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A review on preparation of nanocellulose for new green working fluid in heat transfer application

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Abstract. Cellulose is one of the sustainable raw materials and natural polymers as it can be abundantly found in biomass products. Nanocellulose consist of three categories which are bacterial nanocellulose (BNC), cellulose nanofibrils (CNF), and cellulose nanocrystalline (CNC) has the potential to be used for working fluid in heat transfer application with nanofluid characteristics. Nanofluid is defined as a fluid that has nanoparticle dispersed in it. This nanoparticle will enhance the thermophysical properties of the base fluid. Many types of nanofluid have been used widely in heat transfer applications such as Al2O3, TiO2, CuO and CNT but only a few studies had been done using green and environmentally friendly materials. Therefore, nanocellulose is one of the best candidates as one of the nanofluid's materials which has green and environmentally friendly properties. The preparation of nanofluid is very crucial because improper preparation will lead to deterioration of nanofluid and decrease the thermal performance as heat transfer working fluid. Other than that, low stability cause nanoparticle to form clusters and sediment due to its strong van der Waals interaction. Therefore, this study will focus on the preparation method of the nanocellulose for heat transfer application. Other than that, methods of stability analysis for nanocellulose also presented in this paper.

1. Introduction

Cellulose is one of the sustainable raw materials and natural polymers as it can be found abundantly in biomass products. Cellulose is a chain of glucose monomer organized into fibrils which is the main constituent of the plant cell wall. The most common sources of cellulose are from economic plantations such as wood and plantation by-products. Due to this, nanocellulose can be categorized as one of the natural nanomaterials. Other than that, despite being green material, nanocellulose also has a large surface area, high strength, and low-density extensive chemical modification. Nanocellulose is classified as cellulosic materials in the nanometer range with at least one dimension. [1]. The introduction of nanocellulose has given benefits to many applications such as medical, environmental, and material engineering. Klemm et al. [1] divided nanocellulose into three categories which are bacterial nanocellulose (BNC), cellulose nanofibrils (CNF), and cellulose nanocrystalline (CNC).

The systematic literature review can be described as quantitatively and qualitatively identifying, combining and interpreting all available data to provide a detailed response to a research question. The systematic literature review provides many advantages over traditional literature reviews, such as straight forward article retrieving process, wider study area, and more important criteria that can control research bias. This improves the literature review to produce quality evidence with more significant results.

Based on nanocellulose characteristics mentioned above, nanocellulose is one of the nanoparticles that can be dispersed in based fluid for heat transfer application. Therefore, this paper aims to gain insights into the nanocellulose dispersion methods in based fluid its thermal performance for heat transfer applications. Hence, this paper comprises the details of the methodology for reviewing resources, results, and discussion for achieving its objectives.

2. Methodology

2.1 Resources
Scopus and Web of Science are used as main databases, given that both databases are reliable and cover more than 256 fields of study like engineering. In particular, Scopus indexes a total of 4551 journals and Web of Science indexes a number of 108 journals related to engineering.

2.2 The systematic review process of articles selected
In selecting a number of relevant articles for the present study, the systematic review process consists of identifying keywords, followed by the search process for related and similar terms. Accordingly, search strings on Scopus and Web of Science databases were developed in October 2019 (Refer Table 1). A total of 20 articles from both databases were successfully retrieved from the current research work.

In the beginning stage of screening, seven duplicated articles were removed. A total of 13 papers were screened in the second screening stage based on several parameters for inclusion and exclusion. The first requirement was chosen to focus only on the source type of journals and conference proceedings as it acts as the primary source providing empirical data. Hence, the conference review is excluded. To improve the probability of retrieving similar articles, papers published in the field of engineering were selected. Additionally, the review concentrated only on articles published in English. Therefore, on the basis of these requirements, a total of two publications were omitted.

A total of 11 articles were prepared for the eligibility stage. At this point, on a more important note, the titles abstracts, and the main components of all the articles were investigated thoroughly to ensure that the criteria are met to be used in this review, thereby achieving the goals of the current research. Consequently, two articles were excluded due to the journal is not open access and out of heat transfer scope. Finally, a total of nine articles remaining are ready for analysis. The summary of the methodology is presented in figure 1.

Table 1. The search string used for the systematic review process.

Database	Search strings
Scopus	TITLE-ABS-KEY ((“heat transfer” OR “thermal analysis” OR “thermal perform*”) AND (nanocellulose OR “cellulose nano*”) AND nanofluid*)
WoS	TS= ((“heat transfer" OR "thermal analysis" OR "thermal perform*”) AND (nanocellulose OR "cellulose nano*”) AND nanofluid*)

3. Results
The review comes up with four main themes and six sub-themes related to the nanocellulose preparation and heat transfer performance. The four main themes are preparation method, stability evaluation, thermophysical properties, and thermal performance. The results provided a comprehensive analysis of the preparation and thermal performance of nanocellulose for heat transfer applications as shown in Table 2.

A total of two pieces of literature has been conducted on the study thermophysical properties of nanocellulose [2, 3], three literature on cutting tools coolant [4-6], three works of literature on car radiator [7-9] and one literature on pool boiling [10]. Furthermore, all studies are based on the experimental type of research. Regarding the years published, five were published in the year 2019, two papers are published in 2018 and 2017.

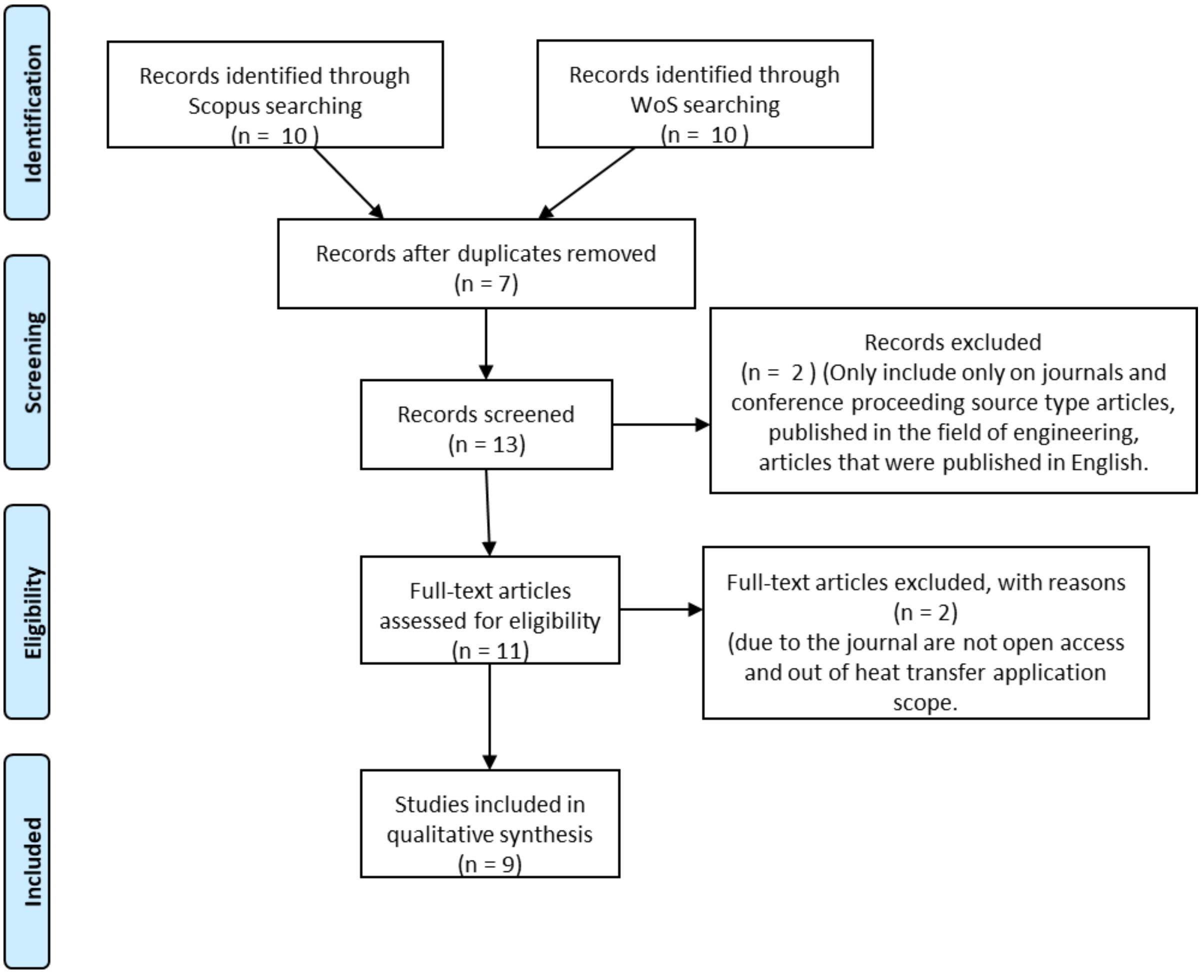

Figure 1. The flow diagram of the study.

Table 2. Table of findings.

Authors	Preparation method		Stability evaluation		Thermophysical properties		Thermal performance
	Conc (%)	SS	Ql	Qt	TC	Vis	
Ramachandran et al. (2017)[2]	0.1-1.3	√		√	√	√	
Ramachandran et al. (2017)[3]	0.1-0.9	√	√	√	√	√	
Anamalai et al. (2018)[4]	0.1-1.3	√		√	√	√	√
Kadirgama et al. (2017)[5]	0.1-1.3	√		√	√	√	√
Samylingam et al. (2018)[6]	0.1-1.5	√	√	√	√		√
Naiman et al. (2019)[7]	0.5	√		√	√		√
Benedict et al. (2019)[8]	0.1-1.3			√			√
Benedict et al. (2019)[9]	0.5	√		√			√
Hwang et al. (2019)[10]	2	√		√			√

Conc = Concentration Ql = Qualitative analysis TC = Thermal conductivity
SS = Stability strategies Qt = Quantitative analysis Vis = Viscosity

3.1 Preparation methods
The preparation of nanocellulose nanofluid involves either a one-step method or two-step method, the value of concentration used and strategies to increase the stability of the nanofluid. Most of the study prepared their nanocellulose nanofluid by using the two-step method, except one study by Hwang et al [10]. Cellulose nanocrystals (CNC) from Blue Goose Biorefineries Inc had mostly purchased in their study [2-9]. To maintain the stability of nanocellulose in based fluid, most of the study used the

ultrasonic vibration strategy to enhance the dispersion of nanocellulose in the based fluid. Ultrasonic vibration consists of the ultrasonic bath and ultrasonic probe.

For the two-step method, the purchased nanocellulose which in gel form was first converted from weight concentration to volume concentration using Eq. 1. Then, the volume needed for dilution, ΔV, used for preferred concentration, ϕ2, can be determined by using Eq. 2 with the original condition of V1 and ϕ1. Then, the nanofluid is stirred by using a magnetic stirrer for 30 minutes for well mixing. Later, the prepared solution is left in the ultrasonic bath for 2 hours. Preparation of 2% nanocellulose nanofluid by using the one-step method had been successfully done by using the TEMPO-mediated oxidized method by Hwang et al [10]. Each individual fiber had a width of 5˜10 nm and a length of several microns.

$$\varphi = \frac{w\rho_{bf}}{\left(1-\frac{w}{100}\right)\rho_{np} + \frac{w}{100}\rho_{bf}} \tag{1}$$

$$\Delta V = (V_2 - V_1) = V_1\left(\frac{\varphi_1}{\varphi_2} - 1\right) \tag{2}$$

3.2 Stability evaluation

Stability evaluation is used to identify and to ensure that the prepared nanocellulose nanofluid is high-quality terms of dispersion of nanocellulose in the based fluid. There are two types of evaluations which are qualitative evaluation and quantitative evaluation. The qualitative evaluation consists of sedimentation analysis which had been done by all studies. Most of the prepared nanocellulose nanofluids were stable for a month or two without any agglomerate and sediment through sedimentation analysis.

Quantitative analysis is used to demonstrate that nanofluid is scientifically stable, such as using the spectrophotometry method's absorbance drop evaluation [3, 6]. Ramachandran et al [3] found that for 1.0% of nanocellulose, the absorbance drops are 8.38% and 12.5% for 0.5 and 0.6 ethylene glycol volume base ratio accordingly. For 0.9%, absorbance drops by 1.24% and 1.83% for 0.5 and 0.6 base ratio respectively. Furthermore, Samylingam et al [6] found that at 0.1% concentration of nanocellulose, the absorbance drops after one month is 5.51%. Nanofluid with an absorbance value decreased by less than 30% as a stable nanofluid [11]. Furthermore, a higher concentration of nanocellulose has more absorbance drop.

3.3 Thermophysical properties

Thermophysical properties are studied to identify the properties of nanocellulose for heat transfer applications. There are two types of reported properties had been done which are thermal conductivity [2-7] and viscosity [2-5]. Thermal conductivity and viscosity studies had been done with respect to temperature and concentration by using KD2 Pro and rheometer.

For the study of thermal conductivity, thermal conductivity is found to increase as the temperature and nanocellulose concentration in the base fluid increases. Other than that, Kadirgama et al [5] noted that 0.5 vol percent nanocellulose nanofluid has a superior thermal conductivity of 0.449 W / m K than 0.267 W / m K thermal conductivity of traditional coolant fluid at 30 ° C. Next, four literatures had studied on viscosity [2-5]. They found that the viscosity decreases with the increases in temperature and increases with the increases in volume concentration.

3.4 Thermal performance

All studies had done the thermal performance analysis for respective heat transfer applications except [2, 3] which focus on the thermophysical properties study only. There are three applications involved, which are cutting tools coolant [4-6], car radiator [7-9] and pool boiling [10]. For the cutting tool's thermal performance, nanocellulose nanofluid has a lower maximum temperature reading recorded at chip compared to the conventional fluid. In the car radiator application, as the concentration on nanocellulose increases, the heat transfer coefficient also increases except for [8] due to stability issues. In the pool boiling application, the critical heat flux (CHF) increases as the concentration of nanocellulose increases. It was recorded that CHF enhances by 40.7%, 45.1%, 54.9%, and 69.4% for 0.01, 0.03, 0.05, and 0.10 wt%.

4. Discussion

This study attempts to systematically discuss the existing literature related to the preparation and heat transfer performance of nanocellulose. Nanocellulose is one of the nanoparticles that exhibit eco-friendly properties and high heat transfer performance compared to conventional fluids. Four main themes and 6 sub-themes have been discussed in this section.

The preparation of nanofluid needs to be good to minimize the stability issue of nanofluid. The two-step method is used in most of the literature as it is much simpler and more economics compared to the one-step method. Unfortunately, Yu and Xie [12] stated that the two-step method has lower stability of nanofluid compared to the one-step method. The one-step method cannot synthesize nanofluids on a large scale and high cost. It is proven that the nanocellulose nanofluid prepared by Hwang et al [10] has a higher concentration compared with others. Most of the literature avoid using higher concentration due to the stability issue and could cause clogging during operation [13]. Furthermore, the stability strategy that has been used in the literature is ultrasonic vibration. Ultrasonic vibration emits supersonic waves travel longitudinally within the liquid medium and caused the alternate positive and negative pressure waves in the liquid [14]. Thus, this will increase the stability of nanofluid. In the literature, only two to three hours of vibration is needed to prepare a stable suspension of nanocellulose nanofluid due to its high stability. Furthermore, nanocellulose need ultrasonic vibration only to fully dispersed in a based fluid. Other nanofluids reported in literature need both ultrasonic vibration and surfactant reaction to fully dispersed in a based fluid. Therefore, the preparation of nanocellulose nanofluid is more cost-effective compared to other nanofluids.

Next, stability evaluation needs to be done to measure the stability of nanofluid prepared. From the reported literature reviews, there is quantitative and qualitative analysis. Quantitative analysis involves sedimentation analysis where most of the prepared nanocellulose nanofluids were successfully stable for 3 to 6 months which can be considered high stability. This analysis shows that nanocellulose has a lower tendency to agglomerates and sediment in the based fluid. Next, qualitative analysis is to measure the stability of nanofluid scientifically. Two literatures used Ultraviolet-Visible spectrophotometry method to study the absorbance drop throughout time. Although the higher concentration of nanocellulose has more absorbance drop, all concentration has lower than 30% absorbance which can be considered high stability after one month. The effect of the adjacent particle phenomenon causes the based fluid to generate an upward flow which pushes the nanoparticle and prevents it from falling due to gravity acceleration as stable nanofluid is achieved [15].

Based on six literatures study on thermal conductivity, nanocellulose has better thermal conductivity compared to the based fluid. Furthermore, a higher concentration of nanocellulose and temperature, higher the thermal conductivity due to the Brownian motion. As the temperature increases, the molecules in the nanofluid would start to collide with each other due to high kinetic energy [16]. The higher the kinetic energy, the higher the thermal conductivity properties of the fluid. This shows that nanocellulose can increase the heat transfer performance of the based fluid. Next, based on the four literatures study on the viscosity of nanocellulose nanofluid, it is observed that the viscosity decreases as the temperature increases due to enhanced movement between atoms and particles at subatomic level [17]. However, the viscosity increased as the volume concentration increase is due to the increased liquid interior shear pressure [4].

Next, seven literatures of using nanocellulose in heat transfer application show the enhancement of heat transfer performance compared to the respective based fluid. This is because the nanocellulose nanofluid has higher thermal conductivity compared to the based fluid. High concentration promotes better heat transfer performance due to Brownian motion that increases the molecules contact caused by high kinetic energy [18]. Furthermore, due to this phenomenon, nanocellulose nanofluid has decreased the maximum temperature of cutting tools and increases the heat transfer coefficient and CHF enhancement. This shows that nanocellulose is proven to be an alternative to increase the heat transfer performance of the based fluid.

5. Conclusion
This review has highlighted the preparation of nanocellulose and its heat transfer performance in heat transfer applications. Based on the review that has been done, nanocellulose is suitable to be used in heat transfer applications based on its preparation, stability of nanocellulose in based fluid, thermal properties, and thermal performance of past applications. Due to its high stability and thermal properties of nanocellulose, nanocellulose can be used to increase the heat transfer performance of the based fluid. Furthermore, this paper only highlighted the application of nanocellulose in cutting coolants, car radiators, and pool boiling.

6. Future outlook
Further studies that can be done to explore the heat transfer performance of nanocellulose are using various types of based fluid to dispersed the nanocellulose. This is because other based fluid has differents thermal enhancement rates using nanocellulose. Furthermore, other heat transfer applications such as in electronic cooling or renewable technology sectors can be done to identify the thermal performance of nanocellulose. More simulation work using nanocellulose as a coolant can be studied to verify the thermal performance of nanocellulose.

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