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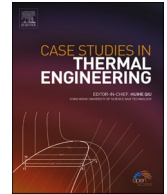
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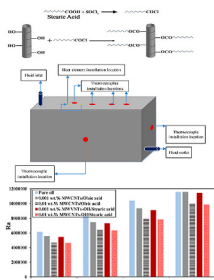
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Experimental investigation on thermal performance of covalently functionalized hydroxylated and non-covalently functionalized multi-walled carbon nanotubes/transformer oil nanofluid

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GRAPHICAL ABSTRACT



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ABSTRACT

The study investigated the effect of adding covalently functionalized-hydroxylated multi-walled carbon nanotubes (MWCNTs-OH) and non-covalently-functionalized MWCNTs on the breakdown voltage and thermal properties of transformer oil in a rectangular chamber. The novelty of the present study is the use of covalently functionalized hydroxylated and non-covalently functionalized multi-walled carbon nanotubes in transformer oil, and the reason for the selection of these nanoparticles is the high intrinsic thermal conductivity compared to other nanoparticles. Between both studied nanofluids, the thermal and electrical performance of covalently functionalized MWCNTs-OH was better due to the highest increase in heat transfer coefficient of free transfer and using fan was related to covalently functionalized MWCNTs-OH, which increased by 26.23% and 30.08%, respectively. Also, by measuring the breakdown voltage, it was found that the

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MWCNTs-OH of 0.001 wt% had the lowest reduction compared to the base fluid and was equal to 55.6 kV, which showed good performance because the specified standard for transformer oil breakdown voltage property is between 30 kV and 70 kV. According to the results, covalently functionalized hydroxylated MWCNTs-OH/transformer oil nanofluid has better thermal performance than pure oil, which prevents the transformer from rising in temperature and can also be used as electrical insulation in transformers.

Nomenclature

A	Heat transfer area
C_p	Specific heat capacity
CNTs	Carbon nanotubes
FTIR	Fourier-transform infrared spectroscopy
Gr	Grashof number
G	Gravitational acceleration
H	Convection heat transfer coefficient
I	Electric current
K	Thermal conductivity coefficient
L	Length
Nu	Nusselt number
MWCNTs	Multi-walled carbon nanotubes
MWCNTs-OH	Functionalized-hydroxylated multi-walled carbon nanotubes
Pr	Prandtl number
Q	Input power
Q	Heat flux
R	Radius
R	Quantity
Ra	Rayleigh number
T	Temperature
TEM	Transmission electron microscope
U_{Ri}	Maximum possible error
U_{xi}	Measurement error
V	Voltage
W	Particle weight fraction
x_i	Measurable parameter

Greek letters

β	Coefficient of volume expansion
ν	Kinematic viscosity
ρ	Density
μ	Dynamic viscosity
φ	Particle volume fraction

Subscripts

Bf	Base fluids
C	Cold
H	Hot
Nf	Nanofluids

1. Introduction

Transformers are one of the most widely used electrical equipment applied in many different industries. Cooling of the transformer is of special importance because if the temperature of the transformer oil rises too much, not only will it damage the components of the transformer and reduce its performance, but if the temperature exceeds a certain limit, the transformer will be completely out of circuit. Therefore, trying to improve the thermal performance of the transformer not only increases its efficiency but also increases the life of the device and reduces its size, which in turn reduces manufacturing costs [1]. In oil transformers, special oil is used to dissipate

the heat generated by the coils and other factors, that the heat of the parts is transferred to the oil and this heat is transferred to the outside through the tank walls that are in contact with the environment [2,3]. During this process, the oil begins to heat up until the amount of heat generated equals the amount of heat transferred by the oil. Important parameters of transformer oil include thermal coefficient, dielectric coefficient and breakdown voltage, on which the performance of the oil is strongly dependent. Today, various industries are looking for new ways to improve heat transfer processes [4,5].

One of the solutions is to use nanoparticles as additives to improve the properties of energy-carrying fluids [6]. Nanofluids are a group of fluids obtained by suspending nanoparticles within the base fluid. Nanofluids are generally prepared by two distinct methods, one-step and two-step respectively. With the one step method, nanoparticles are directly produced within the base fluid from laser ablation, direct evaporation method or chemical methods [7]. With the two-step method, nanoparticles, nanotubes, nanofibers, or other nanomaterials are dispersed in the fluid from mechanical and/or sonication processes [8]. In this study, a two-stage method has been used. Nanomaterials are an effective method to enhance the efficiency of transformers. The more sophisticated lubricants currently available typically include both pure oils and additives. Base oil is the major component of the lubricant that specifies its main attributes. The percentage of additives in fluids is much lower than in base fluids, but additives play a key role in creating novel attributes or compensating for the disadvantages of pure fluids [9,10]. Several kinds of nano-additives, such as carbon-based nanoparticles (such as MWCNTs and graphene) [11,12], metal oxides (such as Al_2O_3 , ZnO , and Fe_3O_4 nanoparticles) [13–15], and metal nanoparticles (such as Fe, Ni, Ag, and Cu) have been applied as nano-additives [16,17]. The main problem in the use of nanoparticles is the highest tendency to agglomeration, which is due to the ratio of surface to the high volume of nanoparticles and therefore the high surface energy of nanoparticles. Nanoparticle surface modification has been used to minimize the surface energy of nanoparticles and decrease their tendency to form agglomeration [18]. Ideal surface modification means improving the surface properties of nanoparticles without changing their major properties [19]. Modification of the surface of carbon nanotubes can be done by physical and chemical methods. Physical modification is done by using surfactants or macromolecules that are adsorbed on the surface of nanoparticles. Adsorption of surfactants on the particle surface has occurred by electrostatic interactions of polar groups. Chemical modification occurs by modifying covalent bonding to the nanoparticle surface [20,21]. In this study, both covalent and non-covalent methods have been used to modify the surface of carbon nanotubes. Carbon-based materials such as MWCNTs and MWCNTs-OH are other additives that have been of interest to researchers due to their high thermal attributes [22,23].

Many researchers have studied the impact of MWCNTs on the thermophysical and rheological features of pure oils. Based on their results, MWCNTs are recognized as the best nano-additives to improve the thermal attributes of working fluid. The following are examples of studies examining the impact of MWCNTs and other additives on the thermal performance of transformer oil.

Aberoumand et al. [24] studied the impact of Tungsten (III) oxide (WO_3)–Silver nano additives on the stability, thermal conductivity, and dielectric strength of transformer oil. They reported that the thermal conductivity of the applied hybrid nanofluid at 4 wt % at 100 °C increased by 41% and also the nanofluids had long-term stability. On the other hand, the dielectric strength of hybrid nanofluids was reduced from 53 kV to 35 kV by adding 4 wt% of nanoparticles. Beheshti et al. [3] examined the effect of oxidized MWCNT additions on transformer oil's flash point, thermal conductivity, and electrical conductivity. They found that adding 0.001 wt % oxidized MWCNT to transformer oil increased the flash point temperature by 4.6% compared to the base oil and increased the thermal conductivity for nanofluids by 0.01 wt% and 0.001 wt% of nanoparticles by 5.7% and 7.7%, respectively. Electrical conductivity, on the other hand, has not altered significantly as nanoparticle concentration and temperature have increased. Amiri et al. [25] by adding hexylamine-activated carbon nanotubes covalently to transformer oil acknowledged that at a low concentration of 0.005 wt%, the thermal conductivity of the nanofluid increased by about 10%. The free convection heat transfer coefficient and forced convection heat transfer coefficient increased by about 23% and 28%, respectively. They reported that the flash point increased with the increasing concentration of nanoparticles in transformer oil. Also, at a concentration of 0.005 wt%, the breakdown voltage decreased by about 11%. Amiri et al. [26] investigated the effect of graphene particles on the rheological and thermal properties of transformer oil. Comprehensive rheological and thermal analysis of nanofluids compared to pure transformer oil did not show any significant increase in viscosity, which is a great advantage of the new generation of transformer oil. On the other hand, nanofluids based on transformer oil with the addition of graphene nano-additives have significant potential as a new generation of high efficiency cooling for use in transformers. Fontes et al. [27] examined the impact of carbon nanotubes (CNTs) and diamond nanoparticles on the viscosity, thermal conductivity, and breakdown voltage of transformer oil. Experimental results indicated that the thermal conductivity and dynamic viscosity are enhanced with increasing particle concentration, so that by adding 0.05 wt% of CNTs and diamond nanoparticles, the thermal conductivity of oil increases by 27% and 23%, respectively. On the other hand, adding these nanoparticles to the pure oil reduces the breakdown voltage of the oil. Choi et al. [28] prepared nanofluids by dispersing Al_2O_3 and AlN nanoparticles in transformer oil and then investigated the thermal properties of the fluids. The results showed that by adding nanoparticles to transformer oil, the thermal conductivity increases. For example, AlN nanoparticles in a volume fraction of 0.5% can increase the thermal conductivity of transformer oil by 8% and the overall heat transfer coefficient by 20%. Mehrvarz et al. [29] in their simulation study of transformer oil, they reported that the addition of 1 vol% TiO_2 nanoparticles to transformer oil causes an improvement in the heat transfer coefficient, the Nusselt number increases from 2.17 to 2.49, while the maximum temperature of the transformer components decreases from 47.20 °C to 43.05 °C.

The following are examples of studies examining the impact of carbon base additives on the thermal performance of the various base fluids.

Amiri et al. [21] investigated the influence of hexylamine-functionalized CNTs on laminar convective heat transfer attributes of turbine oil. They reported that the heat transfer was enhanced at different concentrations of HA-treated CNTs, which is ascribed to the decline of the thermal boundary layer by CNTs. Ilyas et al. [30] evaluated the influence of functionalized alumina on thermal, stability, and rheological properties of thermal oil. They reported that the oleic acid coated alumina is prepared and then mixed in the oil to

overcome the agglomeration of additives in pure oil. The viscosity of the fluid is enhanced by the increase in nano-additive concentrations. The effective thermal conductivity of the nano-lubricant is enhanced as compared to thermal oil. Pourpasha et al. [31,32] experimentally investigated the impact of MWCNTs and TiO₂ nanoparticles on the thermophysical attributes of turbine meter oil. Their results illustrated that the thermophysical attributes of nano lubricants improved compared to pure oil. For example, by adding 0.3 wt % of MWCNTs and TiO₂ nano additives to turbine meter oil, the viscosity index of nanofluids was increased by 2.43% and 6.65%, respectively. By adding 0.3 wt% of MWCNTs and TiO₂ nanoparticles to turbine meter oil, the flash point of nanofluids was enhanced by 1.77% and 4.4%, respectively. Naddaf et al. [33] investigated the effect of modified graphene nanoparticles and carbon nanotubes on the thermal properties of diesel oil. Their results indicated that the thermal conductivity and electrical conductivity of all nanofluids increased at all weight concentrations relative to pure diesel oil. In addition, with increasing temperature, thermal conductivity and electrical conductivity increase for all weight concentrations. Mohammed et al. [34] studied the thermal performance of a hybrid nanofluid's flow boiling in a rectangular channel. Their results showed that increasing the graphene loading by 0.5 vol% increased thermal conductivity by 4.5% and raising the nanoparticle concentration improves the heat transfer coefficient at the heated surface, as well as the boiling point, which increases both the saturation temperature and vapour pressure. Mohammed et al. [35] reported that using a zinc oxide nanoparticle concentration of less than 1% by volume provides sufficient property enhancement whilst allowing for a stable suspension. On the other hand, the vapour production rate rises with nanoparticle concentration due to the improved thermal diffusion because the nanoparticles cause faster heating of the nanofluid. Mohammed et al. [36] reported that the properties of the nanofluid acetone/ZnBr₂ with graphene, the density, and the specific heat follow the simple mixture combination rule, and the thermal conductivity of the heat transfer coefficient was linearly proportional to the temperature difference between the fluid and the wall.

Based on the several studies conducted based on the fluids, extensive investigations have been led to oils. In contrast, little research has been done on transformer oil. Considering the previous research on transformer oil based nanofluids, the covalently functionalized hydroxylated multi-walled carbon nanotubes/transformer oil nanofluid has not studied and the usage of covalently functionalized hydroxylated MWCNT-OH and non-covalently functionalized MWCNTs in transformer oil is novel in this work, and the key reason for selecting these nanoparticles is their high intrinsic thermal conductivity compared to other additives. In this study, the first MWCNT additive was modified with the non-covalently method of oleic acid, and MWCNTs-OH was modified with the covalently method by stearic acid. Secondly, nanofluids containing non-covalently functionalized MWCNTs and covalently functionalized MWCNTs-OH nano-additives in various concentrations (0.001 wt% and 0.01 wt%) were prepared through a two-step technique. Then, the effect of the nano-additives concentration on breakdown voltage and thermophysical properties, including thermal conductivity coefficient, specific heat capacity, density, and viscosity, were theoretically evaluated. Also, the impact of the nano-additives concentration and input powers (50, 70, 90, and 120 (W)) on thermal properties, including convective heat transfer coefficient, Nusselt number (in the two conditions of air flow, including free air flow and forced air flow out of the system), Rayleigh number, and Grashof number, were studied.

2. Material and method

2.1. Materials

MWCNTs and MWCN-OH as nano-additives to enhance the pure transformer oil properties were purchased from Shenzhen Nanotech Port Co. The characteristics of MWCNTs are shown in Table 1. Oleic acid, stearic acid, thionyl chloride, dichloromethane, and chloroform were purchased from Merck. Table 2 indicates the specifications of transformer oil as the pure oil that was obtained from Nynax, Stockholm, Sweden. The properties of nanoparticles and pure oil were provided by the manufacturer, except for the oil breakdown voltage, which was measured with a breakdown voltage measuring device according to IEC156/95.

2.2. Modification of nano-additives and synthesize nanofluids

In order to functionalize the MWCN-OH with the stearic acid functional group, in the first step, 5 ml of thionyl Chloride (SOCl₂) was added dropwise to a solution containing 0.3 g of stearic acid (C₁₈H₃₆O₂) and then 10 ml of dichloromethane (CH₂Cl₂) was added at a temperature of 80 °C and mixed with a mixer at this temperature for 1 h. After 1 h, the solution temperature reached room temperature, then 1 gr of hydroxyl carbon nanotubes was added and mixed with a magnetic stirrer at 60 °C for 5 h. After this time, the resulting suspension was thoroughly washed with distilled water and chloroform simultaneously with the Polytetrafluoroethylene membrane to remove unreacted thionyl chloride. The resulting sample was placed in an oven at 60 °C for 48 h to dry completely.

To synthesize non-covalently functionalized MWCNTs/transformer oil, nanofluids, a two-step technique was applied. For this aim,

Table 1
Multi walled carbon nanotubes properties.

Nanoparticles	Diameter (nm)	Specific heat capacity (J/kg.K)	Thermal conductivity coefficient (W/m.K)	Purity (%)	Density (kg/m ³)
MWCNTs	10–20	550	2000	>99	2100

Table 2
Transformer oil characteristics.

Fluid	Density (kg/m ³) at 15 °C	Flash point (°C)	Pour point (°C)	Viscosity in 40 °C (cP)	Breakdown voltage (kV)
Transformer oil	855	140	-30	12.82	58

a determined amount of oleic acid (weight ratio of 1 to 1 nano-additive) as a surfactant and transformer oil were mixed with the ultrasonic (Made by Pars Nahand company, model S7500 with a tank capacity of 6 L and dimensions of $15 \times 15 \times 30$ "depth \times width \times length" with a power of 100 W) for 1 h. Then, MWCNTs nanoparticles were added to the solution and mixed with ultrasonic for 6 h. A similar method was used to synthesize covalently functionalized MWCNTs-OH/transformer oil, nanofluids. However, because the carboxyl MWCNTs were modified with stearic acid, oleic acid was not required, and after the addition of covalently functional MWCNTs-OH, the suspension was placed in an ultrasonic bath for 6 h.

2.3. Characterization

In this work, the functional groups of MWCN-OH functionalized by stearic acid were examined using Fourier-transform infrared spectroscopy (FTIR). Transmission electron microscope (TEM) analysis was applied to study the morphology of nanoparticles.

2.4. Thermal properties measurement system explanation

To measure the convection heat transfer coefficient, a system similar to a transformer device has been designed and fabricated in laboratory dimensions, so that the material and thickness of the body of the system were made of the body material of transformers used in industry (Its body is made of St37 brand steel and the thickness of the steel body is 3 mm in the tank part and 4 mm in the tank cap). Fig. 1 indicates a schematic of the experimental setup applied to measure the thermal properties and transformer oil and nanofluids. The experimental setup includes an oil tank, thermocouples (Four thermocouples inside the fluid and two thermocouples on the body), thermostats, and heating element. In the first step of the test, pure transformer oil was used, and 50, 70, 90 and 120 W of power were selected. After transferring the transformer oil in the transformer tank, the temperature of each thermocouple was recorded. Then we turned on the transformer and set its power to minimum power. The temperature increased over time and after the temperature remained constant and reached a constant state, the power increased and the temperature was recorded again. The same method was used to obtain the thermal properties of the nanofluids. Also, forced convection heat transfer is a mechanism of heat transfer in which fluid moves through an external force such as a fan or pump. In this section, to create a forced current, a fan with a speed (2850 rpm) and a power of 40 (W) is used vertically, which is located at a distance of 10 cm from the large wall of the transformer.

2.5. Relevant equations

The model presented by Xue [37] was used to calculate the thermal conductivity coefficient of nanofluids (k_{nf}). This model is the developed Maxwell model which has a good compatibility for oil-based nanofluids with carbon nanotube additives, equation (1) is applied.

$$k_{nf} = \frac{(1 - \varphi) + 2\varphi \frac{k_{CNT}}{k_{CNT} - k_{bf}} \ln \frac{k_{bf} + k_{CNT}}{2k_{bf}}}{(1 - \varphi) + 2\varphi \frac{k_{bf}}{k_{CNT} - k_{bf}} \ln \frac{k_{bf} + k_{CNT}}{2k_{bf}}} k_{bf} \quad (1)$$

where, φ is a particle volume fraction, k_{CNT} is the thermal conductivity of the CNT, and k_{bf} is the thermal conductivity of the transformer oil. Also, specific heat capacity (C_{pnf}) and density (ρ_{nf}) are calculated from equations (2) and (3) [38]:

$$C_{pnf} = \varphi C_{pCNT} + (1 - \varphi) C_{pbf} \quad (2)$$

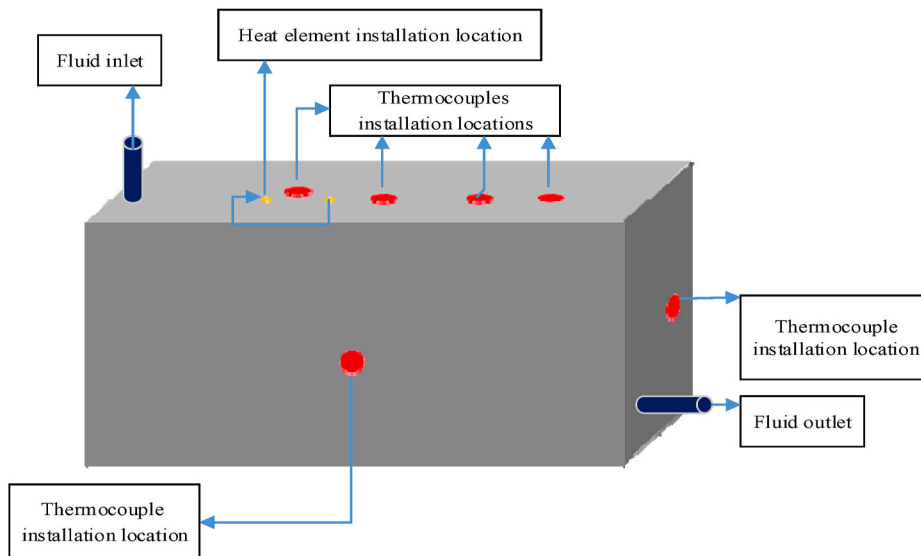


Fig. 1. Schematic of the experimental system.

$$\rho_{nf} = \varphi\rho_{CNT} + (1 - \varphi)\rho_{bf} \quad (3)$$

where $C_{p,CNT}$ is the specific heat capacity of the CNT, $C_{p,bf}$ is the specific heat capacity of the transformer oil, ρ_{CNT} is the density of the CNT, and ρ_{bf} is the density of transformer oil. Equation (4) is used to convert weight fraction to volume fraction. Which w is the weight fraction of the CNT.

$$\varphi = \frac{w\rho_{bf}}{\rho_{CNT}(1 - w) + w\rho_{bf}} \quad (4)$$

where, w is a particle weight fraction. The following equation [39] is used to determine viscosity, which is represented as follows:

$$\mu_{nf} = \mu_{bf}(A + B\varphi + C\varphi^2 + D\varphi^3) \quad (5)$$

At 25 °C, $A = 1.0087$, $B = 0.1553$, $C = -0.0334$, and $D = 0.0631$ are constant values.

During the free convection heat transfer, the displacement process occurs only due to the density difference between various layers of the fluid due to its temperature variation. In this way, the density of the lower layers of the fluid decreases due to heating and then the fluid moves upwards. The amount of fluid that moves depends on the kind of fluid, the value of temperature difference, and the volume of space in which the fluid is located. The force that causes fluids to move is the buoyancy force. Grashof (Gr) number is a dimensionless number in natural convection heat transfer, which is defined as the ratio of buoyancy force to viscous force and is obtained from the following equation:

$$Gr = \frac{g\beta(T_h - T_c)L^3}{\nu^2} \quad (6)$$

T_h is the temperature of the hot wall, T_c is the temperature of the cold wall, g is the gravitational acceleration equal to 9.81, β is the coefficient of volume expansion equal to 0.00075 (1/°C), L is the distance between the hot and the cold walls equal to 5 cm, and ν is the kinematic viscosity defined by:

$$\nu = \frac{\mu}{\rho} \quad (7)$$

In equation (7), μ is the dynamic viscosity and ρ is the density. Another dimensionless number in natural convection heat transfer is the Nusselt number (Nu), which depends on Gr and Prandtl (Pr) in free convection heat transfer. The Nu and Pr numbers are defined as follows:

$$Nu = \frac{hL}{k} \quad (8)$$

$$Pr = \frac{C_p \mu}{k} \quad (9)$$

In equation (8), h is the convection heat transfer coefficient. Another dimensionless number called the Rayleigh number (Ra) is used in the natural heat transfer. Ra number is as follows:

$$Ra = Gr.Pr = \frac{g\rho^2 C_p \beta (T_h - T_c) L^3}{k\mu} \quad (10)$$

The value of input power in this experiment is calculated from equation (11):

$$Q = V.I \quad (11)$$

I is the value of electric current and V is the voltage applied to the device. The average heat transfer coefficient under constant heat flux is calculated as follows. The heat transfer between the hot wall, which is hypothetically considered inside the transformer, and the cold wall is investigated. The material of these walls is steel; the high thermal conductivity of the steel causes the conduction heat transfer from the walls to be negligible and also due to the low heat flux, the heat loss is neglected due to the conductive heat transfer.

$$\bar{h} = \frac{Q}{A(T_h - T_c)} \quad (12)$$

where A is the heat transfer area of the system, which is equal to 0.130 m². Temperatures T_h and T_c are obtained by thermocouples located on the body and inside the chamber. The mean Nusselt is obtained from the following equation:

$$\overline{Nu} = \frac{\bar{h}.l}{k} \quad (13)$$

2.6. Uncertainty analysis

In this study, various parameters have been measured, each of which has a different accuracy, so it is necessary to evaluate the impact of measurement error of these parameters on the results. If the quantity R is a function of the parameters x_1, x_2, \dots, x_n , the

impact of the measurement error of the parameter x_i on the quantity R is calculated as follows [40].

$$U_{Ri} = \frac{x_i}{R} \frac{\partial R}{\partial x_i} U_{xi} \tag{14}$$

where x_i is measurable parameter, R is quantity calculated from measurable parameters, U_{xi} is measurement error, and U_{Ri} is maximum possible error in calculating a quantity. The total error of the quantity R due to the errors of the parameters x_i is calculated as follows

$$U_R = \mp (U_{R1})^2 + (U_{R2})^2 + \dots + (U_{Rn})^2 \tag{15}$$

The convective heat transfer coefficient (h) is calculated from the following equation.

$$h = \frac{q}{\Delta T} \tag{16}$$

The value of the convective heat transfer coefficient error (U_h) is obtained from equation (17). The heat flux (q) and its error value (U_q) are obtained from equations (18) and (19).

$$U_h = \sqrt{(U_q)^2 + (-U_{\Delta T})^2} \tag{17}$$

$$q = \frac{Q}{A} = \frac{Q}{2\pi rL} \tag{18}$$

$$U_q = \sqrt{(U_Q)^2 + (-U_r)^2 + (-U_L)^2} \tag{19}$$

The input power is calculated from equation (11) and the error rate is calculated from the following equation.

$$U_Q = \sqrt{(U_V)^2 + (U_I)^2} \tag{20}$$

The Nusselt, Rayleigh, and Grashof number errors (U_{Nu} , U_{Ra} , and U_{Gr}) calculated as follows.

$$U_{Nu} = \sqrt{(U_h)^2 + (U_L)^2 + (-U_k)^2} \tag{21}$$

$$U_{Ra} = \sqrt{(U_{\Delta T})^2 + 2(U_\rho)^2 + (U_{cp})^2 + 3(U_I)^2 + (-U_\mu)^2 + (-U_k)^2} \tag{22}$$

$$U_{Gr} = \sqrt{(U_{\Delta T})^2 + 2(U_\rho)^2 + 3(U_I)^2 + 2(-U_\mu)^2} \tag{23}$$

The value of the measurement error of different parameters is given in Table 3.

3. Result

In this part, firstly, the Characteristics of nanoparticles and stability of nanofluids have been investigated. Then the effect of increasing non-covalently MWCNT (MWCNTs/Oleic acid) and covalently functionalized MWCN-OH (MWCNTs/Stearic acid) nano-additives on transformer oil properties, including the natural convective heat transfer properties and forced convective heat transfer properties, and breakdown voltage, has been experimentally investigated in detail. Also, the effect of inlet powers (50, 70, 90, and 120 (W)) on the natural convective heat transfer properties and forced convective heat transfer properties have been studied. The thermophysical properties, including the of nanofluids have been studied theoretically.

Table 3
Parameters error values.

Parameters	Symbol	The amount of error (%)
Electric current	U_I	0.13
Voltage	U_V	2.78
Temperature difference	U_T	2.56
Thermal element length	U_L	1.37
Radius of the thermal element	U_r	0.03
Tank length	U_L	1
Density	U_ρ	2
Specific heat capacity	U_{Cp}	2
Viscosity	U_μ	2
Thermal conductivity coefficient	U_k	2
Convective heat transfer coefficient	U_h	4.02
Nusselt number	U_{Nu}	4.60
Rayleigh number	U_{Ra}	5.43
Grashof number	U_{Gr}	5.05

3.1. Characteristics analysis

Table 4 indicates the approximate areas in which different bonds are adsorbed [41]. Fig. 2 indicates the FTIR result of covalently-functionalized MWCN-OH. Sample of stearic acid-functionalized MWCN-OH shows clear signs of functionalization. According to Fig. 2, peaks 1270 and 1625 correspond to the C-O and C=C bonds, respectively. The wide peak between 3000 and 3500 is related to the hydroxyl (-OH) groups in the sample. The most important peaks are around 1400, 2850 and 2900, which are related to the stretching vibrations of C=O, -CH₃ and -CH₂ for the stearic acid group [41].

Fig. 3(a), (b) illustrate TEM images of MWCN-OH functionalized with stearic acid, In TEM analysis, although TEM cannot show functional groups, it can be evidence of functionalization by showing surface roughness. Stearic acid has been able to improve the stability of nanofluids by modifying the surface of MWCNT-OH, and the homogeneous dispersion of nanotubes may be seen in this form. A TEM picture of non-covalently functionalized MWCNTs is shown in Fig. 3(c).

Fig. 4(a), (b), and (c) illustrate a comparison between transformer oil/MWCNTs at the beginning of dispersion and after 25 days. Fig. 4(d), (e) show a comparison between transformer oil/covalently functionalized MWCNTs-OH at the beginning of dispersion and after 40 days. Nanofluid containing non-covalently functionalized MWCNTs has good stability up to 10 days, after 10 days, nanofluid at 0.01 wt% of non-covalently functionalized MWCNTs, precipitations were started. According to Fig. 4(a), (b), and (c), nanofluids containing non-covalently functionalized MWCNTs nano-additives are precipitated up to 25 days. According to visual observations, nanofluids containing non-covalently functionalized MWCNTs nano-additives are precipitated up to 25 days. According to visual observations, nanofluids containing covalently functionalized MWCNTs-OH nano-additives have good stability up to 40 days and slight precipitations were observed. In the covalent method, the functional groups (such as functional groups of stearic acid) are attached to the main structure of carbon nanotubes by chemical reactions. This is accompanied by the breaking of π bonds, which improves the stability of the nanofluid. Alternatively, the non-covalent method (one of the physical methods) uses various surfactants (such as oleic acid) to improve the stability of the nanofluid. According to the results, the modification of the surface of MWCNTs of covalent method has a better stability compared to the use of surfactant directly in transformer oil because nanoparticle surface modification has been used to minimize the surface energy of nanoparticles and decrease their tendency to form agglomeration.

3.2. Thermophysical results

Table 5 indicates the thermophysical properties of the MWCNTs/transformer oil, nanofluids including thermal conductivity coefficient, specific heat capacity, density, and viscosity. As the concentration of MWCNTs in transformer oil increases, the thermal conductivity coefficient of the nanofluid increases because the thermal conductivity of the MWCNTs is higher than the transformer oil (about 18,000 times) and its presence in the fluid improves the thermal conductivity of the nanofluid. According to Table 5, the thermal conductivity of nanofluid with 0.01 wt% of MWCNTs is equal to 0.118 (W/m.K), which has increased by about 8% compared

Table 4

Vibrational frequencies range of bonds [41].

Functional group	O-H N-H	C-H	C≡C C≡N	C=O	C=N C=C	C-Cl C-O C-N C-C
Visible frequency range	3500–3000	3000–2500	2500–2000	1800–1650	1650–1550	1550–650

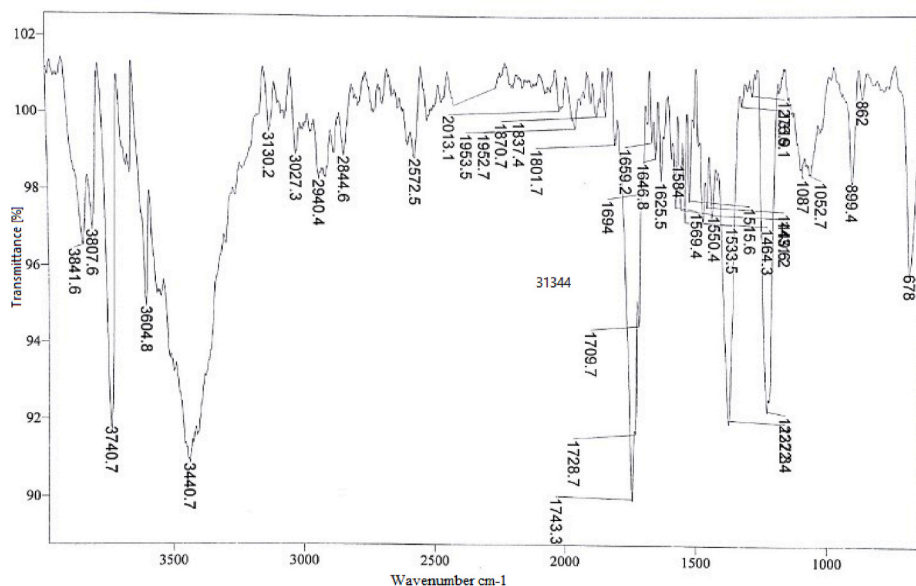


Fig. 2. Infrared spectrum of MWCN-OH functionalized with stearic acid group.

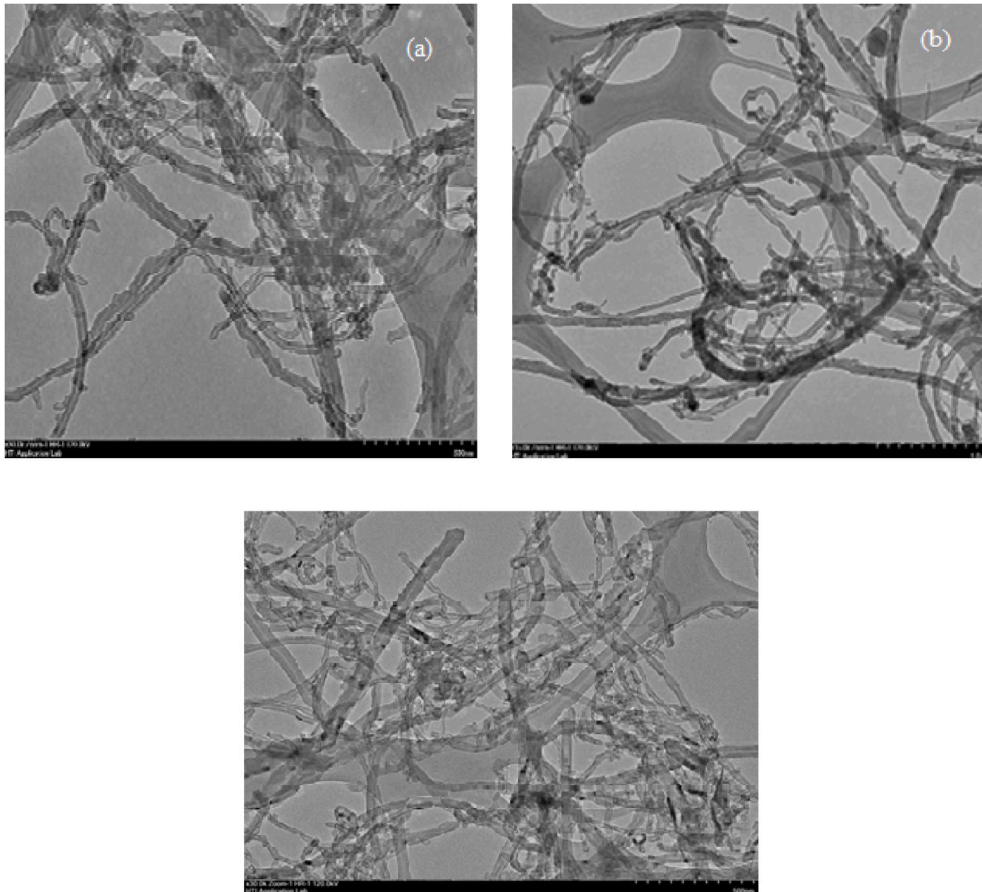


Fig. 3. (a), (b) TEM images of covalently functionalized MWCN-OH (c) TEM image of non-covalently functionalized MWCNs.

to transformer oil. The specific heat capacity of the nano-lubricant decreases with increasing concentration of MWCNTs. Because the specific heat capacity of pure nanofluid is higher than that of carbon nanotubes, and as a result, the presence of nanomaterials in the oil reduces this property of nanofluid. The specific heat capacity of MWCNTs is 550 (J/kg.K), which is about 1340 (J/kg.K) less than pure oil. According to Table 5, the density of the oil increased by adding MWCNTs to the transformer oil. With the addition of MWCNTs additives to the transformer oil, the viscosity of the fluids in the different concentration of nanoparticles relative to the pure oil improved.

3.3. Natural convective heat transfer

Fig. 5 indicates the natural convective heat transfer coefficient of non-covalently MWCNTs/transformer oil (MWCNTs/Oleic acid) and covalently functionalized MWCNTs-OH/transformer oil (MWCNTs-OH/stearic acid) nano-lubricants in the different concentrations of nano-additives (0.001 wt% and 0.01 wt%) in the various input powers 50, 70, 90, 120 (W). The natural heat transfer coefficient is a function of input heat flux and temperature difference. By raising input power and thus increasing the input heat flux, the natural convective heat transfer coefficient for transformer oil and nanofluid increased, also with increasing input power and due to decreasing the temperature difference between hot and cold wall, the natural convective heat transfer coefficient increased. The greatest improvement in convective heat transfer coefficient occurred with increasing the concentration of nanoparticles at an input power of 120 (W) that with the addition of 0.01 wt% and 0.001 wt% of MWCNTs/Oleic acid additives to the transformer oil, the natural convective heat transfer coefficient of nanofluids enhanced relative to the pure oil by 24.62% and 14.48%, respectively. With the addition of 0.01 wt% and 0.001 wt% of covalently functionalized MWCNTs-OH additives to the transformer oil, the natural heat transfer coefficient of nanofluids improved relative to the pure oil by 26.23% and 15.59%, respectively. As the heat transfer coefficient increases, the amount of natural heat transfer increases and this parameter plays a significant role in the amount of heat transfer. Obviously, the higher this parameter, the greater the improvement in the performance of the transformer. The results show that nanofluids and pure fluid both have an increasing trend in heat transfer coefficient, proving that it is a temperature dependent property. The highest natural heat transfer coefficient is related to the concentration of 0.01 wt% of covalently functionalized MWCNTs-OH additives which is equal to 78.59 (W/m².K). By adding nanoparticles to the fluid, the thermal performance of the fluid increases. The main reasons for this increase are:

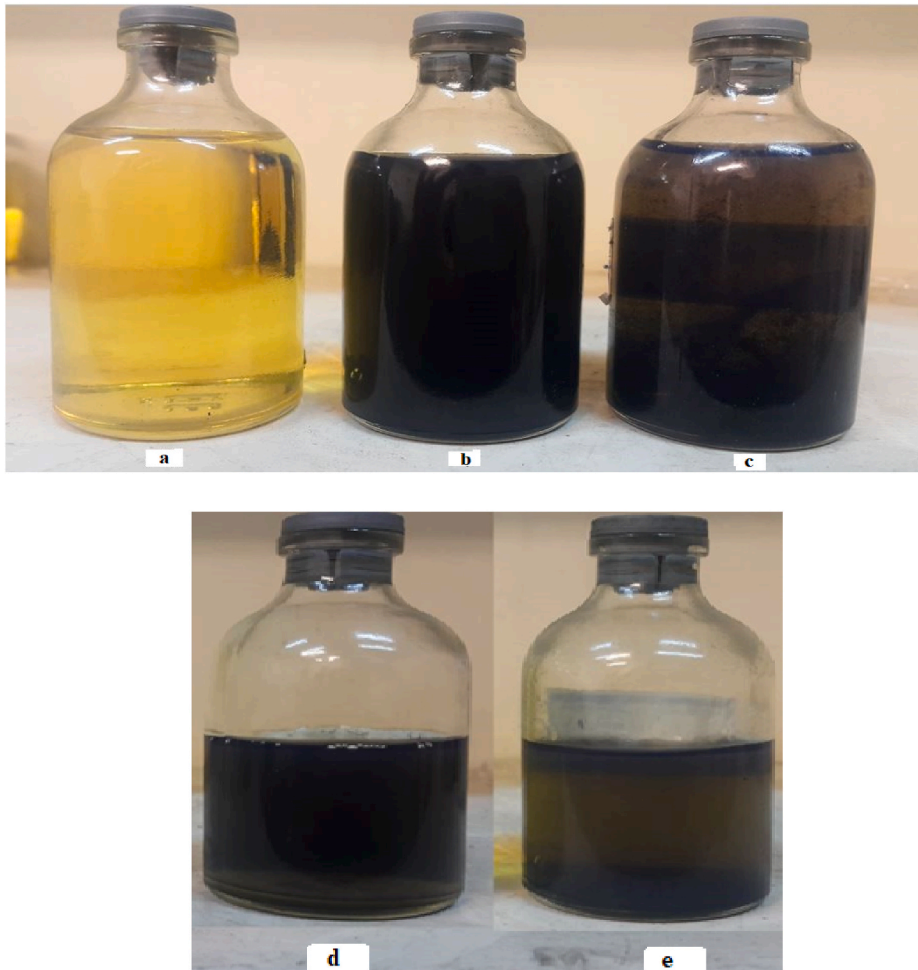


Fig. 4. a) Pure oil, b) Nanofluid with 0.01 wt% of MWCNTs after synthesis, c) Nanofluid with 0.01 wt% of MWCNTs after 25 days, d) Nanofluid with 0.01 wt% of covalently functionalized MWCN-OH after synthesis, e) Nanofluid with 0.01 wt% covalently functionalized MWCN-OH after 40 days.

Table 5

Thermophysical attributes of MWCNTs/transformer oil, nanofluids.

Concentration	Thermal conductivity coefficient (W/m.K)	Specific heat capacity (J/kg.K)	Density (kg/m ³)	Viscosity (cP)
Pure oil (0%)	0.11	1893	855	12.825
0.001 wt%	0.1108	1892.46	855.49	12.938
0.01 wt%	0.118	1887.62	859.98	12.956

- Suspended nanoparticles increase the level of heat transfer.
- The interaction and collision between nanoparticles, fluid and solid surface increases.
- Suspended nanoparticles increase the effective thermal conductivity of the fluid.
- Fluctuation and turbulence of the fluid are intensified by the presence of nanoparticles.
- The dispersion of nanoparticles causes the temperature gradient to spread throughout the fluid.

Fig. 6 shows the effect of MWCNTs/Oleic acid and covalently MWCNTs-OH/Stearic acid nano additives in the various input powers (50, 70, 90, 120 (W)) on the Nusselt number of transformer oil. The Nusselt number is a function of the convective heat transfer coefficient and the conductive heat transfer coefficient. The conduction heat transfer coefficient is much lower than the convective heat transfer coefficient. According to Fig. 6, increasing the input power leads to increasing the input heat flux and thus increasing the Nusselt number. As a result, the Nusselt number also increases with increasing heat flux. The highest increase in Nusselt number for nanofluid is related to input power of 120 (W) and concentration of 0.01 wt% MWCNTs/Oleic acid with a growth of about 16%. Also, the maximum enhancement in Nusselt number for nanofluid is related to input power of 120 (W) and concentration of 0.01 wt% covalently functionalized MWCNTs-OH/Stearic acid with an increase of about 17.6%. According to the results, covalently

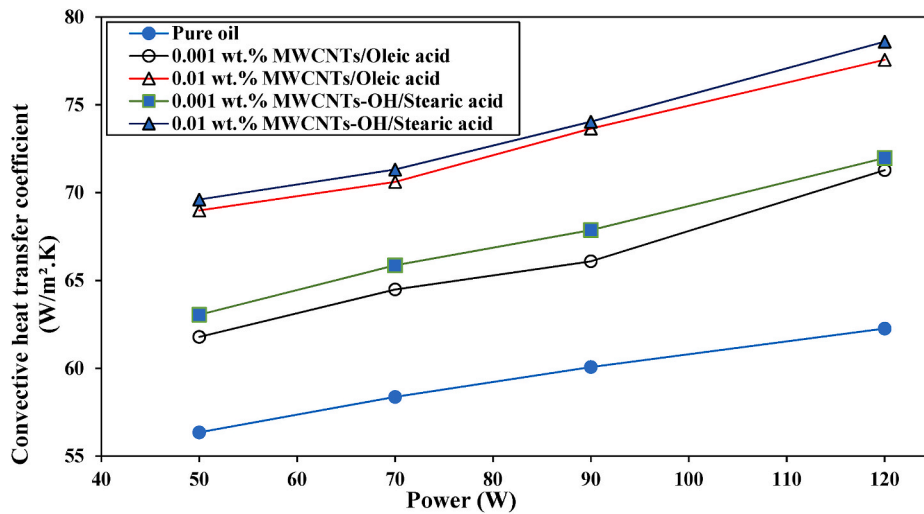


Fig. 5. Comparison of natural heat transfer coefficient of MWCNTs/transformer oil and covalently functionalized MWCNTs-OH/transformer oil nanofluids in different input powers.

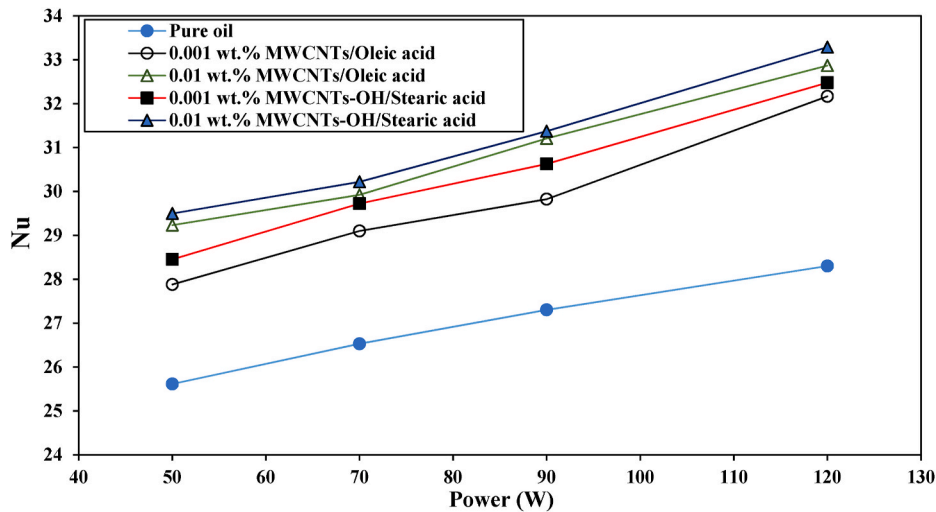


Fig. 6. Experimental Nusselt number versus input powers for MWCNTs/transformer oil and covalently functionalized MWCNTs-OH/transformer oil nanofluids at different concentration.

functionalized MWCNTs-OH has shown better performance, which can be due to better dispersion and stability and stronger bonding of stearic acid with MWCNTs-OH.

Fig. 7 indicates the impact of MWCNTs/Oleic acid and covalently MWCNTs-OH/Stearic acid nano additives in the various input powers (50, 70, 90, 120 (W)) on the Rayleigh number of transformer oil. According to Fig. 7, with raising the input power to the transformer and thus increasing the heat flux has led to an increase in the Rayleigh number. Also, according to equation (10), the Rayleigh number is directly related to the temperature difference between hot and cold walls, by increasing the concentration of MWCNTs cause to reduce temperature difference and as a result the Rayleigh number decreases. Also, since the Rayleigh number is the ratio of buoyancy force to thermal diffusivity the heat transfer increases with the increase of nanomaterials, so the Rayleigh number reduced by adding MWCNTs. With increasing concentration of nanoparticles, the Rayleigh number for nanofluids with covalently functionalized MWCNTs-OH additive decreased more than the nanofluids with MWCNTs/Oleic acid and base oil, which further decrease in Rayleigh number indicates high natural heat transfer for nanofluid with 0.01 wt% covalently functionalized MWCNTs-OH additive.

Fig. 8 indicates Grashof number variations with input power in different mass percentages of covalently functionalized MWCNTs-OH and MWCNTs/Oleic acid. The Grashof number is a dimensionless number that defines a ratio of buoyant forces to viscous forces a fluid. According to Fig. 8, in all concentrations, the Grashof number increased by raising input power due to the increase in temperature difference between hot and cold walls. Also, with increasing the concentration of MWCNTs/Oleic acid and MWCNTs-OH/

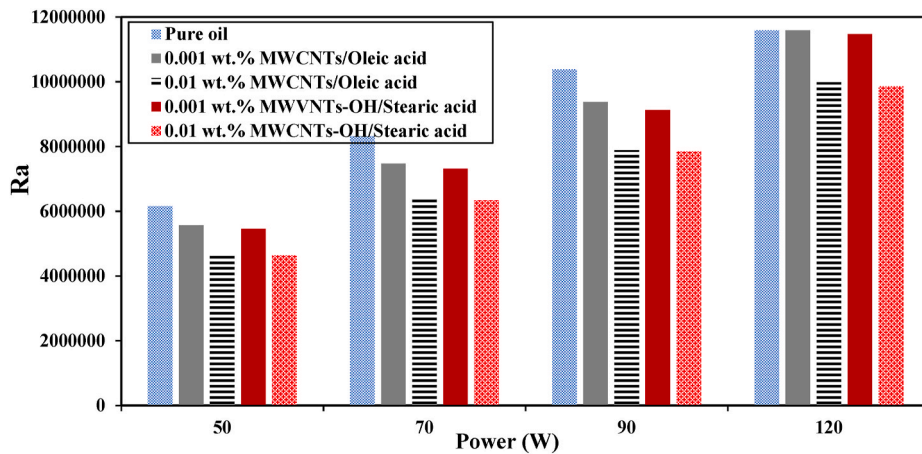


Fig. 7. Comparison of Rayleigh number of MWCNTs/transformer oil and covalently functionalized MWCNTs-OH/transformer oil nanofluids in different input powers.

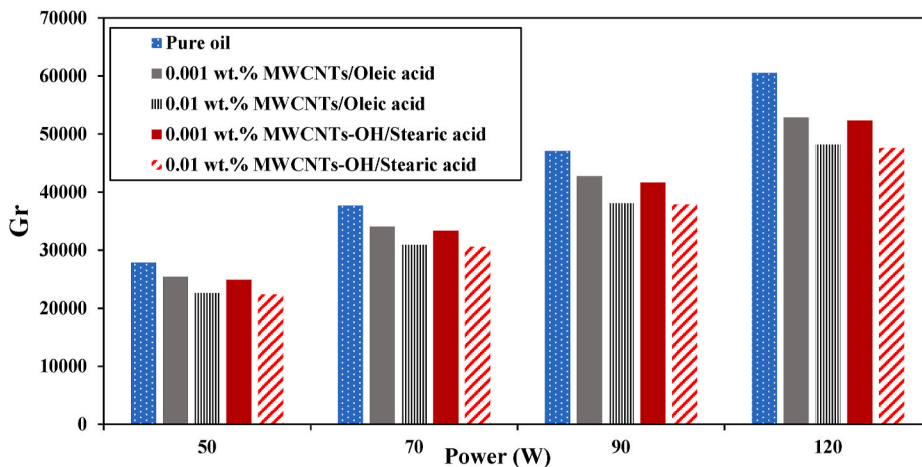


Fig. 8. Comparison of Grashof number of MWCNTs/transformer oil and covalently functionalized MWCNTs-OH/transformer oil nanofluids in different input powers.

Stearic acid in different mass fraction, due to the increase in heat transfer, the temperature difference between the hot and the cold walls decreases and the Grashof number decreases. At 120 (W) of input power, the Grashof number of transformer oil is equal to 60535.41, which by adding covalently activated MWCNTs-OH to the pure oil at a concentration of 0.01 wt%, the Grashof number is equal to 47577.77, which decreased by 21.40%.

3.4. The effect of forced air flow on free heat transfer of transformer oil

In the forced air flow, Nusselt number and heat transfer coefficient were investigated. The variations in the convective heat transfer coefficient by applying the external force of the fan with various input powers at two concentrations of 0.001 wt% and 0.01 wt% of MWCNTs/Oleic acid and covalently functionalized MWCNTs-OH/Stearic acid are shown in Fig. 9. In the forced air flow, such as free air flow, with raising input power and thus increasing the input heat flux, the convective heat transfer coefficient for transformer oil and nanofluid increased. The greatest improvement in convective heat transfer coefficient occurred with increasing the concentration of nanoparticles at an input power of 70 (W) that with the addition of 0.01 wt% and 0.001 wt% of MWCNTs/Oleic acid additives to the transformer oil, the convective heat transfer coefficient of nanofluids enhanced relative to the pure oil by 27.91% and 16.73%, respectively. With the addition of 0.01 wt% and 0.001 wt% of covalently functionalized MWCNTs-OH additives to the transformer oil, the natural heat transfer coefficient of nanofluids improved relative to the pure oil by 30.08% and 18.99%, respectively. According to the results, the convective heat transfer coefficient in the forced air flow is higher than the free air flow.

The variations in the Nusselt number by applying the external force of the fan with various input powers at two concentrations of 0.001 wt% and 0.01 wt% of MWCNTs/Oleic acid and covalently functionalized MWCNTs-OH/Stearic acid are shown in Fig. 10. The Nusselt number is a function of the convective heat transfer coefficient and the conductive heat transfer coefficient. The conduction heat transfer coefficient is much lower than the convective heat transfer coefficient. As a result, the Nusselt number changes are more a function of the convective heat transfer coefficient. The maximum enhancement in the Nusselt number occurred with increasing the concentration of nano-additives at an input power of 70 (W) that with the addition of 0.01 wt% and 0.001 wt% of MWCNTs/Oleic acid

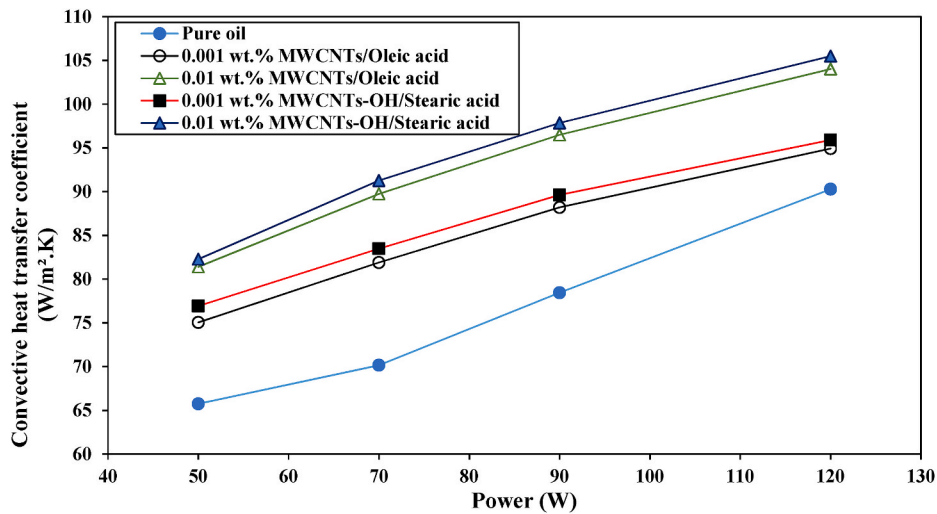


Fig. 9. The effect of forced air flow on the free convection heat transfer of MWCNTs/transformer oil and covalently functionalized MWCNTs-OH/transformer oil nanofluids in different input powers.

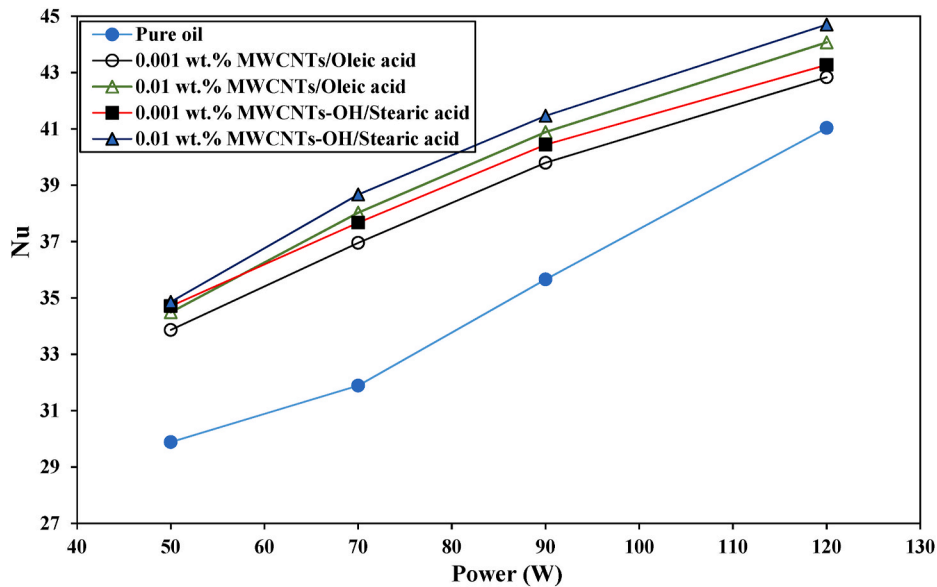


Fig. 10. The effect of forced air flow on the Nusselt number of MWCNTs/transformer oil and covalently functionalized MWCNTs-OH/transformer oil nanofluids in different input powers.

additives to the transformer oil, the Nusselt number of nanofluids increased relative to the pure oil by 19.24% and 15.88%, respectively. With the addition of 0.01 wt% and 0.001 wt% of covalently functionalized MWCNTs-OH additives to the transformer oil, the Nusselt number of nanofluids enhanced relative to the pure oil by 21.26% and 18.13%, respectively.

3.5. Comparison between convective heat transfer coefficient and Nusselt number in the forced air flow and free air flow

The changes in convective heat transfer coefficient and Nusselt number with input power of both the forced air flow and free air flow are given in Table 6. Convective heat transfer coefficient and Nusselt number by applying the external force of the fan due to the presence of external applied force in all nanofluid concentrations is higher than the convective heat transfer coefficient and Nusselt number in the free air flow. According to the experimental results, the effect of forced air flow on the heat transfer coefficient and Nusselt number increased by raising input powers from 50 to 120 (W). The greatest enhancement in the Nusselt number and the convective heat transfer coefficient for nanofluids with 0.001 wt% and 0.01 wt% of covalently functionalized MWCNTs-OH at an input power of 120 (W) (i.e., 33.24% and 34.21%, respectively) obtained by applying the external force compared with the same conditions without applying external force (Free air flow). The maximum increment in the Nusselt number and the convective heat transfer

Table 6
Comparison between convective heat transfer coefficient and Nusselt number in the forced air flow and free air flow.

Fluid	Input power (W)	h_{Free} (W/m ² .K)	h_{Forced} (W/m ² .K)	Nu_{Free}	Nu_{Forced}	Improvement%
Pure oil	50	56.353	65.746	25.615	29.884	16.66
Pure oil	70	58.369	70.157	26.531	31.889	20.18
Pure oil	90	60.070	78.448	27.304	35.658	30.06
Pure oil	120	62.264	90.276	28.302	41.034	44.98
0.001 wt% non-covalently MWCNTs	50	61.785	75.047	27.881	33.866	21.46
0.001 wt% non-covalently MWCNTs	70	64.486	81.895	29.100	36.956	26.99
0.001 wt% non-covalently MWCNTs	90	66.091	88.192	29.824	39.798	33.44
0.001 wt% non-covalently MWCNTs	120	71.280	94.918	32.166	42.833	33.16
0.01 wt% non-covalently MWCNTs	50	68.989	81.400	29.233	34.491	17.98
0.01 wt% non-covalently MWCNTs	70	70.617	89.743	30.220	38.027	27.08
0.01 wt% non-covalently MWCNTs	90	73.649	96.488	31.207	40.885	31.01
0.01 wt% non-covalently MWCNTs	120	77.569	104.08	32.868	44.071	34.17
0.001 wt% covalently MWCNTs-OH	50	63.051	76.923	28.452	34.712	22
0.001 wt% covalently MWCNTs-OH	70	65.866	83.482	29.723	37.672	26.74
0.001 wt% covalently MWCNTs-OH	90	67.870	89.619	30.628	40.441	32.04
0.001 wt% covalently MWCNTs-OH	120	71.974	95.904	32.479	43.271	33.24
0.01 wt% covalently MWCNTs-OH	50	69.613	82.270	29.497	34.860	18.18
0.01 wt% covalently MWCNTs-OH	70	71.319	91.264	30.220	38.671	27.96
0.01 wt% covalently MWCNTs-OH	90	74.034	97.852	31.374	41.463	32.17
0.01 wt% covalently MWCNTs-OH	120	78.599	105.494	33.288	44.701	34.21

coefficient for nanofluids with 0.001 wt% and 0.01 wt% of non-covalently functionalized MWCNTs at the inputs of 90 (W) and 120 (W) (i.e., 33.44% and 34.17%, respectively) obtained by applying the external force compared with the same conditions without applying external force.

3.6. Breakdown voltage

The breakdown voltage of a dielectric fluid is the minimum voltage that causes a part of a dielectric to become electrically conductive and conduct electrical current. Breakdown voltage is a very important parameter in transformer oils, so the fluid used in the transformer should have a higher breakdown voltage. By adding covalently activated MWCNTs-OH and non-covalently activated MWCNTs material due to the high conductivity of these materials, they affect the breakdown voltage of transformer oil. The breakdown voltage measuring device has two electrodes in a chamber in which the transformer oil is poured into the device. The voltage is automatically increased, the voltage at which the fluid goes out of its dielectric state is displayed as the breakdown voltage, and to be sure, this test was performed up to three times and its average was reported as breakdown voltage. These data are given in Table 7. According to the results, covalently functionalized MWCNTs-OH illustrated better performance due to higher dispersion, stronger bonding and better dielectric properties of stearic acid. The nanofluid with 0.001 wt% of covalently activated MWCNTs-OH has the best performance with a decrease of 4.1%, which indicates the low effect of the presence of covalently functionalized MWCNTs-OH at a concentration of 0.001 wt% on the breakdown voltage. The minimum value of breakdown voltage is related to 0.01 wt% non-covalently functionalized MWCNTs with a 28.44% decline in breakdown voltage.

4. Conclusion

The influence of covalently functionalized MWCNT-OH/transformer oil nanofluids and non-covalently functionalized MWCNTs/transformer oil nanofluids with the various concentration of MWCNTs and MWCNTs-OH (0.001 wt% and 0.01 wt%) and input powers range from 50 (W) to 120 (W) on the free convective heat transfer coefficient, Nusselt number, Rayleigh number, and Grashof number at free air flow condition, were investigated. Also, The effect of forced air flow on free heat transfer of transformer oil were investigated. Then the effect of increasing the concentration of nanoparticles on the thermophysical properties of transformer oil has been studied. The following conclusions are made:

1. The nanofluid with 0.001 wt% of covalently functionalized MWCNT-OH has the best performance with a decrease of 4.1%, which indicates the low effect of the presence of covalently functionalized MWCNTs-OH at a concentration of 0.001 wt% on the breakdown voltage. The minimum value of breakdown voltage is related to 0.01 wt% non-covalently functionalized MWCNTs with a 28.44% decline in breakdown voltage.

Table 7
The effect of adding functionalized carbon nanotubes on the breakdown voltage.

Fluid	Breakdown voltage (kV)
Pure oil	58
Nanofluid with 0.001 wt% of covalently functionalized MWCNTs-OH	55.6
Nanofluid with 0.01 wt% of covalently functionalized MWCNTs-OH	44.4
Nanofluid with 0.001 wt% of non-covalently functionalized MWCNTs	51.1
Nanofluid with 0.01 wt% of non-covalently functionalized MWCNTs	41.5

- The highest increase in heat transfer coefficient of free transfer and using fan was related to covalently functionalized MWCNT-OH at concentrations of 0.01 wt% and 0.001 wt% increased by 26.23% and 30.08%, respectively.
- The Rayleigh number for nanofluids with covalently functionalized MWCNTs-OH additive decreased more than the nanofluids with MWCNTs/Oleic acid and base oil, which further decrease in Rayleigh number indicates high natural heat transfer for nanofluid with 0.01 wt% covalently functionalized MWCNTs-OH additive.
- At free air flow, the maximum enhances in Nusselt number for nanofluid is related to input power of 120 (W) and concentration of 0.01 wt% covalently functionalized MWCNTs-OH with an increase of about 17.6%.
- At higher heat fluxes, the buoyancy force is increased for the nanofluid, which leads to an increase in the rotational motion of the fluid, which may be one way to control and reduce the precipitation of nanoparticles in the nanofluid.

Author contribution

Hojjat Alizadeh performed the experiments and drafted the manuscript, and provided the figures and tables as his M.S. final project. Hadi Pourpasha: Methodology, Writing - review & editing. Saeed Zeinali Heris was supervisor of M.S. thesis and all calculations and simulations were done under his supervision and manuscript text, figures and tables were drafted based on his comments. Patrice Estellé checked the manuscript and commented for improving it and also proposed to add some new experimental results.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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