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1 **Advanced applications of tunable ferrofluids in energy systems and energy harvesters: a**
2 **critical review**

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10
11 **Abstract**

12 Ferrofluids or Magnetic nanofluids (MNFs) are the suspensions of magnetic nanoparticles and
13 non-magnetic base fluid. The heat transfer performance of a magnetic nano-suspension is
14 influenced by the strength and orientation of an applied magnetic field. The main attraction of
15 these types of nanofluids is that they not only enhance the fluids' thermophysical properties
16 but also possess both magnetic characteristics like the other magnetic materials and flow ability
17 similar to any other fluids. Such an exclusive feature enables to control heat transfer, fluid flow
18 and movement of the nanoparticles by using the external magnetic fields. This review paper
19 summarises the recent investigations of magnetic nanofluids with the aim of identifying the
20 effects of major parameters on the performance of heat transfer. In addition, this study also
21 acknowledged the novel application of ferrofluids in the electromagnetic energy harvesters,
22 and its challenges as well as the potentiality in the future research.

23

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24 **Keywords:** Magnetic nanofluid; ferrofluid; electromagnetic energy harvester; stability; heat
 25 transfer enhancement; electromechanical systems (MEMS)

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47

48 **Nomenclature**

A	constant	Re	Reynolds number
C_p	specific heat capacity ($J/kg K$)	T	temperature (K)

d	sample size (m)	V	velocity (m/s)
\vec{f}	vector sum of the gravitational body force (N/m^3)	Greek symbols	
g	gravitational acceleration (m/s^2)	φ	particles volume fraction
g^*	effective acceleration (m/s^2)	ρ	density (kg/m^3)
Gr	total potential energy (J)	$\Delta\rho$	density difference (kg/m^3)
h	liquid height (m)	μ	viscosity ($N s/m^2$)
H	magnetic field (A/m)	μ_0	vacuum permeability (H/m)
k_B	Boltzmann constant (J/K)	β	relative coefficient
K	thermal conductivity ($W/m \cdot K$)	γ	system dependent coefficient
M_0	Magnetisation (A/m)	Subscripts	
Pr	Prandtl number	f	base fluid
ΔP	pressure drop (Pa)	L	liquid
R	radius of the container (m)	m	magnetic
R'	radius of the spherical particle (m)	p	particle
Ra	Rayleigh number	T	thermal

49

50 1. Introduction

51 Currently, cooling is one of the most important scientific challenges in production related
52 industries, such as transportation, manufacturing and microelectronics. Technological
53 advancements have led to increased thermal loads and thus, improvements to cooling systems
54 have become a necessity. Maximising the surface area of heat exchanger systems is the
55 conventional approach to enhance heat dissipation. However, this method needs an unwanted
56 rise in the size of thermal management systems, thus, there is an urgency for novel coolants
57 with enhanced performance [1]. One such coolant is the innovative concept of ‘nanofluids’,
58 which are a mixture of metallic/nonmetallic nanoparticles in a base fluid. The term nanofluid
59 was originally introduced by Choi at Argonne National Laboratory [2]. A substantial
60 improvement in liquid thermal conductivity, specific heat and viscosity are the unique features
61 of nanofluids. The relatively large overall surface area of nanoparticles not only improves heat

62 transfer capabilities but also increases the stability of the suspension by alleviating particle
63 settling phenomenon. There are also several potential benefits from nano-suspension testing,
64 specifically: better long-standing stability compared to the millimetre or even micrometre sized
65 particle suspensions and lower erosion and pressure drop, especially in micro-channels [3].
66 Nanofluids are very potential fluids for heat transfer application [4-7].

67 A unique class of nano-suspensions named magnetic nanofluids demonstrate both fluid and
68 magnetic properties. Magnetic nanofluids have drawn considerable attention due to the
69 possibility of tuning their heat transfer and flow properties through the application of an
70 external magnetic field [8, 9]. The nanoparticles can be either ferromagnetic materials, such as
71 cobalt or iron, or ferrimagnetic materials such as magnetite. Magnetite nanoparticles are not
72 susceptible to oxidation and therefore are a far better alternative to iron or cobalt nanoparticles
73 which tend to lose their magnetic characteristics over time because of oxidation [10].
74 Magnetite-based nanofluids with particle sizes less than 10 nm, known as ferrofluids, were
75 initially introduced by Stephen Pappell in the 1960s (at NASA) as an advanced method for
76 controlling fluid in space [11]. He concluded that magnetite nanofluids have a wide range of
77 applications from lubricating rotary shafts to biomedicine. These types of nanofluids have
78 improved thermal properties (such as heat capacity, thermal conductivity, viscosity etc.), as
79 well as magnetic properties, both of those tunable characteristics help to control the heat
80 transfer and the movement of the particle by applying the magnetic fields. As a result, they are
81 believed to be one of the promising fluids in different engineering fields such as
82 bioengineering, thermal engineering, electronics, and energy harvests [12, 13].

83 However, when subjected to an external magnetic field, the thermal conductivity of magnetite
84 nanofluid can be raised to levels much higher than any other nanofluid. In one such example,
85 Philip et al. [14] have shown remarkable enhancements in thermal conductivity of up to 300%

86 for a magnetite based nanofluid. This high rise in thermal conductivity is associated with the
87 effective heat conduction through the chain-like structures induced in the magnetite nanofluid.
88 The advantage of using a magnetically polarisable nanofluid, such as magnetite nanofluid, is
89 that the size, shape and form of aggregates can be precisely controlled by the external magnetic
90 field. More importantly, unlike other nanofluids, the aggregation observed in magnetite
91 nanofluids is perfectly reversible due to the super-paramagnetic nature of particles [15]. This
92 tunable nature offers great opportunities for resolving the inherent problems associated with
93 conventional nanofluids, such as lower heat transfer capacity, clogging and blockage of the
94 flow passage. The ferrofluid with magnetic fields in the application of heat transfer is
95 considered as the compound heat transfer method [16]. The magnetic nanofluids offer the
96 following advantages compared to the nonmagnetic nanofluids [17];

97 (a) the temperature gradient and non-uniform magnetic field are induced by using a
98 magnetic field, which may initiate a flow in the fluid. Such phenomenon is called
99 thermomagnetic convection [18] and it is readily handled;

100 (b) the thermomagnetic convection is higher compare to the gravitational convection;

101 (c) the possibility of changing thermal properties of ferrofluids by applying external
102 magnets/solenoids [19, 20]

103 Furthermore, a ferrofluid is a suspension of solid-liquid, made-up from nano-sized permanent
104 magnetic dipoles [21]. The magnetic nanofluids are potential fluids for vibrational energy
105 harvesting application due to their fluidity and magnetic properties in which they act as a soft
106 ferromagnetic substance [22, 23]. Energy harvesting is an alteration of the environmental
107 energy to the electrical energy at a small scale and was originally introduced in 1966 [24]. The
108 energy sources for energy harvesters are freely available in the environment. Examples of
109 energy sources include vibration, wind energy, wave energy, and thermal temperature

110 gradients. Nowadays energy harvesting has become a hot topic for research because of its
111 exceptional advantages. The cost of the battery supply, in particular, the maintenance cost to
112 replace the discharged batteries, make the energy harvesters a very attractive option. The
113 conventional energy harvesting techniques only capable to generate a few micro-watts,
114 whereas vibration energy harvesters demonstrate high performance in less vibration by
115 replacing the solid magnets with ferrofluid.

116 Recently magnetite nanofluids have come to the attention of the research community after
117 showing high enhancement in heat transfer by applying external magnetic field as well as their
118 usage in the electromagnetic energy harvesters to generate more power with a small vibration
119 with compare to the traditional harvesters. The objective of this paper is to concentrate on the
120 recent investigations of magnetic nanofluids in the application of heat transfer and energy
121 harvesting. It is anticipated that this review paper will help to provide a clear idea about the
122 current status of ferrofluid, and also specify the recommendations for the future study.

123

124 **2. Preparation and characterisation of magnetic nanofluids**

125 The magnetic nanofluids are prepared by dispersing of super-paramagnetic nanoparticles into
126 a non-magnetic base fluid such as water, hydrocarbon oil and so on [25]. Generally, the
127 preparation of ferrofluids consist of two steps: (a) the magnetic nanoparticles preparation, (b)
128 the dispersion of the synthesised magnetic nanoparticles in different polar/non-polar carrier
129 liquids. In the first step, the nano-sized magnetic particles can be produced by various processes
130 such as ball milling [26], sonochemically synthesised [27], sol-gel method [28, 29], reverse
131 micelle technique [30], and thermal decomposition [31]. Moreover, the metal oxide magnetic
132 nanoparticles are synthesised by micro-emulsion, chemical co-precipitation and phase transfer

133 methods [25, 29, 32-34]. In the second step, the magnetic nanoparticles are coated by various
134 methods such as co-precipitation [29], core-shell [35], and then dispersed in the base fluids.

135 Currently some researches have completed experiments to synthesise metal and metal oxide
136 magnetic nanoparticles with a specific size distribution [32]. Metallic nanoparticles were
137 prepared using different techniques such as the thermal decomposition of metal carbonyl
138 complexes, the simple reduction of metal salts, thermolysis of metal-polymer complexes,
139 submerged arc nanoparticle synthesis system, and gas phase reduction of metal complexes [32,
140 36]. Among the metal oxide nanoparticles, Fe_3O_4 , $\gamma\text{-Fe}_2\text{O}_3$ and spinel type ferrites (MFe_2O_4 ,
141 with $\text{M} = \text{Mn, Co, Zn, Ni, etc.}$) are most commonly used nanoparticles because of their chemical
142 stability. The most efficient method for the ferrite nanoparticles and subsequent magnetic fluid
143 preparation is chemical precipitation [37]. The preparation of magnetic nanofluids consist the
144 following steps;

- 145 • co-precipitation ($\approx 80\text{ }^\circ\text{C}$) of magnetite from aqueous solutions of Fe^{3+} and Fe^{2+} ions in
146 solutions of sodium hydroxide (NaOH), ammonium hydroxide (NH_4OH) or potassium
147 hydroxide (KOH) [38]; and ammonium hydroxide has better crystallinity, smaller size
148 and higher magnetic saturation among them [39]
- 149 • subdomain magnetite particles
- 150 • steric stabilisation (chemisorption of lauric acid, myristic acid or oleic acid; $80\text{--}82\text{ }^\circ\text{C}$)
- 151 • phase separation, and magnetic decantation as well as repeated washing
- 152 • mono-layer covered magnetite nanoparticles + free surfactant
- 153 • extraction of mono-layer covered magnetite nanoparticles (acetone addition;
154 flocculation)

155 Cobalt ferrite and Fe_2O_3 nanoparticles were prepared using NaOH instead of NH_4OH .

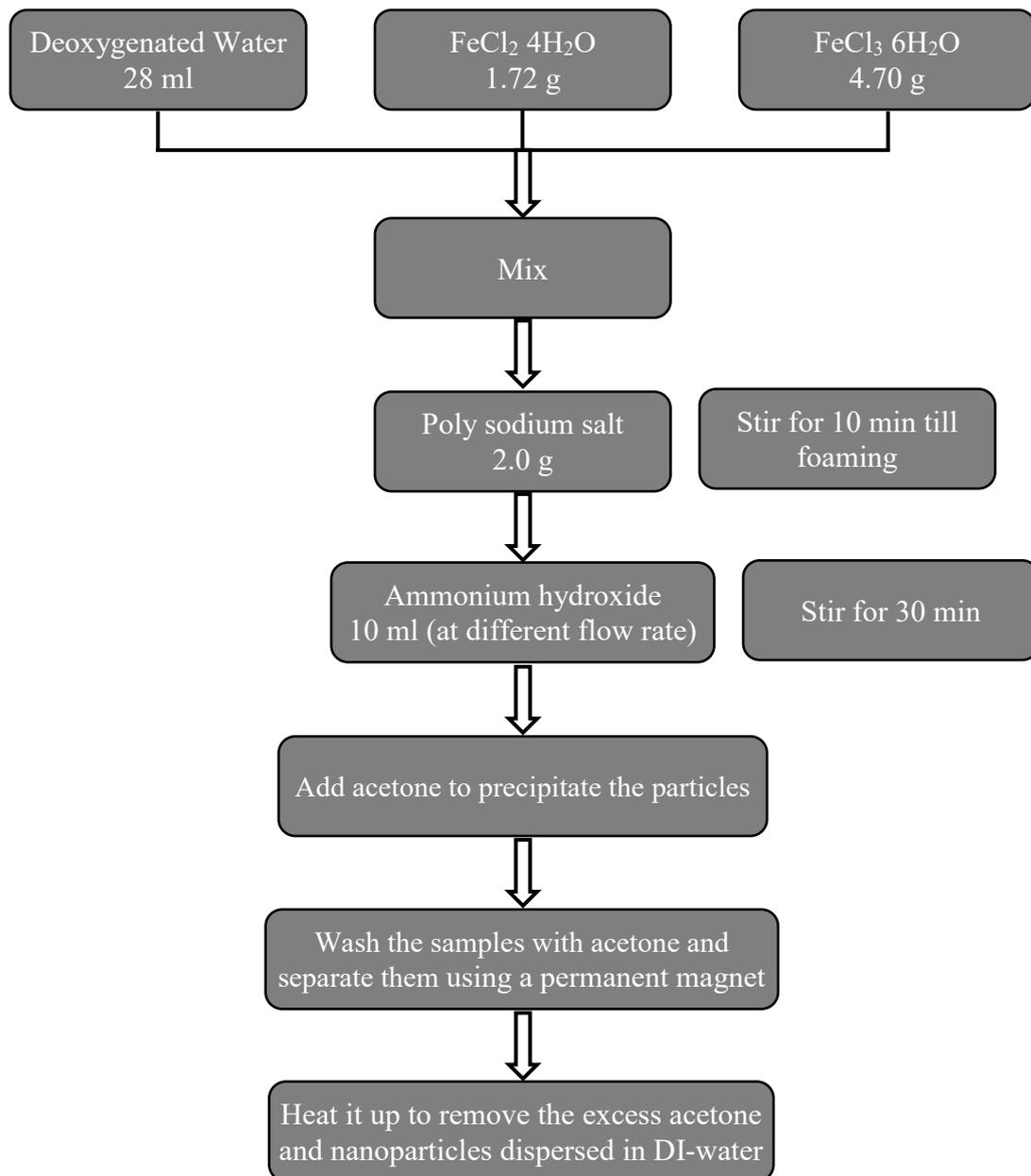
156 Finally, the dispersion of lauric acid, myristic acid or oleic acid mono-layer coated synthesised
157 magnetic nanoparticle in non-polar carriers (such as kerosene, toluene, cyclohexane,
158 transformer oil, etc) are demonstrated as follows;

- 159 • the dispersion process of nanoparticles in non-polar carriers carried out at the temperature
160 of 120-130 °C [40]
- 161 • magnetic decantation/filtration; and repeated flocculation and redispersion of magnetic
162 nanoparticles for the elimination of free surfactant called advanced purification process

163 whereas, the dispersion method of magnetic nanofluids in organic polar carriers such as
164 alcohol, diester, ketone, amine, different mixtures of mineral and synthetic oils involves;

- 165 • primary magnetic fluid on light hydrocarbon carrier
- 166 • repeated flocculation and redispersion of magnetic nanoparticles to eliminate the free
167 surfactant known as advanced purification
- 168 • mono-layer stabilised magnetic nanoparticles
- 169 • dispersion in a polar solvent (stabilisation with a secondary surfactant, e.g.
170 dodecylbenzenesulphonic acid, physically adsorbed to the first layer)

171



172

173

174

Fig. 1. Methodology of ferrofluids production [34]

175 The details of the multi-step procedures applied to prepare the magnetite nanofluids at the

176 University of Newcastle described in Fig. 1 [34], which were refined and optimised for the

177 synthesis of magnetic fluids for various applications, such as vibration based energy harvesters

178 and as a highly efficient heat transfer fluids. The magnetic nanoparticles and nanofluids are

179 characterised using different techniques and methods, as shown in Table 1.

180 **Table 1.** Physicochemical characterisation of magnetic nanoparticles

Characteri- sation	Method and technique	Measured parameter
Geometry	Transmission Electron Microscopy (HRTEM) and Field Emission Scanning Electron Microscopy (FESEM) [41-43]	Size and the shape of magnetic particles
	Dynamic Light Scattering (DLS), Quasielastic Light Scattering (QELS) [43, 44] X-ray Diffraction and Mössbauer Spectroscopy [45]	Mean particle size and size distributions
Structure	X-ray diffraction, thermal analysis, and Mössbauer and infrared spectroscopy [44]	Nanoparticle structure
	Thermogravimetric and differential thermogravimetric analysis (TGA), Differential Scanning Calorimetry (DSC), Fourier Transform Infrared spectroscopy (FTIR) and Statistic Secondary Ion Mass Spectra (SSIMS) [45-47]	The structures of composite particles made of iron oxide and organic molecules The nature of the interactions between the iron oxide core and the associated molecular structures within the composite
	Atomic Force Microscopy (AFM) and Chemical Force Microscopy (CFM) [48]	The morphological changes occurring on the surface of iron oxide nanoparticles upon exposure to a coating material
Colloidal Stability	Dynamic Light Scattering (DLS) [49, 50]	The colloidal stability is based on the hydrodynamic particle size
	Turbidity measurements [51, 52]	Stability of magnetic nanoparticles by measuring the aggregation kinetics

181

182 3. Stability of magnetic nanofluids

183 A nanofluids' stability relies on a number of aspects [53]: a) nanofluids are multiphase
184 dispersion systems with high surface energies and hence, are thermodynamically unstable, b)
185 nanoparticles dispersed in the nanofluids have strong Brownian motions. The nanoparticles'
186 movement can offset their sedimentation due to gravity, c) the dispersed nanoparticles in the
187 fluids may settle out with time because of nanoparticle aggregation, which is initiated by van
188 der Waals forces, d) no chemical reactions among the suspended nanoparticles nor between the

189 base fluid and nanoparticles are expected. Consequently, aggregation and sedimentation are
190 the two critical phenomena relating to the stability of a nanofluid.

191 The sedimentation of the particle in the magnetic nanofluid may occur due to the effects of
192 magnetic field, gravitational field and/or magnetic field gradient, because of the external
193 magnetic field is directly associated with the size distribution of magnetic nanoparticles [54].

194 The sedimentation of the particle has a significant impact on the stability of magnetic
195 nanofluids. The stability against the particle sedimentation can assure when the thermal energy
196 of the nanoparticles goes higher than the gravitational and magnetic energies, accordingly. The
197 maximum size of the nanoparticle was calculated by using the Odenbach [54] formulas;

198 $d < (6k_B T / \mu_0 M_0 \pi H)^{1/3}$ in the presence of magnetic field and $d < (k_B T / \Delta \rho g h \pi)^{1/3}$ under

199 the gravitational field, where $k_B, T, \mu_0, M_0, H, \Delta \rho, g$ and d denote the Boltzmann constant,
200 temperature, vacuum permeability, the spontaneous magnetisation of the magnetic material,
201 magnetic field, density difference between nanoparticle and the base fluid, gravitational
202 acceleration, and size of the sample, respectively. In principle, the particle aggregation rises
203 the active diameter, hence a destabilisation of the suspension occurs due to the sedimentation.

204 In this case, the maximum diameter (d) of the particle was estimated as,

205 $d < (144k_B T / \mu_0 M_0^2)^{1/3}$ corresponding to the maximum interaction of energy [54].

206 The use of super-paramagnetic particles in magnetic nano-suspension is not necessarily
207 definitive of the stability of magnetic nanofluids [55]. Magnetic nanoparticles in a base fluid
208 will not be stable because of the existence of London-van der Waals and magnetic forces,
209 contributing to the irreversible aggregation of nanoparticles. Thus, magnetic nanofluids
210 introduce the repulsive forces between the magnetic nanoparticles to repel the dipole-dipole
211 magnetic interactions and London-van der Waals force. The repulsive force between the
212 nanoparticles can be attained by using a polymer surfactant as a coating around the particles,

213 which could produce an entropic repulsion, and/or by introducing a coulombian repulsion from
214 the variation of the nanoparticle surface [55, 56]. Generally, the dispersion process of the
215 magnetic nanofluids is carried out by ultrasonic homogenization method in the presence of a
216 surfactant.

217 Finely divided iron is very sensitive in the presence of water or humid air and oxidising agents.
218 Therefore, protection of magnetic nanoparticles is the prior requirement to obtain chemically
219 and physically stable colloidal systems, and such protection may be attained by surface coating
220 around the nanoparticle as shown in Fig. 2 [57]. A couple of methods have been suggested to
221 attain stable nanofluids, such as chemical or physical treatment. These treatments may involve
222 the modification of the dispersed nanoparticle surface, the addition of an extra surfactant, or
223 enforcing strong forces on the agglomerated nanoparticles. The active surface agent was
224 applied for the modification of hydrophobic materials, thus allow dispersion in an aqueous
225 solution [58]. The chemical treatment changes the suspension stability through the resultant
226 surface potential, as well as surface charge states [59]. Otherwise, sedimentation, aggregation,
227 and clogging may occur and hamper the thermal properties of the nanofluids. It is well known
228 that the theory of aggregation and clustering is one of the primary causes of stability and
229 unexpected increase of nanosuspensions thermal conductivity [60]. Philip et al. [14] and Evans
230 et al. [60] showed that the greater aspect ratio structure of the fractal-like aggregates is a
231 fundamental factor that allowed for enhanced heat flow over large distances.

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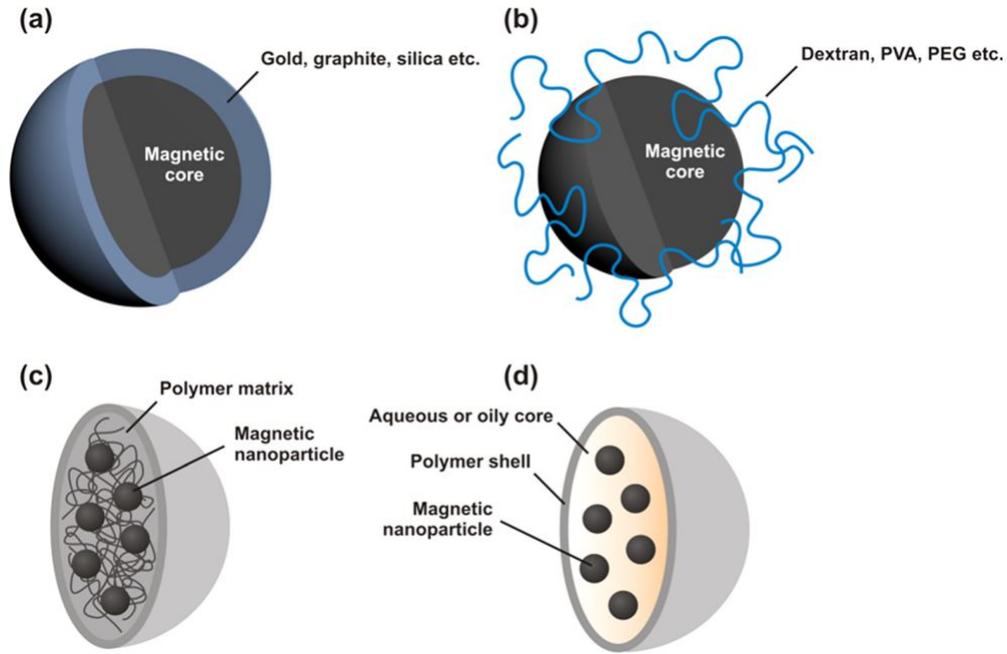


Fig. 2. Schematic representation of the stabilisation of magnetic nanoparticles by surface coating (a) inorganic material, (b) organic material, (c) encapsulation into nanospheres, (d) nanocapsules [57]

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238 Normally, nanoparticles need to fulfill the two principles in order to gain a high-quality
239 suspension, the diffusion principle as well as the zeta potential principle [61]. The stability of
240 a colloidal suspension is highly influenced by its zeta potential value or surface charge [62]. It
241 is well known that the dividing line between stable and unstable suspensions is generally
242 around ± 30 mV and nanoparticles with a zeta potential more negative or positive than ± 30 mV
243 accordingly are considered physically stable [53, 63]. In addition, zeta potential of a colloidal
244 suspension is highly dependent on the pH value of the solution [7].

245 In a stationary state, a nanoparticles' sedimentation velocity in a nano-suspension complies
246 with Stokes Law [64]:

247
$$V = \frac{2R^2}{9\mu}(\rho_p - \rho_L)g \quad (1)$$

248 where V is the sedimentation velocity of nanoparticles, R' is the radius of spherical particle, μ
249 is the viscosity of the liquid medium, ρ_p and ρ_L are the density of particle and the liquid medium,
250 respectively and g is the acceleration of gravity. Eq. (1) exposes a balance of the viscous drag,
251 buoyancy force and gravity, those are actively influencing the dispersed nanoparticles. The
252 following actions might be helpful to reduce the sedimentation speed of nanoparticles in nano-
253 suspension, and henceforward to produce an improvement in nanofluids stability: (a) reducing
254 nanoparticles size/radius, (b) increasing the viscosity of the fluid medium and (c) minimising
255 the difference between nanoparticle and base fluid density. It is obvious that the reduction in
256 particle size should significantly reduce the nanoparticles sedimentation speed, as a result
257 enhance the nanofluids' stability, since V is proportional to the square of R . According to the
258 colloid chemistry theory, when the nanoparticle size reduces to a critical size, there is no
259 sedimentation due to the Brownian motion of the particle [65]. However, nanoparticles have
260 the possibility to aggregate due to its higher surface energy. Therefore, the preparation of a
261 stable nanofluid is associated with using the smaller nanoparticles to prevent the aggregation
262 process concurrently [65].

263

264 **4. Mechanism of heat transfer enhancement using magnetic nanofluids**

265 Numerous methods have been identified on the mechanism of the heat transfer enhancement
266 in the different studies. The thermal properties especially the thermal conductivity of magnetic
267 nanofluids was the main focus of some investigations, but the mechanisms to justify the
268 experimental data in the presence and absence of external magnetic field are still desirable. The
269 Brownian motion [66-69], liquid layering on the particle-liquid interface, and the effects of
270 nanoparticles clustering is the much debated among all of the proposed heat transfer
271 mechanisms [14, 69].

272

273 *4.1 Brownian motion*

274 Brownian motion can be expressed as a random motion induces from the collision of the base
275 fluid molecules, consequently particle experiences a random walk motion [70, 71]. This
276 parameter has a significant impact on the heat transfer and flow features of nanosuspensions
277 [72]. Generally, nanofluids containing smaller particles are offering more improvement of heat
278 transfer with compare to the bigger particles [73]. The Brownian motion can enhance the heat
279 transfer in two different ways, firstly, the motion of the nanoparticles help to transport the heat
280 (diffusion of nanoparticles) through direct contribution and the indirect contribution because
281 of the micro-convection is acting on the surrounding of the individual nanoparticles [71, 74].
282 Some authors [68, 75] were recommended that the thermal conductivity is controlled by
283 Brownian motion at nano and micro levels. The thermal conductivity and particle motion rise
284 with the increment of temperature [68]. Besides, the motion of nanoparticles due to the
285 Brownian motion was noticeably slow in heat transfer, because a particle needs to travel
286 massive distances to achieve a perfect place and this distance may be a shorter one.
287 Consequently, the random motion could not be a significant phenomenon in heat transfer.

288 Current analyses refused the hypothesis of Brownian motion, reporting that the enhancement
289 of thermal conductivity of magnetic nanofluids can be described from the theory of particle
290 clustering [14, 20]. Philip et al. [20] presented that the micro-convection model overvalued the
291 thermal conductivity, thus the micro-convection of the fluid medium around randomly moving
292 nanoparticles did not have any effect on nanofluids thermal conductivity. The diffusion of
293 magnetic nanoparticles exhibits a vital role at low nanoparticle volume concentrations ($\phi < 2$
294), which can be described by the effective medium (Maxwell) theory instead of the mechanism
295 related to the Brownian motion induced hydrodynamics. The improvement in thermal

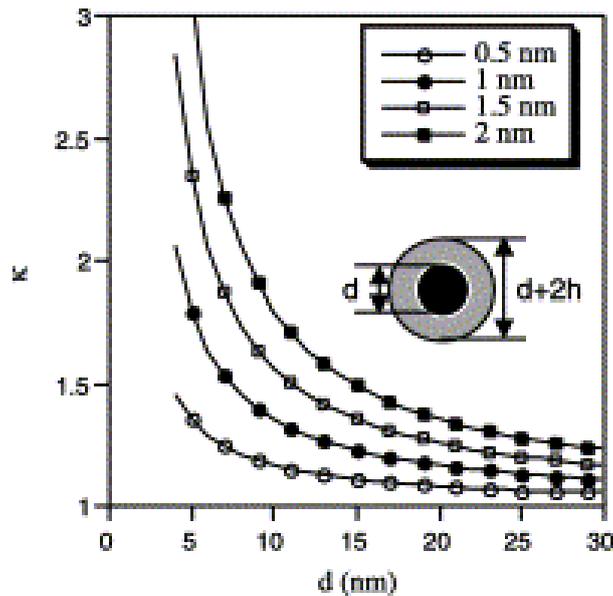
296 conductivity of magnetic nanofluids at the high-volume fraction ($\phi > 2$) was associated with
297 the presence of dimmers or trimmers in the field. These experimental results had a reasonable
298 agreement with the Maxwell-Garnet model, especially at a greater nanoparticle volume
299 concentrations [20]. Tsai et al. [76] examined the effect of viscosity on the thermal conductivity
300 of magnetic nanofluids. They concluded that the experimental value of the nanofluids thermal
301 conductivity progressively matches the value of thermal conductivity predicted by using the
302 Maxwell equation. In addition, diffusion of magnetic nanoparticles could be one of the
303 potential reasons for thermal conductivity improvement.

304

305 *4.2 Liquid layering on the nanoparticle-liquid interface*

306 Previous studies clearly showed that the liquid molecules may form layers around the
307 nanoparticle due to the strong force of the nanoparticle and the atomic structure of liquid layer
308 is more arranged compared with the bulk liquid. Since phonon transfers in crystalline solid is
309 very effective, such ordered layer in the liquid shows higher thermal conductivity and as a
310 result, the thermal conductivity of nanofluids was enhanced [77]. Koblinski et al. [78]
311 suggested that the resultant higher effective volume of the particle-layered-liquid structure can
312 improve the value of thermal conductivity (refer to Fig. 3). Yu et al. [79] explained the
313 existence of liquid molecules near to a solid surface. Ren et al. [80] confirmed the thermal
314 conductivity increment with this liquid layer, maximum 165% enhancement in thermal
315 conductivity was found for 3 nm liquid layer compared to 1 nm. Yu et al. [81] also clarified
316 the significance of the solid-liquid interfacial layers in the enhancement of nanoparticle (<10
317 nm) thermal conductivity.

318



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Fig. 3. Thermal conductivity improvement due to the formation of highly conductive liquid structure at liquid/particle interface for various layer thickness (h) as a function of particle diameter (d) [78]

324 4.3 Effects of nanoparticles clustering

325 Aggregation of nanoparticles is considered as one of the main mechanisms of heat transfer
326 enhancement using nanofluids, and nowadays it receives a wide interest of research community
327 [71, 82, 83]. The particle clusters may involve the fastest heat transport along the long distance,
328 because of the high heat conductivity of the solid particle compare to the base fluid [84, 85].

329 Bishop et al. [86] showed that the magnetic nanofluids have the magnetic interaction to
330 introduce a self-assembled aggregation even though in the absence of an external magnetic
331 field. The structures of aggregations of magnetic nanoparticles control the heat conduction in
332 the fluid. Without the application of the external magnetic field, the nanoparticles distribution
333 in the nanofluid is disordered and the thermal conductivity is isotropic [16]. Magnetic
334 nanoparticle tries to align its magnetic moments toward the direction of the local magnetic field
335 because of the external magnetic field or the neighbouring nanoparticles [54, 87]. This allows
336 magnetic nanoparticles to form micron-sized, one-dimensional chains, rings, two-dimensional

337 aggregates or even three-dimensional super lattices. The mechanisms of the aggregate
338 formation, manipulation of nanofluids and distribution morphology in the presence of external
339 magnetic fields are required to understand the thermal conduction mechanism of magnetic
340 nanofluids.

341

342 **5. An overview of thermal properties of magnetic nanofluids**

343 Investigations on magnetic nanofluids without the application of external magnet exhibit that
344 thermophysical properties of nanofluids are affected by different factors such as nanoparticle
345 size, types and intensities of magnetic fields, nanoparticle volume concentration, base fluid
346 properties, temperature, the chemical composition of the nanoparticle, the coating around the
347 nanoparticles etc. The properties of magnetic nanofluids and their effects on the volume
348 fraction of nanoparticle and temperature have been discussed in most of the studies. Syam
349 Sundar et al. [88] studied the viscosity and effective thermal conductivity of Fe₃O₄/water
350 nanofluids and showed that the nanofluid demonstrates Newtonian behavior, and the thermal
351 conductivity increases with the rise of nanoparticle volume fraction as well as temperature.
352 Pastoriza-Gallego et al. [89] concluded that a linear improvement of the thermal conductivity
353 with rising the particle volume concentration and the nanofluids was nearly temperature
354 independent. Magnetic nanofluids were prepared by dispersing magnetite nanoparticle in the
355 water using a surfactant called tetramethyl ammonium hydroxide, and the highest thermal
356 conductivity enhancement was found 11.5% with 3% volume concentration of nanoparticle at
357 40 °C [90]. Many studies have been focused on the thermal conductivity of nanofluids and
358 proposed various correlations to calculate its value, Table 2 shows the frequently used models
359 for the determination of the thermal conductivity of magnetic nanofluids.

360

361 **Table 2.** Summary of most common thermal conductivity models of magnetic nanofluids

Model	Expression	Description
Maxwell-Gannet model [91]	$\frac{K_p + 2K_f + 2\varphi(K_p - K_f)}{K_p + 2K_f - \varphi(K_p - K_f)}$	Spherical particles with low volume fraction φ
Modified Maxwell-Gannet model [92]	$\frac{K_i}{K_f} = \frac{K_p + 2K_f + 2\varphi_i(K_p - K_f)}{K_p + 2K_f - \varphi_i(K_p - K_f)}$	Spherical particles with volume fraction φ_i ; $i = x, y, z$
Microconvection model [93]	$\left(1 + A Re^\gamma Pr^{0.003} \varphi\right) \left[\frac{K_p + 2K_f + 2\varphi(K_p - K_f)}{K_p + 2K_f - \varphi(K_p - K_f)} \right]$	A is constant, Re and Pr represents Reynolds and Prandlt numbers, correspondingly, and γ denotes system dependent coefficient
Bruggeman model [94]	$\frac{K}{K_f} = \frac{1}{4} \left[(3\varphi - 1) \frac{K_p}{K_f} + (2 - 3\varphi) \right] + \frac{K_f}{4} \sqrt{\Delta}$ with $\Delta = (3\varphi - 1)^2 \left(\frac{K_p}{K_f} \right)^2 + 2(2 + 9\varphi - 9\varphi^2) \left(\frac{K_p}{K_f} \right)$	Spherical-sized particles with high volume fraction of the particle
Jeffrey model [95]	$\frac{K}{K_f} = 1 + 3\varphi + \varphi^2 \left[3\kappa^2 + \frac{3\kappa^3}{4} + \frac{9\kappa^3}{16} \left(\frac{\alpha + 2}{2\alpha + 3} \right) + \frac{3\kappa^4}{64} + \dots \right]$ with $\kappa = \frac{\alpha - 1}{\alpha + 2}$ and $\alpha = \frac{K_p}{K_f}$	High order terms represent pair interaction of randomly dispersed particles
Rayleigh model [96]	$\frac{K}{K_f} = K_f + 3\varphi \frac{K_p K_f}{2K_f + K_p \varphi [1 + 3.939\varphi^2 (K_p K_f) / 4K_f + 3K_p] K_p K_f}$	Suspensions of spherical particles with a regular particle distribution
Murshed model [97]	$K = \left\{ K_f \frac{Q_p \omega (K_p - \omega K_f) (2\gamma_1^3 - \gamma^3 + 1) (K_p + 2\omega K_f) \gamma_1^3 [Q_p \gamma^3 (\omega - 1) + 1]}{\gamma_1^3 (K_p + 2\omega K_f) - (K_p - \omega K_f) Q_p (\gamma_1^3 - \gamma^3 + 1)} \right\}$ $+ \left\{ Q_p^6 \gamma^6 K_f \left[3\Lambda^2 + \frac{3\Lambda^2}{4} + \frac{9\Lambda^3}{16} \left(\frac{K_{cp} + 2K_f}{2K_{cp} + 3K_f} \right) + \frac{3\Lambda^4}{2^6} \right] \right\}$ $+ \left\{ \frac{1}{2} \rho_{cp} C_{p-cp} d_s \left[\sqrt{\frac{3\kappa_B T (1 - 1.5\gamma^3 Q_p)}{2\pi_{cp} \gamma^3 r_p^3}} + \frac{G_T}{6\pi_{cp} d_s} \right] \right\}$ with $\gamma = 1 + \frac{t}{r_p}$, $\gamma_1 = 1 + \frac{t}{2r_p}$, $\Lambda = \frac{K_{cp} - K_f}{K_{cp} + 2K_f}$, $K_{Cp} = K_{lr} \frac{2(K_p - K_{lr}) + \gamma^3 (K_p + 2K_{lr})}{(K_{lr} - K_p) + \gamma^3 (2K_{lr} + K_p)}$	Murshed model considers the effects of nanolayer, size and movements of the particle, and surface chemistry of nanoparticles. C_{p-p} and C_{p-f} represent the specific heat capacity at constant pressure for nanoparticles and

ρ_{cp} and C_{p-cp} are density and specific heat of complex particle,

respectively, and they are given by $\rho_{cp} = \frac{1}{\gamma^3} \rho_p + \left(1 - \frac{1}{\gamma^3}\right)$

$$\left\{ \frac{3\rho_p}{pb^3} (2t^2 + 2br_p t + b^2 r_p^2) - \frac{3\rho_f}{pb^3} [(2 + 2b + b^2) + br_p (br_p + 2bt + t)] \right\}$$

$$C_{p-cp} = \frac{1}{\gamma^3} C_{p-p} + \left(1 - \frac{1}{\gamma^3}\right)$$

$$\left\{ \frac{3C_{p-p}}{pb^3} (2t^2 + 2b'r_p t + b'^2 r_p^2) - \frac{3C_{p-f}}{pb'^3} [(2 + 2b' + b'^2) + b'r_p (b'r_p + 2b't + t)] \right\}$$

where $p = 3r_p^2 + 3r_p t + t^2$; $b = \ln\left(\frac{\rho_p}{\rho_f}\right)$; $b' = \ln\left(\frac{C_{p-p}}{C_{p-f}}\right)$

base fluid,
respectively. Gr
denotes the total
potential energy
between two
interacting
nanoparticles.

362

363 5.1 Effects of nanoparticle size

364 A number of researchers have been investigated the effect of magnetic nanoparticle cluster and
365 size on the thermal properties of nanofluids. Zhu et al. [98] stated that the clustering, as well
366 as nanoparticle alignment was one of the main reasons for the irregular improvement of thermal
367 conductivity. They conclude that the Fe₃O₄ nanofluid with 4% volume fraction of nanoparticle
368 shows 38% enhancement in thermal conductivity. Jiang and Wang [99] synthesized a magnetic
369 nanofluid by one step phase transfer, showed that the particle coating on the surface of the
370 nanoparticle was an important parameter in the cluster formation. They concluded that the
371 thermal conductivity of magnetic nanofluid can be influenced by using a stabiliser, hence it
372 controls the structure of the aggregates. Besides, the thermal conductivity decreases in case of
373 well-dispersed composites. There is no recognisable standard for the superlative mix of
374 combining methods. The techniques are (a) the surfactant or activator addition, (b) pH control
375 (surface chemical effect), and (c) ultrasonic vibration [100]. According to the literature, three
376 effective techniques are applied to achieve suspension stability against nanoparticles
377 sedimentation. Some researchers used all of these tactics to achieve better stability [101-103]
378 but others applied just one [104] or two techniques [105-107] with satisfaction. Wang et al.
379 [108] experimentally examined the effect of nanoparticle size (4~44 nm) on the thermal

380 conductivities and exhibited that the magnetic nanofluids showed greater thermal
381 conductivities with compare to the heat transfer oils, and the enhancement of thermal
382 conductivity was found with reducing the size of the nanoparticle. The improvement in thermal
383 conductivity has showed up to 26.4% at 4 vol% of Fe_3O_4 nanofluids with 4 nm size of the
384 particle. Additionally, the values of viscosity for all nanofluids were significantly lower than
385 that of the carrier fluid. Hong et al. [109] investigated the effect of nanoparticles clustering for
386 a glycol based magnetic nanofluids in the absence of magnetic field, and it was found that
387 thermal conductivity decreased with sonication time. Thus, the size of the clusters formed by
388 the nanoparticles had a major influence on the thermal conductivity. The non-linearity was
389 attributed to the rapid clustering of nanoparticles in the condensed nanofluids. Moreover, the
390 carrier fluid effect on the ferrofluids properties was demonstrated in some analyses. Chen et al.
391 [110] suggested that the aggregation of nanoparticles was one of the main reasons for the
392 enhancement in viscosity of nanofluids beyond the classical Einestien equation. They proposed
393 their model by adding an aggregation effect to the Krieger and Dougherty [111] model, finding
394 good agreement between their experimental data and the proposed model.

395

396 *5.2 Effects of magnetic field*

397 Applying a magnetic field can affect the thermophysical properties of magnetic nanofluids, as
398 a result, some researchers were analysed the properties of magnetic nanofluids in the presence
399 of magnetic field. The effect of external magnetic fields and its directions and strengths on the
400 thermophysical properties of nanofluids have also been assessed. Results revealed that the
401 thermophysical properties of magnetic nanofluids are easily changeable by varying the strength
402 and direction of the magnetic field. Krichler and Odenbach [112] used an advanced measuring
403 device (a hot plate technique with a non-stationary mode) for measuring the thermal

404 conductivity of ferrofluid by changing the direction and strength of the magnetic fields. The
405 thermal conductivity improved with the increment of the magnetic field strength for parallel
406 alignment of the magnetic field and heat flux. On the other hand, the relation between the
407 thermal conductivity and magnetic field strength was totally opposite in the case of
408 perpendicular alignment. Therefore, the strength, parameters, and orientation of the external
409 magnetic field can also affect the properties of magnetic nanofluids because of the structure
410 formation of the magnetic particles in the magnetic nanofluid depends on the types of magnetic
411 fields. Azizian et al. [84] used electromagnets and solenoids to apply the parallel and
412 perpendicular magnetic fields to the temperature gradient, accordingly (as shown in Fig. 4);
413 and illustrated the influence of nanoparticle aggregation on the thermal conductivity of
414 nanofluids. They concluded that the maximum enhancement of thermal conductivity of
415 $\text{Fe}_3\text{O}_4/\text{water}$ nanofluid is 167% under external magnetic field configuration.

416

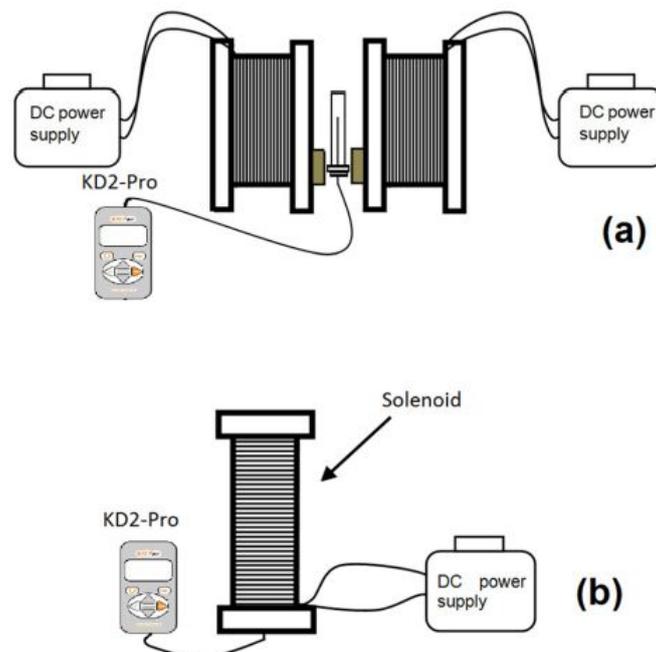
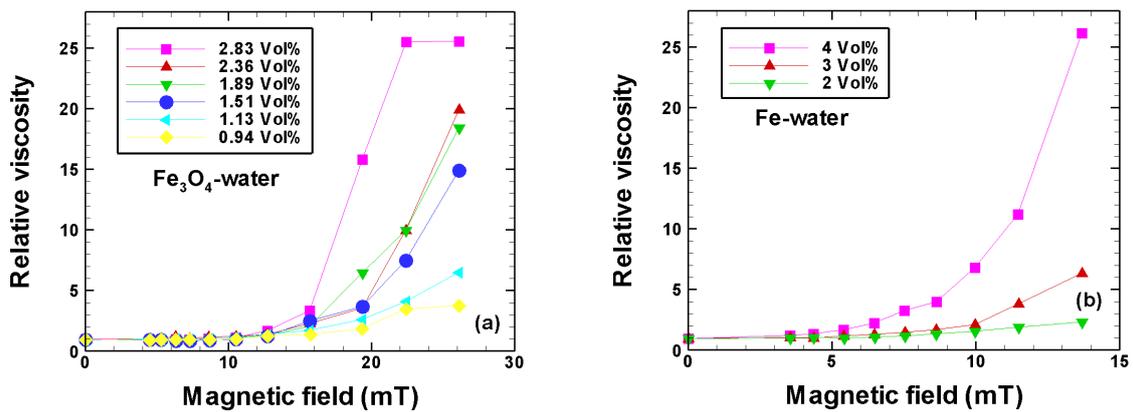


Fig. 4. Schematic diagram of (a) electromagnet and (b) solenoid

417
418
419

420 Li et al. [113] shows the enhancement in relative viscosity of aqueous Fe and Fe₃O₄ nanofluids
 421 by applying the magnetic field, as shown in Fig. 5. The higher viscosity of the magnetite
 422 nanofluid under an externally applied uniform magnetic field caused a reduction in fluid
 423 velocity.

424



425 **Fig. 5.** Relative viscosity as a function of perpendicular external magnetic field strength (a)
 426 Fe₃O₄/water, (b) Fe/water [113]

427

428 Shima and Philip [114] examined the effect of magnetic field on the thermal properties of
 429 magnetic nanofluids and showed that the thermal properties can be easily adjusted from very
 430 low to high values by changing the strength and orientation of the magnetic field. Baby and
 431 Ramaprabhu [115] investigated the effect of the application of magnets on the thermal
 432 conductivity of the magnetic nanofluid. It was confirmed that a small augmentation in thermal
 433 conductivity in the absence of magnetic field, whereas a significant rise in the thermal
 434 conductivity was noticed in the presence of magnetic field. They specified that the aligned
 435 nature of Fe₂O₃/MWNT toward the direction of the magnetic field may one of the possible
 436 reasons for the improvement of thermal conductivity under the application of magnetic field.
 437 Parekh and Lee [116] concluded that the enhancement in thermal conductivity of Mn-Zn and
 438 magnetic nanofluids exhibit 45% and 17%, respectively without the existence of the magnetic

439 field. However, the magnetite nanofluid demonstrated a further development in thermal
440 conductivity in the presence of a transverse magnetic field, although no variation was detected
441 for Mn-Zn ferrite. Gavili et al. [8] showed that the enhancement of thermal conductivity was
442 not significant in the absence of magnetic field. However, a maximum 200% improvement in
443 thermal conductivity of ferrofluid was observed under the effect of the magnetic field. Mehrali
444 et al. [117] investigated the thermophysical and magnetic properties of hybrid graphene/Fe₃O₄
445 ferro-nanofluid in the presence of a magnetic field, and thermal conductivity has shown an
446 development of 11%. Chiu et al. [118] analysed the influence of magnetic field and temperature
447 on the specific heat of a water based magnetic nanofluid. The specific heat did not depend on
448 the temperature under the lower magnetic field strength, but it decreased noticeably with the
449 rise of temperature in the presence of higher magnetic field strength. This may be due to the
450 reduction in the degrees of freedom of the ferrite nanoparticle in the magnetic nanofluid under
451 the higher magnetic field strength.

452

453 *5.3 Effects of chain-like structures of the magnetic nanoparticle*

454 Studies have been carried out on the thermophysical properties of magnetic nanofluids, and
455 their structural changes with the application of the external magnets, such as chain-like
456 structures formation, have been suggested as one of the efficient factors to enhance the heat
457 transfer. Nkurikiyimfura et al. [93] studied the influence of chain-like aggregation of magnetic
458 nanoparticle on the thermal conductivity of magnetic nanosuspensions in the presence of the
459 magnetic fields. They concluded that a significant enhancement of thermal conductivity with
460 the existence of the magnetic field parallel to the temperature gradient because of the
461 development of chain-like magnetic nanoparticle aggregates. Fu & Gao [91] introduced the
462 two-step homogenisation method for studying the anisotropic thermal conductivity of magnetic

463 nanofluids by considering the effect of physical anisotropy as well as the magnetic field. The
464 result showed that the aspect ratio of the chain-like aggregates of magnetic nanoparticle
465 exposed a vital role in the improvement of anisotropic thermal conductivity. Philip et al. [20]
466 found the highest thermal conductivity enhancement about 300% and 216% under the influence
467 of external magnetic fields for Fe₃O₄-kerosene and Fe₃O₄-hexadecane nanofluids, accordingly.
468 Such a large improvement was found due to the formation of chain-like structures of magnetic
469 nanoparticles. Parekh and Lee [119] also reported approximate 30% of thermal conductivity
470 increment at 4.7% of volume concentration of magnetite nanofluids in the presence of a
471 magnetic field and this may happen due to the continuous development of three-dimensional
472 zipper-like structures of nanoparticle inside the magnetic fluid.

473 Furthermore, the nanoparticle size, magnetic field as well as chain-like structure of magnetic
474 nanoparticle are the important parameters to change the thermal properties of magnetic
475 nanofluids in particle application. It is very important to consider the viscosity and thermal
476 conductivity of magnetic nanofluids at the same time. Because, Prasher et al. [120] have shown
477 that if the increase in viscosity becomes more than four times that of a comparable increase in
478 thermal conductivity of nanofluid, then the use of nanofluid is not economically viable.
479 Venerus et al. [121] also ran a benchmark study on the viscosity behaviour of ten different
480 nanofluids. They concluded that all of the nanofluids used in their study clearly failed the
481 qualifications of practical nanofluids as proposed by Prasher et al. [120].

482

483 **6. Convective heat transfer and pressure drop characteristics of magnetic nanofluids**

484 Nanofluids are dilute suspensions of functionalised nanoparticles and their objective is to
485 enhance the heat transfer performance of coolants/fluids, and nowadays has evolved into a
486 promising nanotechnological area [122, 123]. Although thermal conductivity of magnetic

487 nanofluids in the absence and presence of magnetic field has been the subject of many past
488 studies, relatively little effort has been focused on the convective heat transfer of magnetic
489 nanofluids. It has been proven that the thermal conductivity of magnetic nanofluids increases
490 under applied magnetic field parallel to the temperature gradient. If this is the case,
491 enhancement in heat transfer coefficient is expected. A review of studies related to the
492 convective heat transfer of magnetic nanofluid in the absence and presence of an external
493 magnetic field is provided in this section.

494 One of the main attractions of magnetic nanofluids application is the induction of a body force
495 due to the application of magnetic fields which allows the magnetic nanofluids to control the
496 fluid motion. The thermogravitation phenomenon may define as the natural movement of the
497 fluid because of the buoyancy force, where the reasons for the fluid motion are the presence of
498 temperature gradient and density difference. Moreover, these types of magnetic nanofluids are
499 temperature sensitive, and its magnetisation reduces with the rise of temperature. The
500 temperature gradient involves a non-equilibrium state in the magnetisation of the fluid under
501 the magnetic field. As a result, a net magnetic driving force induces, which may initiate a flow
502 in the fluid. Such phenomenon is called thermomagnetic convection.

503 When the approximation of equilibrium magnetisation with the magnetic susceptibility is only
504 depend on the local external magnetic field as well as the density then the gravitational and
505 magnetic forces are potential. The Kelvin body force produces a static pressure field in the fluid
506 flow, which is symmetric about the external magnetic field creating an irrotational force field.
507 This type of symmetric field does not change the velocity profile and the convection inside the
508 fluid could not rise [124]. The non-potentiality in the bulk forces is coming into the scenario if
509 a fluid experiences a non-uniformity of the magnetisation and density because of their
510 dependence on the volume fraction of nanoparticle or temperature [125]. Afterwards, the

511 condition is fulfilled for non-isothermal systems with an asymmetric temperature distribution
 512 about the applied magnetic field. The resultant Kelvin body force induces a field force, which
 513 contributes to the self-organised adjective motion of the magnetic nanofluid across isotherms.
 514 The condition for the free convection to develop is estimated by the following Eq. (2) [125]:

$$515 \quad \vec{\nabla} \times \vec{f} = \vec{\nabla} T \times \left[\beta_T \rho_o \vec{g} \pm \mu_o \beta_m \frac{\chi_o(H)}{2} \nabla H^2 \right] \neq 0 \quad (2)$$

516 where \vec{f} represents the vector sum of the gravitational body force and Kelvin body force; β_T
 517 denotes the relative volumetric expansion coefficient, the \pm sign means the anti-parallel and
 518 parallel alignment of the magnetic field gradient with respect to the gravitational force and β_m
 519 is the relative pyromagnetic coefficient of the fluid.

520 The intensity of the heat transfer is calculated by the Rayleigh number Ra , which is the
 521 combination of thermogravitational and thermomagnetic parts as follows;

$$522 \quad Ra = Ra_T + Ra_m = \frac{\rho c_p l^4}{\eta K} \frac{dT}{dz} \left(\beta_T \rho g + \mu_o \beta_m M \frac{dH}{dz} \right) \quad (3)$$

523 where Ra_T and Ra_m represent the thermal and magnetic Rayleigh number, accordingly. The
 524 efficiency of a device by applying the magnetic nanofluid gives the magnetic field, the
 525 temperature distributions fields as well as the tunable thermal properties of ferrofluid and the
 526 pyromagnetic coefficient. The pyromagnetic coefficient of a magnetic nanofluid is expressed
 527 as follows [17];

$$528 \quad \beta = \beta_T M + \left(\frac{\partial M}{\partial m} \right) \left| \frac{dm}{dT} \right| + \left| \frac{\partial M}{\partial T} \right| \quad (4)$$

529 where, thermal expansion coefficient, $\beta_T = -\frac{1}{\rho} \frac{d\rho}{dT}$, and ρ denote the density of magnetic
530 nanofluid. Almost all of the studies have been discussed the influence of temperature on the
531 magnetic nanofluids, which involves the dependency of magnetisation on the fluid temperature,
532 thus the existence of thermomagnetic phenomenon [18, 126, 127]. It is considered that the
533 thermal properties of ferrofluids are independent as well as constant under the external
534 magnetic field [18]. Though some of the inquiries have been concluded that the external
535 magnetic fields can influence the thermal properties of ferrofluids, thus has a significant impact
536 on heat transfer performance of nanofluids. The magnetic nanofluid varies the value of
537 viscosity under the application of magnetic field, known as the magneto-viscous effect.
538 Recently, some of the studies considered the effects of thermal properties on the enhancement
539 of heat transfer as well as pressure drop of magnetic nanofluids in the absence and presence of
540 the external magnetic field, shown in Table 3.

541

542 **Table 3.** Recent development on magnetic nanofluid as a heat transfer fluid

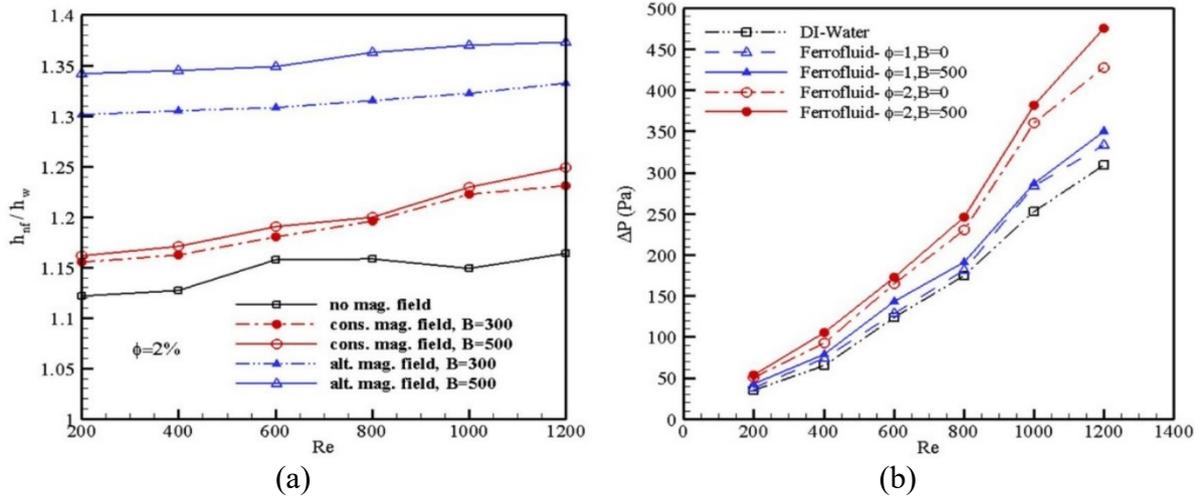
Author	Types of magnetic field	Result
Mehrali et al. [117]	Constant magnetic field	The local convective heat transfer coefficient increased up to 4% The thermal efficiency index was only 1 in the absence of the magnetic field and it was increased between 1.1 and 1.7 after applying a magnetic field. In addition, the total entropy generation rate was reduced up to 41% compared to distilled water
Goharkhah et al. [128]	Oscillating and constant magnetic field	The heat transfer coefficients were enhanced by 37.3% and 24.9% under alternating and constant magnetic field, accordingly. The rise in pressure drop under the alternating magnetic field was not significant compared to heat transfer enhancement

Goharkhah et al. [129]	Constant and alternating magnetic field	The average convective heat transfer grows up to 18.9% and 31.4% by application of constant and alternating magnetic field
Yarahmadi et al. [130]	Constant magnetic field with different magnetic field arrangements	The local convective heat transfer coefficient enhanced by 19.8% with $Re = 465$ and the concentration of 5%
Azizian et al. [131]	Constant magnetic field with different arrangements of magnets	The local heat transfer coefficient of magnetite nanofluids enhanced noticeably up to 300%, whereas, only 7.5% increment in the pressure drop under applied magnetic field
Ghofrani et al. [132]	Alternating magnetic field	A maximum of 27.6% enhancement in the convection heat transfer was observed
Sundar et al. [133]	Without magnetic field	The heat transfer coefficient and friction factor were improved 30.96% and 10.01%, respectively
Lajvardi et al. [134]	Constant magnetic field	The use of magnetic particles dispersed in distilled water cannot improve the convective heat transfer in the absence of magnetic field

543

544 Goharkhah et al. [128] studied the convective heat transfer and hydrodynamic features of
545 magnetite nanofluid under the influence of magnetic fields. The improvement of heat transfer
546 in the presence of the alternating magnetic field was compared with the heat transfer at the
547 constant magnetic field as well as in the absence of magnetic field configurations in Fig. 6a. In
548 the absence of magnetic field, a maximum 16.4% enhancement was found in magnetite
549 nanofluid compare to DI-water, although, it was enhanced up to 37.3% and 24.8% in the
550 presence of alternating and constant magnetic fields, correspondingly. Fig. 6b shows the
551 influence of magnetic field on the pressure drop of the magnetic nanofluid at different
552 nanoparticle volume concentrations. The pressure drop rises with the increase of ferrofluid
553 volume concentration and magnetic field because of the increase in fluid viscosity as well as
554 the tendency of chain alignment of magnetic particle in base fluid [128].

555

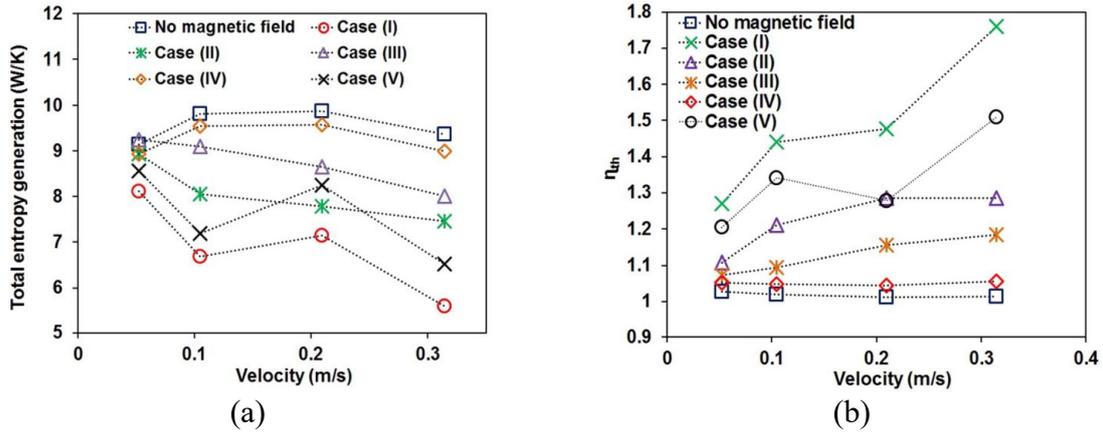


556 **Fig. 6.** The $\text{Fe}_3\text{O}_4/\text{water}$ nanofluid under different magnetic field (a) heat transfer
 557 enhancement versus Reynolds number, (b) pressure drop versus Reynolds number [128]

558

559 Mehrali et al. [117] analysed the hybrid graphene/ Fe_3O_4 ferro-nanofluid flow under the
 560 influence of a magnetic field. Fig. 7a shows that the total entropy generation reduces as the rise
 561 of fluid velocities and the effect of frictional entropy generations were low (the maximum value
 562 of frictional entropy generations were <1 in all cases). The effect of magnetic field
 563 arrangements on the thermal efficiency index is also shown in Fig. 7b. The maximum thermal
 564 efficiency index was only 1 in the absence of the magnetic field. After applying a magnetic
 565 field, this amount increased between 1.1 and 1.7. Therefore, use of hybrid magnetic nanofluids
 566 could be beneficial for improvement of heat transfer coefficient under the effect of magnetic
 567 fields.

568



569 **Fig. 7.** The rGO-Fe₃O₄ nanofluid for different arrangements of magnetic bars (a) total entropy
 570 generation rate versus velocity, (b) thermal efficiency versus velocity [117]

571

572 Li and Xuan [135] studied the heat transfer coefficient of magnetic nanofluids under the
 573 application of the external magnetic field in the laminar flow regime. The maximum Nusselt
 574 number enhancement of 40.6% under the application of an external magnetic field compared
 575 to the result in the absence of an external magnetic field.

576 From the above literature review, it is expected that the thermomagnetic convection and
 577 magneto-viscous effect should be assessed together and not separately in the future studies.
 578 Therefore, this may allow to understand the mechanism of the occurrence of thermomagnetic
 579 convection. Moreover, the uniform magnetic fields have been studied in the most of the current
 580 literature. Although the application of uniform magnetic field is essential and beneficial to get
 581 a clear idea about the mechanism of thermomagnetic convection, using electromagnets and
 582 Helmholtz coils to provide uniform magnetic fields looks uneconomical because of the
 583 consumption of the electrical energy. Thus, the phenomenon of thermomagnetic convection
 584 can utilise in a more economical and practical way by applying the magnetic fields using the
 585 permanent magnets. Such magnetic fields are more accessible as well as energy efficient. In
 586 addition, the pressure drop induced during coolant flow is a significant parameter to determine
 587 the efficiency of nanosuspensions. Different researchers have concluded that nanofluids have

588 a higher pressure drop than base fluids and as a result a notable rise in pumping power [136].
589 Hence, the heat transfer enhancement and pressure drop should be evaluated at the same
590 circumstances to characterise the magnetic nanofluids.

591

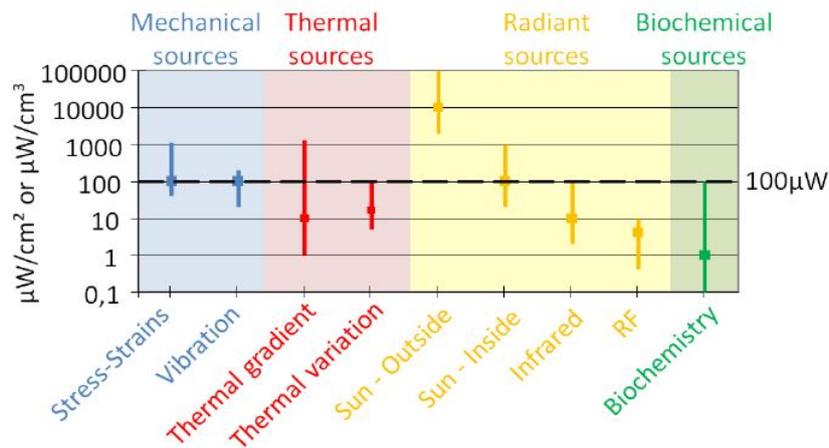
592 **7. Ferrofluid based energy harvester**

593 Over the current years, interest has raised in the development of micro-electromechanical
594 systems (MEMS) and miniaturised system. In this approach, there has been great effort to
595 decrease the energy consumption of these devices from being in the order mW to the order μW
596 [137]. Applications such as sensors in buildings, medical implants, wireless sensors networks,
597 sensors used in military applications, monitoring of the environment, sensors that may provide
598 different information about the maintenance requirements of various industrial equipment and
599 sensors for structural monitoring are just a few of the many examples [137, 138]. Currently,
600 batteries are used to supply the power to such systems, but some limitations of this are their:
601 limited lifetime, large size, difficulty in replacement and containing dangerous and hazardous
602 substances. An energy harvester can be utilised to improve the capability and lifespan of these
603 devices by swapping the usage of the batteries [139, 140]. Energy harvesting devices can use
604 to get vital information on structural and operational conditions by engaging them in remote
605 areas [141]. Hence, an energy harvester can be used to charge or replace the existing battery
606 and enhance the lifetime of the system [137, 142, 143].

607 Energy harvesting devices are classified based on the form of energy they use to scavenge
608 power. For example, piezoelectric harvesters scavenge mechanical energy and change it into
609 electrical energy. Primarily there are four types of ambient energy available in the environment:
610 thermal energy (temperature variations), radiant energy (RF, sun, infrared), mechanical energy
611 (deformations, vibrations) and chemical energy (biochemistry, chemistry). These sources are

612 characterised by different power densities as shown in Fig. 8 [144]. Fig. 8 demonstrates that
 613 the accessible power ranges from $10 \mu W$ to $100 \mu W$ and is a good order of magnitude for a $1cm^2$
 614 or a $1cm^3$ energy harvester. Apparently, $10-100 \mu W$ is a small amount of power; yet it can be
 615 adequate for various types of applications, in particular, wireless sensor networks.

616



617

Fig. 8. Power densities of ambient sources before conversion [144]

619

620 The mechanical vibrations are the most dominant and attractive ambient energy source because
 621 of its abundance [145, 146]. Unlike solar cells, a vibration energy harvester can supply the
 622 power during the day as well as the night, whether light is present or not. Here, the focus is on
 623 the vibration which can convert into electric power. Some vibration sources are available in
 624 the environment including electric motor rotation, wind energy, vehicle motion, wave energy,
 625 seismic vibrations and human movement all of which vary widely in both amplitude and
 626 frequency. Vibrational energy is found in most of the built environment [147]. The sources of
 627 vibration vary with the variation of dominant frequency and amplitude [148]. Roundy et al.
 628 [149] completed a number of measures of vibration sources [149] and concluded that the
 629 amplitude and frequency of the vibration sources differs from $12 ms^{-2}$ to $0.2 ms^{-2}$ at $200 Hz$

630 and 100 Hz, respectively. Most of the sources were measured to a fundamental frequency in
 631 the range of 50-200 Hz. Table 4 shows a market survey of mechanical vibration based energy
 632 harvesting products [150].

633

634 **Table 4.** Market survey of vibration based energy harvesting products [150]

Product	Max. harvested power (mW)	Frequency (Hz)
Vulture Piezo Energy Harvester-PEH20W (Mide)	20	50-150
PMG27 Microgenerator (Perpetuum)	4	17.2
VEH-APA400M-MD (Cedrat)	95	110
VEH360 (Ferro Solutions)	10.8	60
Energy Harvesting Shoe (Scientific Research Institute)	800 mW of power/shoe at a place of 2 steps/sec	

635

636 An inertial mass is responsible for initiating the movement due to the vibration of a device.
 637 This movement can be transformed into the electrical energy by applying three types of
 638 mechanisms: electromagnetic (inductive), piezoelectric and electrostatic (capacitive) [151].
 639 Electrostatic and piezoelectric energy harvesters have several shortcomings. Electrostatic
 640 generators require an additional power source to charge the capacitor with an initial voltage in
 641 order to initiate the alteration process. Another disadvantage is the difficulty of manufacturing
 642 these converters because capacitor electrodes must not come into contact with each other.
 643 Piezoelectric energy harvesters are not efficient with microelectronics and are more compatible
 644 for moderately high vibrational frequencies. Among the other vibrational energy harvesting
 645 techniques, an electromagnetic energy harvester is able to drive low impedance load with a
 646 high current level [139]. Moreover, vibration energy harvesters are usually assembled on
 647 resonant structures with rigid suspensions such as membranes [152], cantilevers [153] or
 648 springs [154]. Their construction processes are complicated because of the low resonant
 649 frequency and it is difficult to fabricate with microfabrication process. Furthermore, a rigid

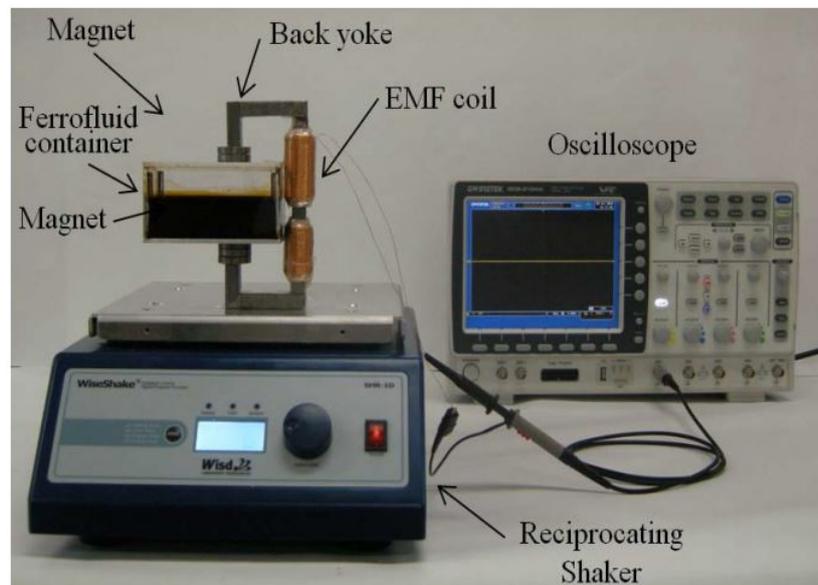
650 suspension may tend to failure or breakage in the presence of strong vibration and in the long-
651 run. Hence, an energy harvester with sturdy suspension structure and low resonant frequency
652 is highly desired.

653 The basic principles of electromagnetic energy harvesters were introduced almost two hundred
654 years ago. The Lorentz force caused by a magnetic field on moving charged carriers is initially
655 introduced by Hans Christian Ørsted in 1920 [155, 156]. Michael Faraday [155, 157] invented
656 the electric motor, and describe the transformation of mechanical to electrical energy and vice
657 versa [158]. The mechanism of the energy harvester depends on the variation of the flux linkage
658 in the coil of the energy harvester because of the natural vibration. According to the Faraday's
659 law of induction, a voltage is found and partly energy is supplied to the connected load and
660 some loss of energy due to the resistance of the coil. Using a permanent magnet is one efficient
661 way to induce an electromagnetic induction for energy harvesting [139]. Since the late 1990s,
662 many researchers [159-161] have acknowledged the different approaches used to generate
663 power from electromagnetic resources. The electromagnetic generator is an enclosed system
664 and has the advantage of being protected from the outside environment. In addition, it is reliable
665 and may reduce the mechanical damping because there is no mechanical contact between the
666 parts as well as the separate voltage source does not necessary [162].

667 Sazonov et al. [163] have described a self-powered wireless system that utilises the vibrations
668 of a bridge induced by passing traffic. The vibrations are converted into the electrical energy
669 by using the electromagnetic generator, which can produce power up to 12.5 *mW* with a
670 vibrational frequency of 3.1 *Hz*. Sterken et al. [164] studied on the electrostatic micro generator
671 and concluded that this system was compatible up to the power of 50 μw for 0.1 cm^2 surface
672 area. Dallago et al. [165] have introduced an active electronic interface for an energy harvester
673 containing an electromagnetic transducer. This transducer can deliver a voltage of 3.25 *V* at

674 resonance frequency of 10.4 Hz. Furthermore, electromagnetic materials are large in size and
675 are complex to integrate with MEMs [166]. Bayrashev et al. [167] and Staley & Flatau [168]
676 focused on energy harvesting from magnetostrictive materials because these types of materials
677 were employed to build sensors and actuators as they were capable of altering magnetic energy
678 into kinetic energy. The magnetostrictive materials are flexible, suitable for the high frequency
679 of vibration as well as overcome the drawbacks of the other vibration sources. Wang & Yuan
680 [166] described magnetostrictive materials which were used to harvest energy and supply
681 power to the wireless sensors in a health monitoring application. El-Hami et al. [143] described
682 a vibration based electromechanical energy harvester that contained a pair of magnets and
683 cantilever beam.

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Fig. 9. Experimental setup for measuring induced electromotive force of electromagnetic energy harvester [146]

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690

Nowadays, electromagnetic generator utilises the sloshing of ferrofluid columns to harvest vibration energy. A ferrofluid is strongly magnetised under the influence of a magnetic field.

691 Ferrofluid is a stable suspension with a mixture of magnetic nanoparticles in a conventional
692 carrier fluid such as water or oil. Generally, the nanoparticles are ferri- or ferromagnetic
693 particles with the diameter of <10 nm and coated with a surfactant layer of 1-2 nm. The
694 difference between magnetorheological fluid and ferrofluid is the size of the particles. Bibo et
695 al. [21] studied the use of ferrofluids in an electromagnetic micro-power generator and
696 determined the influence of the ferrofluid quantity, magnetic field strength and acceleration
697 level on the output voltage of the harvester. They concluded that the harvester output voltage
698 increased with an increase in acceleration level, but the height of the fluid column had a small
699 effect on the output voltage. Oh et al. [146] analysed the effect of ferrofluid sloshing movement
700 on energy harvester to generate an electromotive force (EMF), and the experimental set-up
701 used in this study is shown in Fig. 9. The influence of ferrofluid volume and different strength
702 of permanent magnets on the characteristics of induced electromagnetic force were also
703 examined. Results showed that, output power (μW) increased with an increase in vibration
704 speed (RPM) and a decrease in magnetic flux densities (mT). Alazmi et al. [169] investigated
705 the sloshing motion of ferrofluids, and concluded that the sloshing motion of the magnetised
706 nanofluids creates a time dependent magnetic flux, which can generate an EMF in a coil. Seol
707 et al. [170] studied the ferrofluid based triboelectric-electromagnetic hybrid generator for
708 vibration energy harvesting. The result showed that the hybrid energy harvester was
709 advantageous for harvesting subtle and irregular vibrations in the extremely low threshold
710 amplitude and a wide operating frequency range. Wang et al. [171, 172] examined ferrofluid
711 liquid springs for vibration energy harvesting. The results showed that the ferrofluid liquid
712 spring can attain relatively low resonant frequency when engaging small volume of liquid. It
713 is also compatible in the presence of high input acceleration. The modal frequencies of the fluid
714 column are determined by the following equation [21, 173]:

715
$$f_{mn} = \frac{1}{2\pi} \sqrt{\frac{g^* k_{mn}}{R} \left(1 + \frac{\sigma k_{mn}^2}{\rho g^* R^2} \right) \tanh\left(\frac{k_{mn} h}{R}\right)} \quad (5)$$

716 where, the effective acceleration, $g^* = g - \frac{\chi}{\rho\mu_0} \frac{dB(z)}{dz} B(z) \Big|_{z=h}$, and g represents the
 717 gravitational acceleration, m & n are the different oscillation nodes, h is the liquid height, R is
 718 the radius of the container, μ_0 is the magnetic permeability of vacuum. From Eq. (5), it is clear
 719 that the ferrofluids properties (like density and surface tension) have an important impact on
 720 the modal frequencies as well as the electromagnetic force, accordingly.

721 A large number of studies have been done on conventional electromagnetic energy harvester,
 722 where solid magnets were used to generate electromotive force. Dissimilar to the solid magnets,
 723 ferrofluids can easily change its shape and injected into hard to access locations due to its
 724 fluidity. Moreover, ferrofluid based energy harvester can also respond at infinite closely spaced
 725 modal frequencies which may enhance the performance of energy harvester under the influence
 726 of non-stationary and random excitations. Therefore, a broad study is needed on ferrofluid
 727 based electromagnetic energy harvester in the future to overcome the difficulties in the current
 728 energy harvesters as discussed earlier.

729

730 **8. Conclusion and recommendations for future work**

731 This paper involved the recent development on magnetic nanofluids for energy systems and
 732 energy harvesters. Recent studies have discussed the thermomagnetic convection and
 733 thermomagnetic effects. Few investigations have been done on the other features of heat
 734 transfer such as the improvement and control of thermal properties of ferrofluid under the
 735 external magnetic field. The use of magnetic nanofluids in heat transfer, as well as

736 electromagnetic energy harvester applications, appear promising. Several studies have been
737 investigated the outstanding thermal properties of nanofluids, and showed that the nanofluids
738 exhibit significantly higher thermal conductivities and convective heat transfer properties in
739 comparison to conventional heat transfer fluids (e.g. ethylene glycol, water, and mineral oils,
740 etc.), although some doubts have been raised recently about these exaggerated claims. The
741 overall effectiveness of nanofluids in heat transfer applications can be best evaluated if the
742 enhancements in both thermal conductivity and viscosity are considered at the same time. In
743 comparison, relatively little work has been done on the factors affecting nanofluid stability,
744 specifically the effect of zeta potential and particle size distribution on nanofluid viscosity and
745 thermal conductivity. Stability is an important concern in nanofluids research as attaining the
746 desired stability of nano-suspensions remains a big challenge.

747 The development of magnetic nanofluids as a heat transfer fluid is still challengeable by many
748 aspects ranging from the production and characterisation of nanofluids to the practical
749 applications through the understanding of the mechanisms responsible for the observed heat
750 transfer improvement. Future investigations should emphasise the preparation of ferrofluids
751 and influence of different parameters on the size of the final synthesised nanoparticles. Another
752 important aspect is the control of particle morphology during the preparation of the
753 nanoparticles, because the cluster formation may be affected by the particle morphology. The
754 control of the particle morphology and particle size may enable to prepare a unique ferrofluid
755 with desired thermal properties as well as long-term stability, and further on the development
756 of devices using magnetic nanofluid for practical applications.

757 The ferrofluid based electromagnetic energy harvesters implement ferrofluids, which can
758 easily change shape and respond to very small vibrations. The further research will be needed
759 to examine the science underpinning the behaviour of tunable magnetic nanofluids with a view

760 towards creating a step-change improvement in the performance of electromagnetic energy
761 harvesters. Most of the researches have been done on conventional electromagnetic energy
762 harvester using solid magnets, and very few studies have been performed on ferrofluid based
763 energy harvesters. The effect of fluid quantity, the shape, and size of the fluid container,
764 magnetic field induction, vibration speed on EMF and power output of ferrofluid based energy
765 harvester have been studied. But, the consequence of changing the physicochemical properties
766 (such as pH, the weight concentration of nanoparticles, the size of nanoparticles etc.) of
767 ferrofluids on the output power of the energy harvester will need to be examined and analysed.
768 Then, the feasibility of the proposed method will need to be determined and the characteristics
769 of electromagnetic force should be explained through the use of the experimental result.

770

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