

**OPTIMISATION OF THERMAL
CONDUCTIVITY OF PHASE CHANGE
MATERIALS BY USING TAGUCHI METHOD**

MOHD FAZRY BIN ABDUL AZIZ

MASTER OF MECHANICAL ENGINEERING

**UNIVERSITI PERTAHANAN NASIONAL
MALAYSIA**

2020

**OPTIMISATION OF THERMAL CONDUCTIVITY OF PHASE CHANGE
MATERIALS BY USING TAGUCHI METHOD**

MOHD FAZRY BIN ABDUL AZIZ

Thesis submitted to the Centre for Graduate Studies, Universiti Pertahanan Nasional
Malaysia, in fulfilment of the requirements for the Degree of Master of
Mechanical Engineering.

November 2020

ABSTRACT

To date, although research on phase change materials (PCM) are still widely carried out, study on different type of phase change materials system is yet to be performed due to the complexity of mixture properties. Thus, right parameters are vital to generate a product with high quality especially with balanced thermal properties. In this study, a comprehensive approach known as Design of Experimental (DoE) has been applied efficiently on relevant stages, starting from the material formulation and processing stage until the material characterisation stage. Few control factors directly affecting thermal conductivity were chosen namely types of PCM wax, percentage weight of graphene, mixing time and speed. The optimised composition was obtained through verification experiment whereby the thermal conductivity had increased by as much as 9.4 % compared to pure paraffin. From the Analysis of Mean (ANOM) and Analysis of Variance (ANOVA), weight of graphene and types of wax had the most significant effects for the thermal conductivity with contribution percentages of 60% and 31% respectively. Results shown thermal conductivity increase proportionally with the percentage weight of graphene. Optimum results are obtained when the type of PCM is paraffin wax and percentage weight of graphene is 5wt%, while the mixing time and speed does not show any significant effect on enhancing thermal conductivity. Furthermore, melting point does not show any significant changes. However, there is a decreasing in latent heat compared to the pure PCMs. The experimental optimum result of thermal conductivity performed better than the predicted optimum results while in comparison the addition of silicone oil gives a better latent heat compared to the

addition of mineral oil. This research has shown that the addition of graphene will enhance the thermal conductivity and modify the thermal properties of the PCMs.

ABSTRAK

Sehingga kini, walaupun kajian mengenai bahan perubahan fasa (PCM) masih banyak dilakukan, kajian mengenai pelbagai jenis sistem bahan perubahan fasa masih belum dapat disimpulkan kerana kerumitan sifat campuran. Parameter yang tepat sangat penting untuk menghasilkan produk berkualiti tinggi terutamanya dengan sifat terma yang seimbang. Dalam kajian ini, pendekatan komprehensif dikenali rekabentuk eksperimental (DoE) dilaksanakan mulai dari tahap perumusan dan pemrosesan bahan hingga pencirian bahan. Beberapa faktor kawalan yang mempengaruhi kekonduksian terma dipilih iaitu jenis lilin, peratusan berat GR, masa dan kelajuan pencampuran. Komposisi optimum diperoleh melalui verifikasi eksperimental, kekonduksian terma telah meningkat sebanyak 9.4% berbanding dengan parafin tulen. Dari Analisis Min (ANOM) dan Analisis Varians (ANOVA), peratusan berat GR dan jenis lilin memberi kesan yang paling signifikan dengan peratusan 60% dan 31%. Hasil kekonduksian terma meningkat secara berkadar dengan peratusan berat GR. Hasil optimum yang diperoleh adalah jenis PCM daripada lilin parafin dan berat peratusan GR adalah 5 wt% sementara masa dan kelajuan pencampuran tidak menunjukkan kesan yang signifikan terhadap peningkatan kekonduksian terma. Titik lebur tidak menunjukkan perubahan yang ketara. Walau bagaimanapun, terdapat penurunan haba pendam berbanding PCM tulen. Hasil optimum eksperimen, kekonduksian terma menunjukkan prestasi yang lebih baik daripada hasil optimum yang diramalkan sementara sebagai perbandingan, penambahan minyak silikon memberi haba pendam yang lebih baik berbanding dengan penambahan minyak mineral. Penyelidikan ini menunjukkan penambahan GR akan meningkatkan kekonduksian terma dan mengubah sifat terma PCM.

ACKNOWLEDGEMENTS

I would like to express my sincere gratitude to the Ministry of Defence, Malaysia for endorsement of my study and financing through the Higher Education Scheme. I want to thank National Defence University of Malaysia, Kuala Lumpur for providing the facilities and materials required to generate and finish my dissertation. To my supervisor, Dr. Ku Zarina binti Ku Ahmad, I would like to convey my sincere and heartfelt appreciation for her ongoing assistance at every stage of my study and research, for her guidance, motivation, patience and enormous understanding. Thank you for the advice and time sharing that has assisted me throughout my studies and thesis writing. I am deeply grateful to my fellow laboratory technician, in particular Mr. Zazlin Bin Ismail who volunteered to assist me in preparing of materials for laboratory experiments. Last but not least, I would like to thank my wife Masita binti Mohd Bari for her unwavering support and belief in me, as well as my children Muhammad Rayyan Nufail, Muhammad Raqib Ukail and Mohd Raef Mikail, who have always been there with excellent love and patience and who have been my personal "moral booster" to work on this thesis.

APPROVAL

The Examination Committee has met on **30th October 2020** to conduct the final examination of **Mohd Fazry bin Abdul Aziz** on his degree thesis entitled **‘Optimisation of Thermal Conductivity of Phase Change Materials by Using Taguchi Method’**.

The committee recommends that the student be awarded the Master of Mechanical Engineering.

Members of the Examination Committee were as follows.

Dr. Norwazan Binti Abdul Rahim

Faculty of Engineering

Universiti Pertahanan Nasional Malaysia

(Internal Examiner)

Dr. Raja Nor Izawati binti Raja Othman

Faculty of Engineering

Universiti Pertahanan Nasional Malaysia

(Internal Examiner)

APPROVAL

This thesis was submitted to the Senate of Universiti Pertahanan Nasional Malaysia and has been accepted as fulfilment of the requirements for the degree of **Master of Mechanical Engineering**. The members of the Supervisory Committee were as follows.

Dr. Ku Zarina Ku Ahmad

Faculty of Engineering

Universiti Pertahanan Nasional Malaysia

(Main Supervisor)

UNIVERSITI PERTAHANAN NASIONAL MALAYSIA

DECLARATION OF THESIS

Student's full name : MOHD FAZRY BIN ABDUL AZIZ
Date of birth : 5 NOVEMBER 1982
Title : OPTIMISATION OF THERMAL CONDUCTIVITY OF PHASE CHANGE MATERIALS BY USING TAGUCHI METHOD
Academic session : 2019/2020

I hereby declare that the work in this thesis is my own except for quotations and summaries which have been duly acknowledged.

I further declare that this thesis is classified as:

- CONFIDENTIAL** (Contains confidential information under the official Secret Act 1972)*
- RESTRICTED** (Contains restricted information as specified by the organisation where research was done)*
- OPEN ACCESS** I agree that my thesis to be published as online open access (full text)

I acknowledge that Universiti Pertahanan Nasional Malaysia reserves the right as follows.

1. The thesis is the property of Universiti Pertahanan Nasional Malaysia.
2. The library of Universiti Pertahanan Nasional Malaysia has the right to make copies for the purpose of research only.
3. The library has the right to make copies of the thesis for academic exchange.

Signature

**Signature of Supervisor/Dean of CGS/
Chief Librarian

IC/Passport No.

**Name of Supervisor/Dean of CGS/
Chief Librarian

Date:

Date:

Note: *If the thesis is CONFIDENTIAL OR RESTRICTED, please attach the letter from the organisation stating the period and reasons for confidentiality and restriction.

TABLE OF CONTENTS

	Page
ABSTRACT	ii
ABSTRAK	iv
ACKNOWLEDGEMENTS	v
APPROVAL	vii
DECLARATION	viii
TABLE OF CONTENTS	ix
LIST OF TABLES	xi
LIST OF FIGURES	xii
LIST OF ABBREVIATIONS	xiii
CHAPTER	
1	INTRODUCTION 1
	1.1 Background 1
	1.2 Problem Statement 3
	1.3 Objectives 4
	1.4 Scope and Limitation 5
	1.5 Importance of Study 6
2	LITERATURE REVIEW 7
	2.1 Introduction and Overview 7
	2.2 Thermal Energy Storage 9
	2.2.1 Sensible Heat Thermal Energy Storage 10
	2.2.2 Latent Heat Thermal Energy Storage 10
	2.3 Phase Change Material 11
	2.3.1 Paraffin Wax 14
	2.3.2 Non-Paraffin 16
	2.3.2.1 Beeswax 17
	2.3.2.2 Carnauba Wax 17
	2.4 Additives 18
	2.5 Surfactant 19
	2.6 Application of Phase Change Material 19
	2.7 Previous Studies/Researches About PCMs with Graphene (GR) as Additives 22
	2.8 Table of Literature/Research Gap 25
	2.9 Taguchi Method 27
3	METHODOLOGY/MATERIALS AND METHODS 29
	3.1 Flow Chart of Research 31
	3.2 Materials 32

	3.2.1 Phase Change Materials	32
	3.2.2 Graphene Nanoplatelets	33
	3.2.1 Surfactant	34
	3.2.2 Mineral and Silicone Oil	35
	3.3 Method and Experimental Setup	36
	3.3.1 Design of Experiment	37
	3.3.2 PCMs Fabrication Process	39
	3.4 PCM Characterization Methods	41
	3.4.1 Thermal Conductivity Analysis	42
	3.4.2 Thermal Characterisation Analysis	43
4	RESULTS AND DISCUSSION	45
	4.1 Introduction and Overview	45
	4.2 Orthogonal Array	46
	4.3 Thermal Conductivity Analysis	47
	4.3.1 Determination of Thermal Conductivity	47
	4.3.2 ANOVA and ANOM	48
	4.3.3 Determining the Optimum Levels	50
	4.4 Characterisation of Phase Change Materials	52
	4.5 Analysis of Optimum Sample	56
	4.5.1 Determination of Optimum Sample PCM Thermal Conductivity	57
	4.5.2 Characterisation of Optimum Sample PCM	57
5	CONCLUSIONS AND RECOMMENDATIONS	61
	5.1 Introduction and Overview	61
	5.2 Conclusions	61
	5.3 Recommendations	63
	REFERENCES	65

LIST OF TABLES

TABLE NO.	TITLE	PAGE
2.1	Criteria for ideal phase change material	11
2.2	Comparisons classification of PCMs	13
2.3	Commercial paraffin waxes with their thermal properties	15
2.4	Specification of commercial grades of carnauba wax	18
2.5	Comparison of previous studies on PCM and graphene as additive	26
3.1	Properties of graphene nanoplatelets	34
3.2	Original template of L9 orthogonal array	37
3.3	Actual weight for material, additive, mixing time and speed	38
4.1	Level of process parameters	46
4.2	Thermal conductivity of phase change materials	48
4.3	Contribution percentage of control factors	50
4.4	Thermal properties of experiment PCMs	55
4.5	Thermal properties of pure PCMs.	56
4.6	Thermal conductivity result for optimum sample	57
4.7	Latent heat and melting temperature comparison of optimum sample with and without oil	59

LIST OF FIGURES

FIGURE NO.	TITLE	PAGE
2.1	Different types of thermal energy storage	9
2.2	Classification of phase change materials	12
2.3	Non-paraffin properties	16
2.4	Solar phase changes thermal storage integrated device	20
2.5	PCM heat sink schematic	21
2.6	Taguchi method algorithm	28
3.1	Flow chart of the experiment	31
3.2	Paraffin wax block	33
3.3	Beeswax block	33
3.4	Carnauba wax Grade T4	33
3.5	Graphene nanoplatelets powder	34
3.6	Sodium Dodecyl Benzenes Sulfonate powder	35
3.7	PVC pipe with fully fabricated mould	39
3.8	Experimental setup during mixing process	40
3.9	Analytical balance	41
3.10	K2D Pro device setup for thermal conductivity analysis	43
3.11	Simultaneous Thermal Analyser (STA 8000)	43
4.1	Pareto percentage of factor effects of control factors	50
4.2	S/N chart for factor effect of thermal conductivity	52
4.3	Graph of heat flow endo-up versus temperature of experiments	55
4.4	Graph of heat flow endo-up versus temperature of pure PCMs	56
4.5	Graph of heat flow endo-up versus temperature of optimum sample	58

LIST OF ABBREVIATIONS

PCMs	Phase Change Materials
TES	Thermal Energy Storage
LHTES	Latent Heat Thermal Energy Storage
DOE	Design of Experiment
SDBS	Sodium Dodecyl Benzenes Sulfonate
PV	Photovoltaic
EP	Expanded Perlite
xGNP	Exfoliated Graphene Nanoplatelets
GR	Graphene
EG	Graphite
GO	Graphene Oxide
STA	Simultaneous Thermal Analyzer
UPNM	Universiti Pertahanan Nasional Malaysia

CHAPTER 1

INTRODUCTION

1.1 Background

In line with increasing population and ever-changing lifestyle, demand of energy has steadily increased throughout this new era. Generally, energy resource is a necessity for almost all sectors including transportations, manufacturing, industrial and residential, and have been significantly utilised in our daily lives. The primary sources of energy are mainly fossil-based energy such as oil, natural gas and coal which are on the top list of high demand resources. Based on statistics from International Energy Agency 2019, primary energy consumption rose by 1.8% in 2018 relative to 2017 levels of consumption. More than 123 million barrels per day of fossil-based energy are expected to be produced by 2025 (Imtiaz Hussain et al.,2018). Hence, the increasing demand of fossil-based energy from time to time basis leads to excessive use of it and will soon be depleted. Thus, this has contributed to the increase in green house emission and pollution (Zecca & Chiari., 2010).

However, an alternative source of a renewable energy has been discovered and currently been developed and grown to produce energy from natural sources to

ensure that the energy will be constantly replenished. This innovation of the renewable energy brings down the costs and looks promising as a clean energy in future. This means that renewable energy is increasingly displacing fossil fuels while offering benefits of lower emissions of carbon and other types of pollution. In term of renewable energy, phase change materials (PCMs) have been identified and developed as an effective Latent Heat Thermal Energy Storage (LHTES) material. It plays a significant role in improvement of energies efficiency.

LHTES techniques is particularly popular because of its high energy storage capacity and unique abilities to hold heat at constant temperature with related PCM compared to the conventional sensible heat storage (Sharma et al.,2007). The effective way to store solar thermal energy is by using PCM (Mofijur et al.,2019). The solar enables excess energy to store in storage medium and provides it whenever required. LTHES uses the concept of phase transition temperature of PCM. These materials absorb or release latent heat while undergoing phase change process such as melting and boiling. During melting process, heat is transferred into the material and it stores heat energy at constant temperature. Next, the heat is released and solidified the material. The most suitable state of PCM for storing thermal energy is solid liquid state. It can store 5 to 14 times heat energy higher compared to sensible heat storage materials (Boda et al.,2017).

Various types of PCM have been investigated by many researchers for their suitability especially in enhancement of their thermal properties. Naturally, almost all pure PCMs have a low thermal conductivity and substantially decrease their efficiency in term in energy storage and release (Sriharan & Harikrishnan., 2017). In

order to overcome this limitation, combining the PCM with the high thermal conductivity matrix acts as an additive, which is one of the methods to improve the thermal conductivity of PCM (Marcos et al.,2017).

1.2 Problem Statement

Globally, dependency on fossil fuels as a primary energy source continues to rise and this non-renewable energy can no longer bear the amount of demand. Mofijur et al.,2019 noted that the rapid rise in energy usage, continuous increase of fuel cost and greenhouse gas emissions seem to be the major forces for more reliable use of renewable sources of energy. In order to reduce the reliance on fossil fuels, alternative exploitations on energy consumptions have emerged which is renewable energy. Renewable sources of energy such as sun, wind and wave are unlimited resources and expected to fulfill the demands of the growing population today. Thermal energy storage (TES) is among the most major breakthroughs for energy savings (Rawi et al.,2018). Thermal energy storage is defined as the temporary holding of thermal energy for later utilisation. By using PCM as one of the methods to manage energy storage, it is considered to be ideal products for thermal management solutions.

However, due to the climatic changes, the means of storing these types of renewable energy has become urgent. This has resulted in the important use of phase change material (PCM) for developing new efficient and sustainable energy saving methods. There are many types of materials which can be used as PCM. As a matter of fact, identifying the suitable PCM for exact application is an area which needs further research due to different thermal properties of PCM. Nevertheless, pure PCM

thermal properties could still pose as an obstacle since the thermal conductivity is low. But the improvement of thermal properties of PCM composite can be done through incorporating with a high thermal conductivity additive compared to PCM alone. A research needs to be done to find the perfect composition of PCM and additives.

1.3 Objectives

This study embarks on the following objectives:

- a) To investigate the performance of thermal conductivity and thermal properties of PCM with the addition of graphene.
- b) To access the optimum control factor of composite phase change materials in terms of thermal conductivity using Taguchi Method.
- c) To compare the thermal performance properties by incorporating silicone oil and mineral oil into the optimum composition of PCM with and without graphene.

1.4 Scope of Work and Limitation

This research focuses mainly on achieving the outcomes of thermal properties enhancement with addition of nanoparticles which is graphene. Design of Experiment (DOE) will be used in this experiment by constructing the L9 orthogonal array template. Collected information will be inserted in the template. PCMs composite will be fabricated according to the template. Total mass for mixed composite is 70 grams. Thermal conductivity of composite PCMs is measured using KD2 Pro device in unit W/m.K. The characterisation such as melting and solidification temperature and latent heat are analysed and investigated by using Simultaneous Thermal Analyser (STA). 3 types of main PCMs (paraffin wax, beeswax and carnauba wax) with different melting point are used in this research to enhance its thermal properties by incorporating graphene (GR) nanoparticles as an additive into PCM. The percentage weight of GR nanoparticles is 0.05%, 1% and 5%. Different mixing time of 15 minute, 30 minute and 60 minute will be executed with the addition of graphene to obtain homogenous mixture. Next, different mixing speed rate of 100 rpm, 300 rpm and 500 rpm will be used to mix the composite PCM with the addition of the additive. Surfactant such as Sodium Dodecyl Benzenes Sulfonate (SDBS) is used to reduce surface tension of composite. The ratio of surfactant to additive is 0.1 with the mixing time of 15 minutes and speed of 300 rpm which is constant for all experiments during the mixing between PCM with the addition of the surfactant. PVC pipe with dimension of 27mm, 22mm and 127mm for outer diameter, inlet diameter and height respectively are connected with a stopper at one end to be used as the moulds for the composite PCM and capable to insert the TR-1 sensor due to the length of the sensor

is 100mm. A hot plate with temperature between 55 to 100 °C were put underneath the beaker with PCM during fabrication process to prevent it from being solidified.

1.5 Importance of Study

This study will focus on optimisation and characterisation of thermal properties enhancement of PCM through the addition of graphene. Instead of paraffin, PCM made from vegetable sources (carnauba wax) and animal sources (beeswax) will be studied in term of melting and solidification temperature, latent heat and a thermal conductivity. It is believed that non-paraffin organic based PCM such as vegetable and animal can be beneficial due to its being eco-friendly and renewable as compared to petroleum derived paraffin whereby the fossil-based energy may increase the risk of pollution that can cause harmful effects on living things and climate change. Moreover, the results from this study may be applicable in future in the relevant fields suitable for their performance and thermal properties of the composite PCM. Furthermore, the future researcher will be able to compare the different thermal properties resulted in different type of PCM and additives.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction and Overview

Due to huge demands on energy supply, carbon fossil energy has been playing a major role during the past years in supplying various types of energy all across the world. However, fossil fuels tend to have certain disadvantages to the environment. The degradation and the rising reliance on fossil fuels due to continuous urban and industrial development made us aware of the importance to focus on the utilisation of sustainable energy sources which are readily accessible and naturally available. The UN Intergovernmental Panel on Climate Change (UNIPCC) reported that global average temperature rises at around 0.6 K and expected to continue rising at 1.4 - 4.5 K by year 2100 (Kim and Ferreira.,2007). Among the types of energy storage, thermal energy storage (TES) is one of the most relevant. This is especially due to the latent heat thermal energy storage (LHTES) mechanism whereby a higher energy density is stored in a narrower operating temperature range by applying phase change material (PCM). In an attempt to minimise the usage and to conserve this type of energy, phase change materials is introduced as the next Latent Heat Thermal Energy Storage (LHTES).

There are various kinds of PCMs. It is widely used in numerous applications, to name a few, energy management in buildings, food storage and solar energy system in order to improve the overall performance. Guo and Zhang., 2007 stated that PCMs are the best common materials in term of high thermal absorption capacity and non-hazardous. This material is more effective in term of absorption and its capability to release heat compared to conventional (sensible) storage ranging from 5 – 14 times temperature capacity in a small temperature gap (Sharma et al.,2007). Eventually, latent heat storage abilities of PCM are the main properties that need to be measured (Navarro et al.,2010). However, their limitation of lower thermal conductivity in PCM represents the key disadvantages which may increase thermal resistance during the transformation process as the liquid-solid interface shifts away from the surface during heat transfer. But with recent pioneering emerging technologies in nanotechnology, the technical issues are being resolved and capable to disperse nanometrically highly conductive particles in PCM.

2.2 Thermal Energy Storage (TES)

There are various classification of energy storage available and thermal energy storage (TES) is one of them. Thermal energy is a valuable source of energy, commonly available in environmental conditions and a by-product of many applications by using energy conversion. It is promising due to their availability for recycle of abundant thermal energy and energy efficiency improvement (Zhang et al.,2018). The ability of a material to absorb thermal energy can be achieved by cooling, heating, melting, solidifying or vaporising and the energy obtainable as heat materialise during the reversed cycles or process (Pielichowska and Pielichowski., 2014). The different changes in interval energy of a material for TES storage can be in the forms of sensible heat, latent heat and thermochemical or combination of these. Among all forms, latent heat thermal energy storage (LHTES) exhibits a high energy storage capacity and a small variation in operating temperature due to the phase transition of phase change materials (PCMs) at the melting point. Figure 2.1 shows major techniques of thermal energy storage (Sharma et al.,2007).

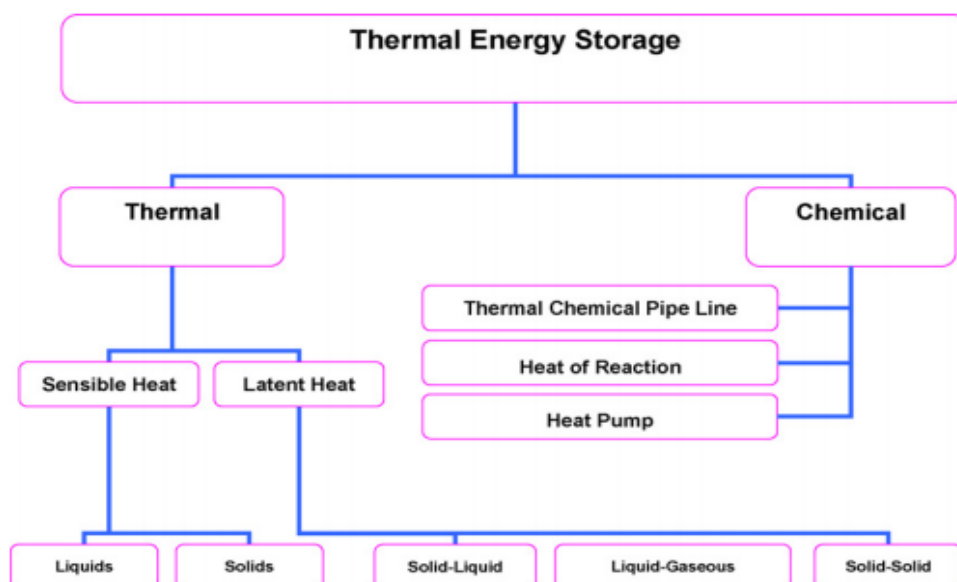


Figure 2.1: Different types of thermal energy storage (Sharma et al.,2007)

2.2.1 Sensible Heat Thermal Energy Storage (SHTES)

Sensible heat is associated with a general change and relies on the specific heat capacity of the storage material. It is achieved by storage from the heating medium (liquid sodium, molten salt or pressurised water) and increasing the energy content, but not altering the accumulation status. Energy is released and absorbed by the medium as the temperature reduces and increases respectively. Sensible heat can be stored in either solid media example as packed beds or liquid state media such as molten salt or pressurised water (Gil el al.,2010). Water seems to be the best SHTES liquid due to low cost and high specific heat (Khyad et al.,2016).

2.2.2 Latent Heat Thermal Energy Storage (LHTES)

Latent heat storage is associated with a phase change of material and is known as the energy put in or taken out to enable the change in phase. Energy required during charging is used to convert a solid material to liquid material or liquid material to gas. Phase change material (PCM) is the best example for LHTES with high number of advantages with particularly small temperature difference between small unit sizes, storage and retrieval cycles, and low weight per unit of storage capacity (El-Dessouky and Al-Juwayhel., 1997). A few certain criteria for the ideal PCM are shown in Table 2.1.

Table 2.1: Criteria for ideal Phase Change Material (Sharma et al.,2007)

Criteria	Remarks
Thermal properties	<ul style="list-style-type: none">• High latent heat of transition temperature• Good heat transfer• Suitable phase-transition temperature
Physical properties	<ul style="list-style-type: none">• Low vapour pressure• Small volume change• Favourable phase equilibrium• High density
Kinetic properties	<ul style="list-style-type: none">• Sufficient crystallisation• No supercooling
Chemical properties	<ul style="list-style-type: none">• Non-flammable and explosive• Compatibility with materials of construction• No fire hazards• Long-term chemical stability
Economics	<ul style="list-style-type: none">• Inexpensive• Available• Abundant

2.3 Phase Change Material (PCM)

Phase change material has been developed as an effective thermal energy storage technology. As seen in Figure 2.2, the general classification of PCM is broken into three categories such as organic, inorganic and eutectic.