A STUDY OF COPPER BASED NANOFLUIDS THERMOPHYSICAL PROPERTIES AS ADVANCED COOLANT IN AUTOMOTIVE RADIATOR

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RESEARCH VOTE NO:

PJP/2012/FKM(10A)/S01085

Fakulti Kejuruteraan Mekanikal

Universiti Teknikal Malaysia Melaka

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ACKNOWLEDGEMENT

Alhamdulillah. All Praise and Thanks to Allah.

Without great support of Universiti Teknikal Malaysia Melaka, (UTeM) it would not possible to complete this research project (Grant No. PJP/2012/FKM(10A)/S01085). I would also like to express my deepest appreciation to all those who have always been very responsive in providing necessary information, ideas and contributions to this project. Furthermore I would also like to acknowledge with much appreciation the crucial role of the technicians; Mr. Asjufri Muhajir and Mr. Ismail Ibrahim who helped us in using all required equipments and the necessary materials to complete the nanofluids production and engine coolant test rig. A special thank goes to my research group members, Mdm Nurhidayah binti Ismail, Mdm Nor Faizah binti Haminudin, and Mdm Nurul Hilwa binti Mohd Zini. Last but not least, many thanks go to the undergraduate students, Mr Amiruddin Mohd Ashari, Mr Romzi Che Yahya, Ms Nur Hamizah Lia Ketalik, Ms Hidayah Md Isa and Mr Muhammad Huzairi Che Hassan whose gave their full efforts in completing this research project.

The Research Principal,

Fadhilah binti Shikh Anuar

Abstract

The objective of this study is to investigate the thermophysical properties of copper (Cu) based fluids; thermal conductivity and dynamic viscosity of the water, ethylene glycol (EG) and a mixture of water and ethylene glycol (water/EG) suspended with Cu particles. Furthermore, in this study, an engine coolant test rig was built to investigate the heat transfer performance of Cu-based fluids as a new engine coolant. Initially, the Cu-based fluids were produced by using two-step method and their thermophysical properties were investigated experimentally based on the Cu particles concentration, temperatures, types of surfactant and particles sizes. The stability of the Cu-based fluids was observed for 12 days. It showed that the thermal conductivity of Cu-based fluids increased with particles concentration and temperatures. In addition, the temperature variations are more significant factor to enhance the Cu-based fluids thermal conductivity than particles sizes. The dynamic viscosity of Cu-based fluids with 0.1 – 0.5 vol.% particles concentration were comparable to the conventional engine coolants such as pure water and water/EG. The surfactants in this study; Polyvinylpyrrolidone (PVP) and Centyltrimenthylammoniumbromide (CTAB) had stabilized the Cu-based fluids up to 14 days based on the particles concentration and the types of base fluids. The Cu-based fluids showed better heat transfer capabilities compared to the conventional engine coolants and it could be considered as advanced engine coolant with further researches on specific areas.

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NOMENCLATURE

C_p	Specific Heat Capacity [kJ/kg.K]
D	Diameter [m]
f	Friction Loss
g	Gravitational Force [m/s^2]
h_L	Head Loss [m]
L	Distance [m]
$\dot{m}$	Mass Flow Rate [kg/s]
ΔP	Pressure Drop [Pa]
ρ	Density [kg/m^3]
Q	Heat Loss [kJ]
ΔT	Temperature Difference [K]
μ	Viscosity [kg/m.s]
V	Velocity [m/s]

CHAPTER 1

INTRODUCTION

1.1 Research Background

Cooling is the most important technical challenges in diverse industries such as transportation, microelectronics, medical and air-conditioning industries. The conventional method to increase the heat dissipation by increasing the surface areas for better heat transfer has achieved its limitation. Small and light devices with excellent heat transfer performances have been preferred nowadays to meet the customers' needs. Therefore, an innovative coolant should be introduced to improve the cooling capacity. Nanofluids are studied to be implemented in many industries to enhance the heat transfer performance of the systems. The production of nanofluids from diverse materials, base fluids and nanoparticles concentration has been done to obtain the most stable nanofluids for a long time usage. After a research of mathematical modeling on radiator nanofluids by Leong et al., (2010), many other researchers are trying to implement the nanofluids in transportation industry to increase heat transfer performance especially in an engine cooling system based on diverse areas and nanomaterials through different approaches (Chintakayala & Rajamanickam, 2013; Peyghambarzadeh et. al., 2011; Vajjha, et. al, 2010). The nanofluids are introduced to replace the conventional coolants; water and a mixture of water and ethylene glycol (water/EG). The thermophysical properties of nanofluids especially thermal conductivity and dynamics viscosity showed promising properties as compared to the conventional coolants (Kwak & Kim, 2005; Mahbubul et. al., 2013). However there are many unsolved problems related to the nanofluids as advanced coolant in the engine cooling system such as preferred nanoparticles materials, nanoparticles concentration, stability, sedimentation and corrosion on engine cooling parts due to particles suspension. These shortcomings should be solved to commercialize the nanofluids as the advanced engine coolant in the near future.

1.2 Objectives

This study was conducted with the purpose to produce Cu-based nanofluids as an advanced coolant with better heat transfer characteristics using two-step method in synthesizing the nanofluids. In addition, the nanofluids thermophysical properties were investigated to have advanced thermal properties to replace the conventional coolants. Primarily, the objectives of this study are stated as follows:-

(i) To investigate the thermal conductivity and dynamic viscosity of Cu-based fluids by considering particles concentration, temperatures, particles sizes and surfactant.

(ii) To investigate the stability of Cu-based fluids with addition of two types of surfactants.

(iii) To study the Cu-based fluids heat transfer capabilities through a car radiator.

1.3 Scope of Study

Nanofluids are introduced with diverse types of nanoparticles material such as metallic, non-metallic, carbon nanotube (CNT) or any other material classes. However, there are uncertainties on the nanoparticles reactions that contributed to better heat transfer process. This study focused on one type of material; copper as the enhancer of thermophysical properties of pure water, EG and a mixture of the water/EG. Only sole manufacturer of EG (the same type of coolant) has been considered throughout the experiment to be mixed with water or suspended with Cu particles. This study considered main thermophysical properties such as the thermal conductivity and dynamic viscosity of Cu-based fluids even though the other thermophysical properties such as density and surface tension may effects the heat transfer characteristics of the advanced coolants. The thermophysical properties of Cu-based fluids were investigated by considering certain temperature limits, surfactants, particles sizes and particles concentration.

In analyzing the stability of Cu-based fluids, this study introduced an observation method and two surfactants; Polyvinylpyrrolidone (PVP) and Centyltrimenthylammoniumbromide (CTAB). The duration of stability is considered up to 14 days, focusing on Cu/water fluids stability. Only 200 ml of Cu-based fluids was produced to be added with 6000 ml of water/EG solution in order to satisfy the minimum volume of heater tank in the engine coolant test rig. The pressure drop and heat loss in the test rig has been discussed as a value added for this study. The high temperature of heat from engine combustion was replaced by the heater with temperature ranges of 60 - 95°C which is lower than the real temperature of engine combustion. However, the heat transfer capabilities of the Cu-based fluids could be obtained from the small range of temperatures by including some necessary assumptions. Other components such as rubber hose, and car water pump are changed accordingly to match the engine coolant test rig design and experimental tests.

1.4 Problem Statement

It is well-known that the nanofluids could show better thermophysical properties compared to conventional fluids such as automotive coolant. However, the heat transfer performance of the nanofluids in the radiator should be investigated so that the capabilities of nanofluids as advanced coolant could be identified precisely. Until now, researchers are trying to identify suitable materials to be suspended into the conventional coolants as water, EG, refrigerant, oil and many other types of fluids to achieve optimum performance of nanofluids. The problem rises are not only because of materials compatibility, but the particles concentration is also becoming an issue that causes sedimentation or instability in dispersing particles into solution. There are different material selections and contradict results on thermophysical properties or efficient methods in synthesizing the nanofluids. Researchers are also believed that lack of theoretical understanding of underlying mechanisms with respect to nanoparticles has developed huge barrier to ensure nanofluids implementation into real practice.

Further researches on thermal conductivity enhancement are required to identify the optimum nanoparticles concentration that shows an outstanding heat transfer performance without any deficiency likes instability, agglomeration, sedimentation or deteriorating the engine cooling system. Several surfactants have been proposed to have better stability of nanofluids, yet the results were inconsistent among different researchers. In fact, prior to nanofluids mass production and real practice application, more researches are necessary especially on production cost, stabilization of particles, and superior performance in existing system or current technologies. Although a number of patents on nanofluids findings, the fundamental mechanisms of nanofluid heat transfer have not yet well understood especially in the car engine cooling system (Fadhilah et al., 2013). Therefore, the use of nanofluids as advanced engine coolants is questionable as uncertainty raised in its performance from all the results (Kothawale, 2013).

This study seeks for further improvement on the fundamental knowledge of the thermophysical properties and heat transfer performance of nanofluids as coolant for the automotive engine cooling system. The heat transfer performance was investigated by constructing an engine coolant test rig as a model of a real engine cooling system. This study should contributes a body of knowledge on nanofluids potentials as a new types of engine coolant in term of thermophysical properties and heat transfer characteristics.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

Energy consumption reduction in every industrial and domestic application is important to save the environment and benefits us in term of money and health. The energy savings could be achieved by innovating an equipment e.g. a heat exchanger especially its heat transfer performance. In past years, different types of geometry and design of heat exchangers have been analyzed in enhancing the heat transfer processes. Besides the design and materials of heat exchangers, further researches have been conducted in producing an advanced fluid as a new approach to increase the heat transfer characteristics in many other applications such as medical, air-conditioning system, electronic device and automotive engine cooling system. The production of nanofluids from diverse materials, conventional fluids and nanoparticles concentration have been done to come out with better thermal properties of the nanofluids for real-life application. The nanofluids are potential fluids with superior properties to replace conventional fluids such as water, deionised water, refrigerant, coolant, lubricant, etc. The term of “nanofluids” has been introduced by Choi in 1995 at Argonne Research Laboratory as an advanced fluid that showed superior heat transfer properties with nanoparticles suspension. The nanofluids were grouped based on their application which occasionally called as nanolubricant or nanorefrigerant, which is also one kind of nanofluids referred to the type of conventional fluids. The superior properties and stability of nanofluids are considered as main research areas nowadays as it challenges the significance of nanofluids implementation in existing application. Suitable material of nanoparticle is crucial to be identified in order to be suspended in different types of conventional coolants.

2.2 Thermal Conductivity and Dynamics Viscosity of Nanofluids

Nanofluids studies mainly involved thermal conductivity and heat transfer coefficient of the nanofluids since the thermophysical properties show a very significant influence on heat transfer processes. Murshed et. al., (2007) stated that the thermal conductivity of nanofluids varies with three attributes; size, shape and material of nanoparticles. Other properties such as viscosity, density and surface tension of nanofluids have also been explored to obtain reliable results for massive implementation in future. The nanofluids have superior thermophysical properties which have been proved since past decades by many researchers due to the suspension of nanoparticles. The nanoparticles could be metal, non-metal or carbon nanotubes (CNTs) which must be dispersed into the conventional fluids to produce nanofluids. Eastman et. al., (1997) proved that the thermal conductivity of nanofluids that contains CuO, Al_2O_3 and Cu nanoparticles with two different conventional fluids; water and HE 200 oil showed 60% improvement for the thermal conductivity compared to the corresponding conventional fluids for only 5 vol.% of nanoparticles suspension. Nanoparticles suspended into the refrigerant (nanorefrigerant) also has higher thermal conductivity than conventional pure refrigerant (Jiang et. al., 2007). Meanwhile, Abu-Nada (2009) investigated the thermal conductivity and viscosity of alumina water nanofluids mathematically using different models to evaluate the heat transfer performance in natural convection. The study claimed that for alumina particles more than 5 vol.% and Rayleigh number more than 10^4 has decreased the Nusselt number but lower particle concentration increased the Nusselt number. The size of nanoparticles, temperature, and optimum concentration of nanoparticles must be considered carefully. These factors are important to be considered so that high thermal conductivity and heat transfer coefficient of nanofluids could be obtained without causing agglomeration, instability, corrosion, high pressure drop and pumping power (Saidur et. al., 2011; Leong et. al., 2010; Han, 2008). The enhanced viscosity proved that the power efficiency would be increased in the engine cooling system. Basically, the dynamic viscosity of nanofluids could be influenced by amount of particles concentration. The minimum amount of volume fraction produced much lower viscosity (Saterlie et. al., 2011). However, Garg (2008) showed that with low Cu particles concentration about 2 vol.% suspended into water had increased almost four times of the viscosity compared to Einstein's law of viscosity. Li et, al., (2011) investigated the temperature effect on the viscosity of nanofluid. The temperature range is 20 - 60ºC with different conventional fluids such as Cu/kerosene, Cu/tolune and Cu/decahydronaphthalene. Even though the dynamic viscosity could be enhanced by the particles concentration, but it is also decreasing with temperature increment. Meanwhile, Corcione et. al,. (2011) proved that the ratio between the dynamic viscosities of the nanofluids and the conventional fluids could be increased by increasing the particles volume fraction and decreasing the particles size. Mahbubul et.al., (2013) also proved that the particle sizes and temperatures changed the viscosity of nanofluids in alumina/R-134a, therefore the viscosity of nanofluids could not be neglected as it might effects the pressure drop and pumping power of the system.

2.3 Nanofluids Stability

Nanofluids production influences the stability of nanoparticles suspension. Usually, there are two methods could be used to produce nanofluids; one-step method and two-step method. In two-step method, the nanoparticles need to be prepared first before dispersing into the base fluids. Basically, the nanoparticles could be produced by using physical method or chemical method. The physical methods included the grinding process and inert gas condensation. Meanwhile, the chemical precipitation method, chemical vapor deposition, thermal spraying, micro-emulsion are classified as chemical method. In addition, when the chemical method cannot produce the size of desired particles, manual grinding process could be one of the solutions. Meanwhile, the one-step method is another way to produce nanofluids. In this method, the nanofluids is simultaneously produced and dispersed directly into base fluids. Usually this method is really good in avoiding any oxidation especially when handling non-metallic such as copper dioxide, alumina, zinc oxide etc.

Many factors influenced the stability of nanofluids such as production method, material selection and types of surfactant introduced to the solution. The production method using two-step method, stirrer, and ultrasonic bath showed poor performance in avoiding particles clustering and agglomeration but high pressure homogizer magnetron sputtering system are better alternative to achieve longer stability of nanofluids due to their capabilities in producing smaller size of particles (Hwang et. al., 2008). In fact, the particles clustering and agglomeration increase the particles size and due to gravity force, the agglomerated particles would cause sedimentation in the system. In addition, the stability of nanofluids also could be influenced by pH values. Huang et. al., (2009) proved that the optimal value of pH resulted in highest stability of Cu/water and alumina/water through Derjaguin-Landau-Verwey-Overbeek (DLVO) calculation. The DLVO calculation considered the stability of a particle in the solution based on the van der Waals attractive and electrical repulsive forces between particles (Yu & Xie, 2012). However, the optimal pH values for different types of material are different such as alumina/water and Cu/water are 8 and 9.5 respectively (Mukherjee & Paria, 2013). Another easier way to determine the stability of nanofluids are through the measurement of particle size distribution using a microscopy and light scattering techniques with very high resolution microscope.

Surfactants added into the nanofluids could lower the surface tension of the conventional fluids and increasing the immersion of particles. The adequate amount of surfactants for each solution is important to make sure its usage is a helpful to stabilize the nanoparticles dispersion and avoiding fast sedimentation. Some importants surfactants included Sodium dodecyl sulfate (SDS), salt and oleic acid, Dodecyltrimethylammonium bromide (DTAB), Hexadecyltrimethylammoniumbromide (HCTAB), Polyvinylpyrrolidone (PVP), and arabic gum which the high temperature envinronment might damage the bonding between surfactant and nanoparticles (Mukherjee & Paria, 2013).

2.4 Nanofluids as Coolant for Engine Cooling System

There are two types of coolants in the engine cooling system; air from the surrounding and liquid (water, EG and a mixture of water/EG) that flowing inside the system. Nowadays, many vehicles used a mixture of water and EG as the engine coolant. The EG has higher boiling point and lower freezing point compared to water. It has been used to stand high temperature from the heat of fuel burning and prevent freezing coolant in winter season in some countries. Even though the properties of EG is good enough as coolant, the action to fill the coolant into the radiator without a mixture of water will cause harm to the engine cooling system. The EG which is corrosive and might leak the radiator tubes or deteriorate the engine body for a long term usage (Ho & Ching, 2010). It is undeniable the important of coolant function to remove heat from the engine. Nowadays trends introduce the use of advanced fluid to remove away heat seem as excellent potentials. The conventional cooling method to increase heat loads by increasing the surface area for heat transfer is not effective due to high cost, size increases and complexity of the system. These factors encourage researchers to investigate the coolant heat transfer capabilities. s

Many researchers are investigating the nanofluids as one of the advanced fluids to exhibit better cooling capacity than conventional fluids. In automative engine cooling system, researchers are trying to find the optimum performance of nanofluids as engine coolants and investigating the effects of the nanofluids on the engine cooling system (Chintakayala & Rajamanickam, 2013; Fadhilah et al., 2013; Peyghambarzadeh et. al., 2011). The researches have been conducted extensively either mathematically or experimentally to determine the potential of nanofluids as better coolant than the conventional coolants. Through mathematical analyses, Fadhilah et. al., (2013) and Leong et. al., (2011) agreed that nanofluids could show better heat transfer capability than conventional coolants after considering the flow of nanofluids in the car radiators. In addition, the heat transfer rate and thermal performance of Cu/EG coolant in an automotive radiator can be enhanced by increasing the particle volume fraction from 0 -

2 % (Leong et. al., 2010). The enhancement of heat transfer depends on air and coolant Reynolds number (Re) which is increasing with nanoparticle concentration. Mare et. al., (2011) proved that the convective heat transfer coefficient of CNTs nanofluid increased about 50 % in comparison to water for the same Reynolds number experimentally. Basically, there are five factors that can enhance the heat transfer; Brownian motion, layering at the solid/liquid interface, Ballistic phonon transport through the particles, nanoparticles clustering, and friction between the nanoparticles and fluid (Wang and Mujumdar, 2007).

The use of nanofluids as advanced coolant should be able to reduce the size of radiator. However, there are many aspects need to be investigated such as the compatibility of nanofluids with the engine cooling system, the stability of nanofluids in high temperature environment and the possible damage and pressure drop of the system due to the effects of nanofluids implemetation. Razi et. al., (2011) investigated the heat transfer and pressure drop of CuO-base oil nanofluid flow inside horizontal flattened tubes under constant heat flux of 2600 W/m^2 and proved that the pressure drop of nanofluids increased with nanoparticle concentration. There is also a withdrawn investigation of nanofluids natural convective heat transfer since the suspension of nanoparticles caused higher viscosity and pressure drop as compared to conventional fluid (Calvin & Peterson, 2010). Therefore, there are a lot of factors that need to be considered when deciding to introduce specified nanofluid as a new alternative for heat transfer enhancement in automotive engine cooling system.

CHAPTER 3

METHODOLOGY

3.1 Introduction

This study intended to produce nanofluids using two-step method. Cu particles were produced by using chemical reduction method and suspended into different types of conventional coolants; water, EG and a mixture of water/EG. In order to enhance the stability of nanofluids, two surfactants were mixed into the copper based solution. This study considered Polyvinylpyrrolidone (PVP) and Cetyltrimethylammoniumbromide (CTAB) for particles volume fraction from 0.1 - 0.5%. The thermophysical properties of Cu-based fluids such as thermal conductivity and viscosity were measured to analyze the capabilities of copper particles to enhance the fluids thermal properties. Regardless the stability on the fluids, it would be tested in the test rig right after production. The performance of the Cu-based fluids were measured accordingly and any damages or sedimentation of Cu particles were considered at the end of the testing process.

3.2 Cu-Based Fluids Synthesis

As mentioned in the Section 2, the Cu-based fluids could be produced either one-step method or two-step method. This study chose two-step method to avoid any impurities at the end of the production of Cu-based fluids and inexpensive. The method started with producing Cu particles at first. The production of copper particles required the following main substances; sodium hydroxide, copper acetate, acetone, hydrazine. The physical properties of the substances could be seen in Table 3 -1.

Table 3-1 Physical properties of substances for producing copper nanoparticle

Material / Physical Property	Copper acetate monohydrate	Sodium hydroxide	Hydrazine	Acetone
Physical state	Solid	Solid	Liquid	Liquid
Molecular weight	199.65 g/mole	40 g/mole	50.06 g/mole	58.08 g/mole
Color	Blue	White	Colorless	Colorless
Boiling point	Decompose	1338ºC	113.5ºC	56.2ºC
Melting point	115ºC	323ºC	-51.7ºC	-95.35ºC

It took about 48 hours to synthesis the copper particles which most time used in drying the solution to extract the particles. The synthesis processes adapted from Zhu et. al., (2004) and changes were expected as higher volume of particles is needed in this study. Initially 160 g of copper acetate and 40 g of sodium hydroxide dissolved in 4 liter of distilled water using magnetic stirrer for 1 hour. Some ice cubes placed outside the beaker while stirring to cool down the temperature solution to 5°C. After adding 25.7 ml hydrazine, the solution changed color from blue to brownish and the stirring process was continued for 12 hours at room temperature. Hydrazine was introduced as a forming agent in arranging polymer foam of copper particles. Then, the copper particles filtered and washed using acetone. Finally, the particles were dried up in an oven (universal oven 321 memmert model UM200) for 20 hours at 50°C. The process of nanoparticles synthesis is shown in Figure 3-1. As the particles were obtained, the morphology of the particles was studied using a scanning electron microscopy (SEM). The diameter and shape of particles are the main factors to be investigated in order to make sure the thermal properties is a promising to be used as an advanced coolant.

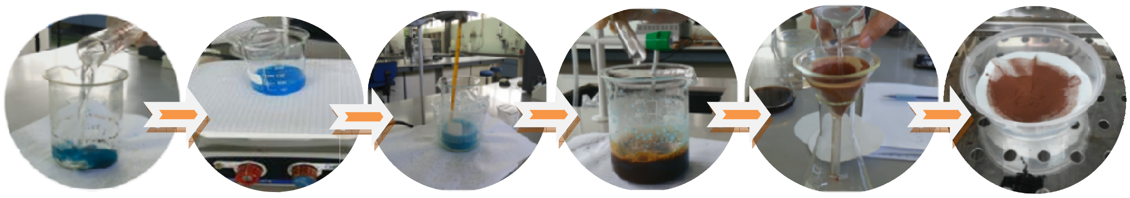

Figure 3-1 Copper particles production process

In producing nanofluids, it is important to calculate the volume fraction of copper particles precisely. This study considered Eq. (3-1) to have the desired volume fraction,

$$Vol(\%) = \frac{\frac{W_{cu}}{cu}}{\frac{W_{cu}}{cu} + \frac{W_f}{f}} \times 100 \qquad (3\text{-}1)$$

where W_{cu} is the weight of copper particles, W_f is the weight of any types of basefluid, ρ_{cu} and ρ_f are the densities of the copper and basefluids respectively. The copper particles concentration, and amount of surfactants used are shown in Table 3-2 and the particles were mixed into three different solution (water, EG and a mixture of water/EG) for homogenizing process at 1000 rpm for 3 minutes using a homogenizer (HG-15D WiseMixTM). The addition of 0.2 g of surfactants for each samples of Cu/water could be helpful in stabilizing the solution. An ultrasonic cleaning unit (Elmasonic s30h series) was used at the end of the process to avoid agglomeration of copper in early stage of nanofluids production. The ultrasonic with high frequency electric energy transformed into mechanical energy, transmitted into the bath. Adequate amount of Cu-based fluid to be used in engine coolant test rig required the manipulation of substances weight to produce Cu particles as stated in Section 3.5.

Table 3-2 Particles concentration used in producing copper based fluids

Copper particles volume fraction (%)	PVP / CTAB (g)	Water (g)
0.1	0.2	39.442
0.2		39.043
0.3		38.723
0.4		38.363
0.5		38.003

3.3 Thermophyiscal Properties Measurement

Basically, the thermal conductivity of pure copper is higher than conventional coolants. Therefore, by adding the copper particles into the conventional coolant (water) with thermal conductivity of 0.578 W/m.K at room temperature, the thermal conductivity of the mixure solution should be higher than the conventional coolants. In this study, the thermal conductivity of mixture solution was measured using KD2 Pro Thermal Properties Analyzer (manufactured by Decagon Devices Inc.) shown in Figure 3-2. The device equipped with handheld controller and several sensors that suitable to measure diverse types of material and fluids. It also used to measure the resistivity of substances. For fluids, the measurement could be done by collecting the fluid into a specimen container (cylinder tube with a cap made of silicon rubber). Then, the needle of the device inserted through the silicone cap without touching the surface or the bottom of the tube. This device needs to be calibrated for each measurement since it is highly sensitive that influence the results accuracy. The procedure of thermal conductivity measurement is listed as follows:-

i. KS-1 sensor was used as it was designed to measure liquid samples and insulating material with thermal conductivity less than 0.1 W/m.K.
ii. The KS-1 sensor was connected to the port on KD2-Pro device and switched on.

iii. The solution was transferred into the specimen container covered with a silicone cap.
iv. Then, the needle must be fully inserted through the silicone cap.
v. The KD2-Pro device was setting to “Auto” mode and “Enter” icon was pressed to begin the measurement.
vi. An indicator bar shown on the screen implies the time required to complete the measurement. Usually it would take a few seconds for the results to be displayed.

Figure 3-2 Thermal conductivity analyzer

This study used Brookfield DV-II+ Pro Viscometer (manufactured by Brookfield Engineering Laboratories, INC, USA) to measure the dynamic viscosity of the Cu-based fluid as shown in Figure 3-3 and it consists of different spindles that could be used in analyzing the dynamic viscosity of diverse fluids. Spindle LV-2 (62) was used to measure the solutions in this study. The investigation of the thermophysical properties, either the thermal conductivity or the viscosity of Cu-based fluids has been conducted by considering the effects of temperature, surfactants and particle concentration. The procedure of dynamic viscosity measurement is stated as follows:-

i. Spindle LV62 was mounted on the DV-II+ Pro Viscometer and then, the spindle was inserted into the beaker.
ii. It must be mounted in the center of beaker and the groove at spindle shaft must be fully immersed into the solution. The spindle shaft should not touch the