCM is water. For temperature below 0 °C, eutectic water salt solutions are used. Salts (> 150 °C) and salt hydrates (5 °C and 130 °C) are used for waste heat and solar energy storage due to their high storage energy densities. The biggest problem with salt hydrates is their phase separation or their incongruent melting with repetitive cycles. This leads to degradation in heat-storing properties and limits the useful life of PCM. Another important problem in hydrated salt type PCMs is very high degree of sub-cooling required for freezing. The sub-cooling problem has been solved to some extent by mixing additives called heterogeneous nucleator. Gelling or thickening is generally used to overcome problem of incongruent melting^{15,16}.

Low-fusion metal alloys (eutectic compositions of *Bi-Pb-In-Cd-Sn-Zn* multicomponent system) are also used as PCM

due to their good thermal conductivity (> 0.5 Watt meter/°K), favourable melting temperature range in 45-120 °C with high heat of fusion per unit volume (250-300 kJ/kg) in applications where space is a constraint like absorptive heat sinks for electronic components^{7,17}.

Organic materials like paraffins, fatty acids^{18,19} are another class of PCM with density less than that of water. These are congruent in melting and chemically stable. They have high specific heat of fusion by mass (200-250 kJ/kg) but very low by volume (125-175 kJ/cc). These generally shows very little tendency to super cooling. These are compatible to all metal containers; however with plastic containers, paraffins have a tendency to infiltrate and soften some plastics. On disadvantage side organic PCMs have low thermal conductivity and a high volume change between the solid and the liquid stages. These cause many problems in container design²⁰. Commercial paraffin does not have sharp well defined melting points and also has comparatively low heat of fusion (50-60 J/g).

Apart from these, new class of materials like sugar alcohols, polyethylene glycol (PEG), poly alcohols, clathrates etc have been tried as PCM in last decade^{21, 22}.

Research in PCM has also been focused to get materials with tailored melting point (fine-tuned to a particular application) or improved properties by mixing or alloying of two or more materials, specially eutectic compositions²³⁻³⁰. A new material has been reported³¹ where water as a PCM is integrated into a three-dimensional network of polyacrylamide.

4. MICROENCAPSULATION OF PHASE CHANGE MATERIALS

Microencapsulation of PCM core of 1 µm to 1000 µm diameter with a shell of 2-3 µm wall thickness of normal plastic or polymer is an important process to contain liquid PCM from flowing out during phase transformation. Chemical processes for *in-situ* encapsulations like complex co-acervation with gelatin and interfacial polycondensation to get a polyamide or polyurethane shell, precipitation due to polycondensation of amino resins, etc have been used to get microcapsules of desired size and cyclic stability³²⁻³⁶. PCM microcapsules are being used to develop textiles with higher thermal mass³ and PCM slurries with high heat storage and transfer properties^{37, 38}.

Table 1. Comparison of properties of commonly used PCMs^{8,10}

Material type	Type of transition	Phase change temp range (°C)	Latent heat (J/cc)	Density (g/cc)	Examples
Organic compounds	Solid-liquid (wet)	-12 to +187	125-400	0.75-1.54	Paraffins, fatty acids, alcohols, sugars, etc
Inorganic: salt hydrates	Solid-liquid (wet)	20-140	270-650	1.5-2.2	$CaCl_2 \cdot 6H_2O$, $Ca(NO_3)_2 \cdot 4H_2O$, $MgSO_4 \cdot 7H_2O$ etc
Inorganic salt	Solid-liquid (wet)	< 150	200- 500	1.7-2.5	$LiNO_3$, $NaNO_3$, $MgCl_2$, K_2CO_3
Inorganic salt solutions	Solid-liquid (wet)	< 0	130- 330	Around 1	KCl 19.5 % + H_2O
Solid-solid organic compounds	Solid-solid (dry)	21-100	140-200	1-1.1	TCC
Metal eutectics	Solid-liquid (wet)	30-125	200-800	6-10	Eutectics of <i>Bi-Pb-Cd-Sn-In</i>
Micro-encap. PCM	Solid-liquid (dry)	6-101	95-186	0.9- 1.1	Micro encap. paraffin, fatty acid

5. PHASE CHANGE MATERIALS COMPOSITES

To overcome problem of leakages of liquid PCM, an alternate route has been taken making its composite with high-density polyethylene^{39,40} and by absorbing it in porous materials like ceramic granuals, tiles and wood fibre board. In composite, PCM is finely distributed in polyethylene/ other porous matrix, which restricts flow of liquid PCM, and therefore can be cut in different shapes easily without any leakages.

Composite making is also being tried to solve the problem of poor thermal conductivity of PCM by mixing it with high conductivity materials like graphite⁴¹⁻⁴⁹. In another approach⁵⁰, a sufficient increase in thermal conductivity has been seen up to 6 W/mK by putting PCM inside metallic foam with 94 per cent porosity.

6. COMMERCIAL PHASE CHANGE MATERIALS PRODUCTS

Many international companies like BASF, Climator, Cristopia, EPS Ltd., Mitsubishi Chemical Corporation, Rubitherm GmbH, TEAP, Witco., etc are marketing PCMs and PCM products. In India also. few companies are marketing their own or licensed PCM products.

Currently more than 50 PCMs are commercially available. Most commercial PCMs are based on modified compositions of salt hydrates, paraffins and eutectic salt water solutions, with agents for nucleating, gelling, and thickening added in the base. Apart from micro- or macro-encapsulated PCMs, these companies are also marketing different PCM products like PCM wallboards, PCM-polymer or PCM-silica dry composite powders by Rubitherm. Recently Dupont Energain panels containing a copolymer and paraffin compound has been launched for building material. Garmisch Partenkirchen, Germany, has introduced pocket heaters for mountain-rescue teams. M/S Climator AB is marketing Cool vest to provide 28 °C temperatures around human body for 3 h. An American company Outlast Thermocules, is marketing fibre- and fabric-containing microencapsulated PCMs. A different application of PCM is in ballistic vests produced by Outlast which protect people from gunshot. Other products for human comfort are underwears to reduce sweating, gloves, shoes, jackets, kidney belts, sleeping bags, etc. Various types of transportation containers to carry medicines and food items to field conditions are being marketed by various companies. PCM products for heat therapy are being marketed by Lavatherm GmbH. A PCM jacket to protect battery from extreme climatic conditions is being marketed by TEAP together with Power Conversion Products and MJM Engineering.

7. POTENTIAL DEFENCE APPLICATIONS

Today's advance war equipments, be these weapon systems, surveillance radars, communication systems, aerospace systems, missiles, etc, use heat-sensitive electronics, microprocessors, sensors, etc. The heat

management in these devices (specially imported ones) gets disturb in extreme Indian climatic conditions (day temperature in summer may be as high as 50 °C) causing malfunctioning or even failure. As latent heat is almost independent of environmental temperature, it can help in maintaining reliability of critical components in extreme hot conditions during short duration or cyclic operations of mission by acting as absorptive heat sink. In literature, PCM has been used as blanket around batteries by absorbing peak or cyclic loads⁴², thus improving their performance, reliability, and life. Microencapsulated PCM has been used to slow down rise in source Temperature in electronics up to 30 min^{17,51}.

PCM-based electronic cooling devices in aerospace and defence are being used successfully by government agencies like NASA or defence industry of advance countries. However these devices are not available off-the-self commercially and design information is a closely-guarded trade secret.

8. PHASE CHANGE MATERIALS RESEARCH AT DL, JODHPUR

Seeing the possible applications of PCM devices in improving reliability of mission-critical defence components during operation in hot climate specially in a desert, an R&D programme for developing PCM materials and devices suitable for defence applications has been initiated at DL, Jodhpur.

In this programme DLJ has developed various low- fusion metal-eutectic alloys of *Pb-Bi-Sn-In-Cd* systems with melting point tunable between 46 °C–120 °C, density 9.5–10.5 g/ml, latent heat between 20–25 J/g. Figure 2 shows differential scanning calorimetric curve of one such alloy having melting rang 57-59 °C. A high capacity heat sink has been developed by filling metal eutectic PCM in aluminum cavity (shown in Fig. 3) for better heat management in electronics. The performance of PCM heat sinks has been tested and compared with reference aluminum sink at different environmental temperatures ranging from 40 °C to 55 °C in a simulated test set up. The performance data at extreme temperature (55 °C) are shown in Fig. 4. PCM heat sink slow down temperature

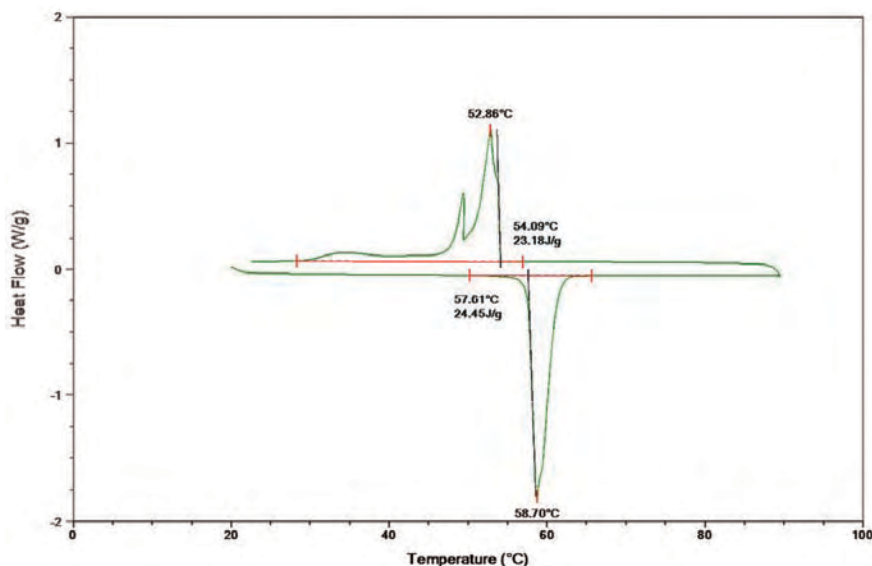


Figure 2. DSC curve for metal-eutectic composition developed at DL, Jodhpur.

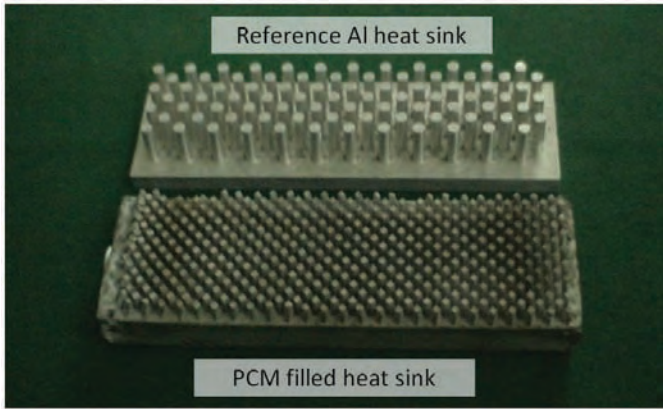


Figure 3. PCM-based absorption heat sink for 20 W SSPA under development at DL, Jodhpur.

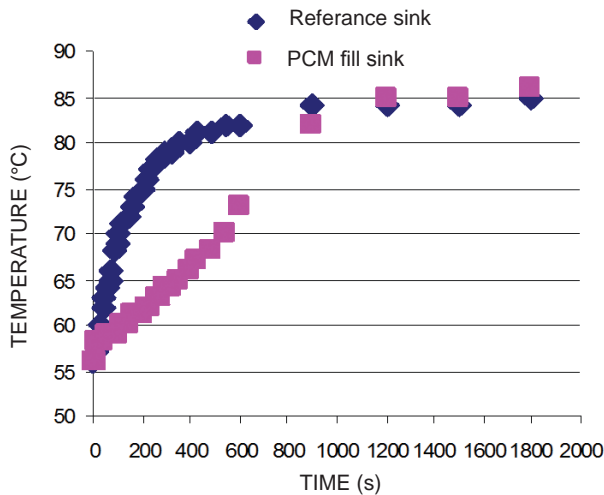


Figure 4. Comparison of performance of PCM heat sink with normal Al heat sink at 55 °C environment temperature.

rise of heating element simulating heat generating electronic component by 10 °C to 15 °C for 10-15 min. Thus PCM heat sink can improve performance and reduce failure probability of sensitive components in cyclic operations.

Working efficiency of soldiers also gets adversely affected by extreme heat of desert or extreme cold of high altitude areas. PCM by absorbing and releasing large quantity of latent heat in narrow temperature band of phase transformation can help in moderating extreme temperatures.

Commercial PCMs show poor phase-reversal and stability problems in harsh desert climates. DLJ has developed a special PCM (Patent application no. 2258/DEL/2007 dt 5th Dec 2007) tuned to extreme hot desert climate. The material melts during daytime by absorbing a large amount of heat as environmental temperature goes above human body temperature and automatically resolidifies during night automatically by releasing heat back to environment.

The material filled in panels (Fig. 5) has been used as internal lining in a prototype cabin of size 240 cm x 120 cm x 180 cm. The internal temperature of cabin was monitored during last three summers. It was observed that even in extreme summer, internal temperature of cabin did not go

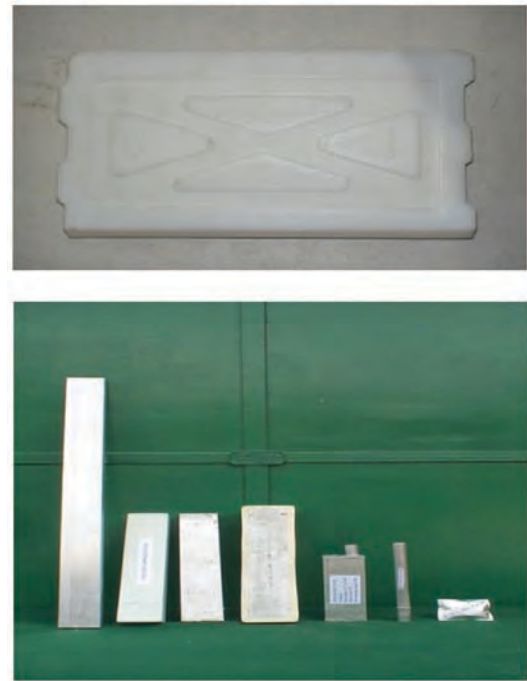


Figure 5. PCM panel developed at DLJ.

beyond 40 °C (Fig. 6), thus providing a relief of 8-15 °C from unlined cabins²³. In addition to its normal function of heat absorption and temperature moderation, the special PCM, being a hydrogenous material, is a good neutron absorber also. Thus PCM panel, if applied inside armoured personnel carrier (APCs), nuclear-hardened bunkers and structures, can provide shielding against both heat as well as radiation pulse generated during nuclear blasts along with its day-to-day functioning of temperature moderation during summer⁵².

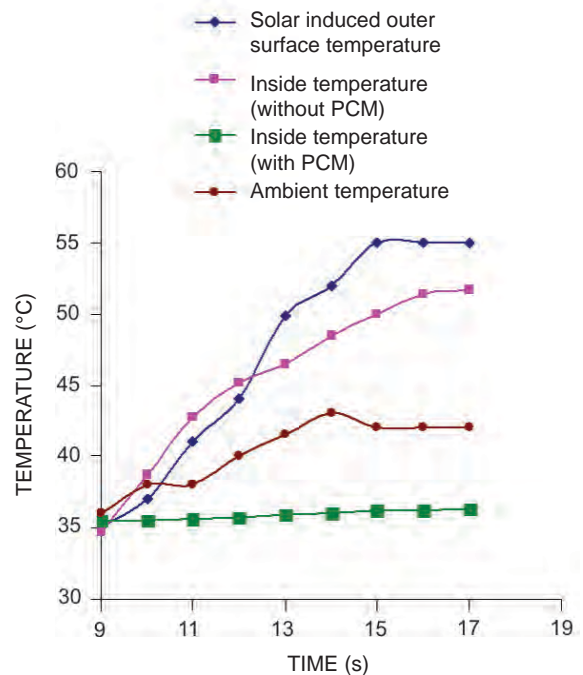


Figure 6. Performance of PCM lined heat efficient cabin at DL Jodhpur.

Other products, PCM-based cool vests and caps, having removable PCMs packs in multiple pockets, have been developed at DLJ for soldiers on field duties. The weight of the vest is 1.5 kg to 2.0 kg and PCM packs needs to be charged in a refrigerator or deep fridge before each application. In a field evaluation inside engine cabin of APC (Russian model BMP) the vest provides less than 30 °C temperature around human torso for more than 2 h (Table 2). During a user trial with Border Security Force (BSF), in May 2011, the cool vest was proved extremely useful for reducing heat stresses of Jawans positioned on Border Observation Posts (BOPs) along Indo- Pak border. Further, soldiers wearing nuclear biological and chemical individual protective gear (NBC-IPG) suits feel highly suffocated and dehydrated due to heat. PCM cool vest underneath NBC-IPG, has reduced heat stress drastically during physiological evaluation at DIPAS, Delhi.

Table 2. Performance of cool vest in BMP engine cabin

Time (h)	Temp of PCM pad (° C)	Temp in the vicinity of human body (° C)	Temp inside BMP driver cabin (° C)
2.30	10.2	20.2	41.0
2.45	10.9	23.1	41.5
3.0	11.5	24.9	41.7
3.15	12.7	25.4	44.1
3.30	12.8	26.8	44.7
3.45	12.9	27.1	45.2
4.00	12.9	27.8	46.5
4.15	13.00	27.9	46.9
4.30	13.00	28.0	47.9

Waste energy/solar energy utilisation to partially meet Army's vast energy requirement is a major concern. In a recent communication from Army Technology Board, non-conventional means like PCM can play an important role in realising this. DLJ is working on this important futuristic technology of solar energy well. As a first step in this direction technology for micro-encapsulation of PCM and PCM slurries (up to 50 % PCM by weight) having 2-3 times more heat-storing capacity than water in 30-50 °C temperature range have been developed.

9. CONCLUSIONS

It is clear from the above discussions that science and technology of PCMs is well developed internationally and PCM products are being increasingly used in various areas for energy storage and temperature smoothening. In defence also, especially in Indian context, PCM can be very useful in overcoming extreme heat/cold-related problems faced by soldiers and equipments alike.

DLJ is pursuing R&D programme to develop expertise in PCM science and technology. Various products for defence applications have been developed or are under advanced stages of development. The special phase-change material developed by DLJ shows minimal under cooling requirements for phase-reversal, and hence, complete its melting-solidification cycle

passively in tune with extreme diurnal cycles observed during hottest summer seasons in deserts. The material also shows excellent cyclic stability and does not degrade with time. In a prototype cabin, the internal lining of DLJ-developed PCM panels was able to maintain internal temperature of cabin below 40 °C passively during last three summer seasons. Efforts are on to apply the technology to develop energy-efficient field shelters for Armed Forces and paramilitary organization like BSF. Other product, PCM-based cool jacket and caps developed by DLJ have successfully completed user trials by BSF and are being induced. Army has also found the product useful for peacetime operations and for training purposes. Further R&D is being carried out to diversify applications of PCMs to address problems of extreme climates being faced by Armed Forces like hot jackets for high altitude areas and high capacity heat sinks for critical instruments.

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