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Preparation Methods and Challenges of Hybrid Nanofluids: A Review



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ARTICLE INFO	ABSTRACT
Article history: Received 16 December 2019 Received in revised form 22 October 2020 Accepted 22 October 2020 Available online 6 December 2020	The recent studies on nanotechnology have reported rapid development of nanofluids in various aspects due to the enhanced thermophysical and heat transfer properties of nanofluids. This paper reviews the preparation methods and some challenging issues of hybrid nanofluids during the Preparation of hybrid nanofluids. One-step and two- step are mainly the preparation methods of hybrid nanofluids. Compared to the one- step method, the two-step method is a widely used technique for preparing nanofluids due to its simplicity, whereas this technique has a complexity of achieving stability of hybrid nanofluids. On the contrary, the one-step is very flexible for achieving uniformity of nanofluids with comparatively high production cost. Some researchers followed various techniques such as surfactant addition, surface treatment, and pH modification for preparing a durable nanofluid. However, these methods also have their limitation, such as degrading the thermal attributes of hybrid nanofluids. So, future studies need to address these challenges along with the cost analysis during preparing the hybrid nanofluids.
Keywords:	
Nanoparticle; hybrid nanofluid;	
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1. Introduction

The demand for effective heat transfer fluid increases with the progress in thermal engineering. Although conventional working fluids such as water and various kinds of oil have been using in various industrial applications, the thermal conductivity of these fluids is very low. Hence, in the advanced thermal engineering devices, these traditional fluids do not maintain standard heat exchange rates. However, recent studies come up with a solution suspending various kinds of solid particles with

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exciting thermophysical properties into traditional fluids [1-6]. The earlier research of suspension of the millimetre and micrometre-sized particles have demonstrated the growing difficulty of pressure drops, particle agglomeration and often equipment erosion. Later on, the researchers include nanometer-sized particles into various fluids which called nanofluid [7-9]. The previous studies showed improvement in terms of dispersibility avoiding much agglomeration and also in thermophysical properties which are expected attributes for better heat transfer applications [10-13]. However, the inclusion of hybrid nanoparticles into the traditional fluid, which is called hybrid nanofluids showed superior thermophysical properties than single nanofluid. In this case, the chemical and physical attributes of composite nanoparticles play a combined influential role to improve the thermophysical properties. Depending on the necessity of a specific application, several combinations of particle materials and fluids are of possible interest of recent nanofluid studies. For example, traditional fluids such as water, various kinds of engine oils, ethylene glycol and propylene glycol can be mixed with various types of nanoparticles such as metallic, non-metallic and carbides [14-17].

Various researchers experimented using various kinds of hybrid nanofluids and showed exciting properties with no adverse effect on pressure drop [18]. For instance, Chen and Ding [19] surveyed the findings on improving the thermal conductivity of various materials and concluded that, with the concentration of nanoparticles, the thermal conductivity of hybrid nanofluids increases linearly. TiO2-SiO2/ water-EG nanofluids revealed 13.8% thermal conductivity enhancement at 70 °C Hamid *et al.,* [4]. Esfe *et al.,* [20] reported 20.10% thermal conductivity improvement at 50 °C while investigating with SiO₂-MWCNT/ EG hybrid nanofluids. In different applications, enhanced thermal conductivity is an important factor for better performance [21]. Like the thermal conductivity, other properties such as viscosity, density, heat transfer coefficient also influenced due to the inclusion of nanoparticles into the base fluid. Esfe *et al.,* [22] found the maximum 83% viscosity enhancement of TiO₂-MWCNT/ water-EG at 0.85% concentration and 10 °C temperature. Besides, the authors also concluded about the Newtonian behaviour of TiO₂-MWCNT hybrid nanofluids for concentrations of 0.05, 0.45 % and non-Newtonian behaviour at 0.85% concentration.

However, for these mentioned impressive properties, the uses of nanofluids in various sectors such as automobile [23], electrical and electronics [24] and biomedicine [25, 26] have been rising dramatically. These hybrid nanofluids can be synthesized following various preparation methods. Depending on the synthesis method, the attributes of nanofluids can be altered. For example, the stability of the hybrid nanofluids is significantly related to the preparation method [27, 28]. Similarly, the thermophysical properties of nanofluids are directly correlated with stability. Hence, the preparation process plays an essential role in the study of nanofluid. However, there is not much systematic review of the synthesis and challenges of nanofluid study in the available literature. The present article, therefore, comprehensively study preparation methods of different nanofluids with difficulties and challenges.

2. Preparation Method of Hybrid Nanofluids

The hybrid nanofluids can be synthesized mainly by one-step or two-step method [12, 27-29]. In the one-step process, the preparation of the nanoparticles and dispersion in the base fluid is done in parallel [30, 31]. The most important advantage of this technique is the reduction of the possibility of agglomeration of nanoparticles [30]. It is, however, difficult to cover it up for manufacturing purpose on a mass scale for higher cost [32, 33], which restricts the implementation of this method [34-37]. Two-step technique is initiated by preparing the nanoparticles and then diffuses them in an appropriate liquid. Under this technique, the high surface energy of nanoparticles leads to



aggregation and eventually sedimentation of nanoparticles deteriorating stability of nanofluid [27]. The two-step approach is used for mass scale processing due to the ease and relatively low cost [28]. Among these two techniques, the two-step method is widely used by researchers and also industrialists [1, 4, 38]. Besides, by Choi and Eastman [7] particularly advises this approach for producing oxide-based nanofluids rather than nanofluids based on metallic nanoparticles. However, various studies incorporated these two techniques for preparing various kinds of nanofluids.

For instance, distilled water-based Fe₃O₄-CNT hybrid nanofluids were synthesized following onestep method Shahsavar et al., [39]. In this study, tetra-methylammonium hydroxide (TMAH) and Gum Arabic (GA) were used to coat Fe₃O₄ and CNT nanoparticles, respectively. Similarly, Jaiswal et al., [40] also followed the single-step method for synthesizing Cu- Pd hybrid nanofluids. Two-step method was applied for water-EG based TiO₂- Cu hybrid nanofluid with the inclusion of SDBS surfactant for achieving the long term durability Khairul et al., [41]. The study also used magnetic stirring and ultrasonication process and eventually found 11% improvement of thermal conductivity. EG based MWCNT- Fe3O4 hybrid nanofluid was also prepared by following two-step method Harandi et al., [42]. In this study, both nanoparticles were mixed in the same proportion, and moderate stability was observed for all samples after stirring and sonication. Afrand et al., [43] and Ramachandran et al., [44] also employed this two-stage technique for Fe₃O₄-Ag/EG and Al₂O₃-CuO/ water hybrid nanofluids, respectively. Similarly, thermal oil-based MWCNT- Al₂O₃ hybrid nanofluid was also synthesized using two-step technique during investigating the thermal properties in the temperature range of 25-50°C and concentration range of 0.125 to 1.50% Asadi et al., [45]. Urmi et al., [46] investigated the thermophysical properties of 40 % EG based TiO₂-Al₂O₃ hybrid nanofluids while using the two-step method. The study included magnetic stirring for mixing the nanoparticles into the base fluid and ultrasonication for getting the durability. Again, TiO₂- SiO₂/ water- EG based hybrid nanofluids were also prepared by Hamid et al., [47] using the same technique for various concentrations and mixing ratios of nanoparticles. This double-stage approach for synthesizing carbon nanotubes containing nanofluids was also followed by several other researchers [48-51].

3. Various Kinds of Hybrid Nanofluids

This section presents a comparative discussion on the various kinds of studied nanofluids in detail and also highlight the issues of preparation procedures. Researchers have been working on various kinds of hybrid nanofluids and their diversified applications in engineering fields.

3.1 MgO-Based Hybrid Nanofluids

For instance, distilled water-based Ag-MgO hybrid nanofluids were prepared by Esfe *et al.*, [52] while the sizes of Ag and MgO nanoparticles were measured by XRD analysis and the sizes were 25 nm and 40 nm respectively. During the study with Ag-MgO hybrid nanofluids, achieving stability was one of the significant contributions. For this purpose, three hours of ultrasonication with the inclusion of Cetyl Trimethyl Ammonium Bromide (CTAB) was conducted. In this study, the concentration of CTAB was very minimum to avoid any interruption in the nanofluids thermophysical properties. Finally, the hybrid nanofluids achieved several hours of stability.

In another study, MgO was mixed with single-walled carbon nanotube (SWCNT), and their mixing ratio was 80:20 (MgO: SWCNT) into the ethylene glycol [53]. The nanoparticles are stirred using a magnetic stirrer to homogenize solution, but the particles appeared agglomerated. Hence, 6 hours of ultrasonication were done and finally, MgO-SWCNT hybrid nanofluids showed three days of stability. Similarly, 80% MgO was suspended into the motor oil (SAE 40) with 20% multi-walled carbon



nanotube (MWCNT) Asadi *et al.*, [54]. The MgO-MWCNT/ SAE 40 hybrid nanofluids showed stability for over two weeks while 2 hours of stirring and 1 hour of sonication was conducted to achieve stability.

3.2 TiO2-Based Hybrid Nanofluids

Hamid *et al.*, [55] prepared water- EG based TiO₂-SiO₂ hybrid nanofluids for various mixing ratios of nanoparticles such as 20:80, 40: 60, 50: 50, 60:40 and 80:20. The authors followed to one hour of mixing and two hours of ultrasonication and found the suspension of nanofluid is stable for almost two weeks. The stability period was confirmed by measuring the absorbency of hybrid nanofluids. Among all the samples of nanofluids, the TiO₂-SiO₂ hybrid nanofluids consisting of 20% TiO₂ and 80% SiO₂ showed maximum improvement of thermophysical properties.

3.3 Al₂O₃-Based Hybrid Nanofluids

Senthilraja *et al.*, [56] also followed the two-step method for synthesizing distilled water-based Al_2O_3 -CuO hybrid nanofluids. At the same time, one hour of magnetic stirring and four hours of ultrasonication was done to achieve durability, but the results showed the prepared nanofluid was stable for several hours. In another study, TiO₂- $Al_2O_3/40\%$ EG hybrid nanofluids were prepared for assessing the stability [57]. The study also conducted magnetic stirring and ultrasonication for getting homogeneity of nanofluids. The results showed that the 80:20 mixture ratio of TiO₂: Al_2O_3 hybrid nanofluids showed maximum stability period for almost three weeks. Among all the blending ratios (20:80, 40: 60, 50: 50, 60:40, 80:20) of nanoparticles, 80:20 showed highest zeta potential value also which proved that the optimum mixing ratio is also a critical issue in the study of nanofluid. Later on, for the investigation on the TiO₂- Al_2O_3 (80:20) /40% EG hybrid nanofluids, Urmi *et al.*, [46] proved that ultrasonication time influenced the durability of nanofluids noticeably. The study was conducted on the various concentrations of nanofluids such as 0.02, 0.04, 0.06, 0.8 and 0.1%. However, the findings revealed that the 7 hours of ultrasonication without any additives could give the highest stability period showing the absorbency of more than 70 %.

3.4 Graphene-Based Hybrid Nanofluids

Like other metallic, non-metallic and oxide-based nanoparticles, graphene hybrid nanofluids also showed exciting improved thermophysical properties. Increased thermal conductivity and higher mechanical strength and electrical conductivity of graphene particles have attracted the scientist in recent years [58]. Besides, it is reasonably simple and price-effective to produce graphene nanoparticles. Due to different methods used to produce one layer or multi-layer graphene, a small variation in graphene properties has been documented, such as graphene oxide layer exfoliation, chemical vapour deposition and mechanical cleavage [58-60]. An experimental investigation has shown that even a single layer of graphene has higher thermal properties than CNT. However, one of the main problems to be solved is the dispersion of graphene with standard stability. Thus, it may be able to prepare stably dispersed graphene-based hybrid nanofluids by using the functionalization process (acid treatment and amino function), proper ultrasonic and solvent [61, 62]. For example, carbon nanotube nanofluid revealed moderate stability after applying simple acid treatment Wen *et al.*, [63].



In another study, graphene nanoparticles were mixed with HNO₃ and H_2SO_4 (1:3) and then dried after washing through distilled water. Finally, the nanoparticles are dispersed into Ag (NO₃) OH solution, which gives 60 days stability period [64]. Hence, the nanofluids can achieve uniformity by converting it slightly acidic or basic [63]. Most significantly, there is a crucial need to preserve the value of the solution's pH in the appropriate range as the highly acidic and basic suspension is not suitable for staff as well as the work environment. Besides, surface corrosion may also place in the equipment using this alkaline and acidic solution [65]. Finally, Table 1 represents the various types of available studied hybrid nanofluids, while Table 2 presents a summary of the preparation methods of hybrid nanofluids. The next section describes various kinds of hybrid nanofluids comprehensively.

Table 1				
Classification of the hybrid nanoparticles				
Type of materials	hybrid nanoparticles			
Metal matrix	Al ₂ O ₃ /Cu			
	Al ₂ O ₃ /Ni			
	MgO/Fe			
	AI/CNT			
	Al ₂ O ₃ /Fe-Cr			
	ND/Ni			
Ceramic matrix	Al ₂ O ₃ /SiO ₂			
	Al ₂ O ₃ /TiO ₂			
	CNT/Fe ₃ O ₄			
	Al ₂ O ₃ /SiC			
	Al ₂ O ₃ /CNT			
	SiO ₂ /Ni			
Polymer matrix	Polymer/layered double hydroxides			
	Polymer/CNT			
	Thermoplastic/thermoset polymer/layered silicates			
	Polyester/TiO2			

Table 2

Summary of preparation methods of hybrid nanofluids

Preparation	Nanoparticles	Base Fluid	Other information during	References
Methods	-		Preparation	
One-Step	Fe ₃ O ₄ -CNT	Distilled water	TMAH and GA coating	Shahsavar <i>et al.,</i> [39]
One-Step	Cu-pd		PVP	[40]
Two-Step	TiO ₂ - Cu	Water-EG	SDBS surfactant	Khairul <i>et al.,</i> [41]
Two-Step	MWCNT- Fe ₃ O ₄	EG		Harandi <i>et al.,</i> [42]
Two-Step	Fe ₃ O ₄ -Ag	EG		Afrand <i>et al.,</i> [43]
Two-Step	Al ₂ O ₃ -CuO	Water		Ramachandran <i>et al.,</i>
				[44]
Two-Step	MWCNT- Al ₂ O ₃	Thermal oil	Stirring+ Ultrasonication	Asadi <i>et al.,</i> [45]
Two-Step	TiO ₂ -Al ₂ O ₃	40% EG	Stirring+ Ultrasonication	Urmi <i>et al.,</i> [46]
Two-Step	TiO ₂ - SiO ₂	Water- EG	Stirring+ Ultrasonication	Hamid <i>et al.,</i> [47]
Two-Step	Ag- MgO	Distilled water	Ultrasonication+ CTAB	Esfe <i>et al.,</i> [52]
Two-Step	Al ₂ O ₃ –CuO	Distilled water	Stirring+ Ultrasonication	Senthilraja <i>et al.,</i> [56]



4. Challenges of Hybrid Nanofluids

Investigation on the emerging heat transfer nanofluid is a fascinating work in the field of nanotechnology. From detail reviewing the literature, it is found that Preparation of standardized suspension remains a methodological challenge; meanwhile, the nanoparticles always form agglomeration because of the substantial van-der-Waals interactions effect. Mostly, the long term stability of nanoparticles dispersion is one of the elementary requirements of hybrid nanofluids applications. The stability of hybrid nanofluids has a good conforming bond with the improvement of thermal behaviour such as the better the dispersion capability, the better the thermophysical attributes of hybrid nanofluids [66]. Consequently, the thermal conductivity, as well as the viscosity of the hybrid nanofluids, is finally affected. Studies have shown that over time, thermal conductivity is decreased [67], which is due to the reduced dispersion capability of nanoparticles with time. Hence, nanoparticles can appear to sediment when it has been preserved for an extended time.

However, the physical and chemical treatment was typically accompanied such as applying an external force on the clustered colloidal nanoparticles, surface modification of the nanoparticles, the addition of surfactant and pH modification, to obtain the stable nanofluids. From the literature, it is shown that as an external force, stirring and ultrasonication can be done. However, the suitable time for stirring and ultrasonication for various nanofluids need to be addressed. Because most of the studies focus only the short study period as nanofluids stability period, but without getting long term stability of nanofluids, it is a hurdle to commercialize the applications of nanofluids. Besides, in case of surfactant addition, proper selection, quantity and function of additives for respective nanofluid is also a need for nanofluid study as some additives degrade the thermophysical properties of nanofluids and start to generating foam at high temperature by degrading the quality of nanofluids [27, 68]. Also, studies on the corrosive behaviour of surfactants and nanoparticles need to be carried out maintaining proper pH value, as no research focuses on this issue. More significantly, there is no report on the total pricing of various forms of nanofluids. Uses of nanofluid instead of large amount traditional fluids may simplify the size and cost of equipment. However, some types of nanoparticles Preparation expenses a lot. Hence, the quantitative expenses of varied categories of nanofluids with the required equipment in different engineering fields can be an exciting area that can be discussed in future research.

5. Conclusions

The review article has attempted to represent the methods of synthesization of hybrid nanofluids from the recent available literature. The recent scientist has been focusing mostly on hybrid nanofluid rather than single nanofluids due to significant increase in thermal conductivity, viscosity, friction factor and pumping power. While many preparation schemes have been developed, branding a uniform and long-term stable hybrid nanofluid with insignificant agglomeration without hindering the thermophysical properties is indeed a challenging issue. A single-step method is ideal to the Preparation of more advanced and durable hybrid nanofluids, but a two-step method can generate a high amount of hybrid nanofluid. The two-step process is suitable for preparing a high volume hybrid nanofluids with low cost. In order to promote heat transfer efficiency of hybrid nanofluids, surfactants may play a vital role in proper dispersion of nanoparticles as nanoparticles cannot be distributed uniformly in the base fluids. Like surfactants, other methods such as surface and pH modification can also be a prosperous solution for achieving a durable solution. In this case, a cautious attempt is needed to choose additive, the proper way to nanoparticles treatment. Because the attributes of nanofluids can be altered due to applying these methods, which can also be harmful



to the system and environment. For example, during pH modification, it is vital to keep the pH value safe both for equipment and also the worker. However, most of the available studies only focus on a few hybrid nanofluids and their thermophysical properties. More studies are required on different kinds of hybrid nanofluids focusing various kinds of preparation methods and most importantly highlighting the facts of achieving long term durability and also economic analysis of applying nanofluids in practical fields which leads to the easy way to commercialize the nanofluids in practical.

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