Chapter 2
MR Fluids

2.1 Introduction

Magnetorheological fluids are a suspension of fine, non-colloidal, low-coercivity ferromagnetic particles in a carrier fluid (Carlson and Wesis 1995). They belong to the class of controllable fluids which reveal the ability to change from a liquid type behaviour to that of a semi-solid with a yield stress when exposed to external magnetic fields. The changes are reversible and fast. MR fluid fulfil the important performance criteria such as low initial viscosity, high shear upon the application of the magnetic field, low hysteresis, low power consumption, temperature stability, and fast response (Ashour et al. 1996). The unique nature of MR fluids have made them suitable for semi-active energy-dissipating applications in particular (Carlson and Chrzan 1994).

In this chapter the authors highlight the general principles of MR fluids. Their rheology, critical parameters are discussed, and the mechanisms governing the so-called MR effect are characterized. For a more in-depth review of modelling efforts, compositions and the influence of critical parameters the reader should refer to e.g. de Vincente et al. (2011) or Bossis et al. (2008).

2.2 Mechanisms of the MR Effect

By far, the most accepted model for the MR fluid magnetization is the particle magnetization model (de Vincente et al. 2011). According to the model, the MR effect occurs due to the mismatch in the permeabilities of the solid phase and the liquid phase, respectively. It is further assumed that particles act as magnetic multi-domains (Agraval et al. 2001). In general, solid particles dispersed in the fluid feature a number of sub-domains; each domain has a randomly aligned dipole moment in the

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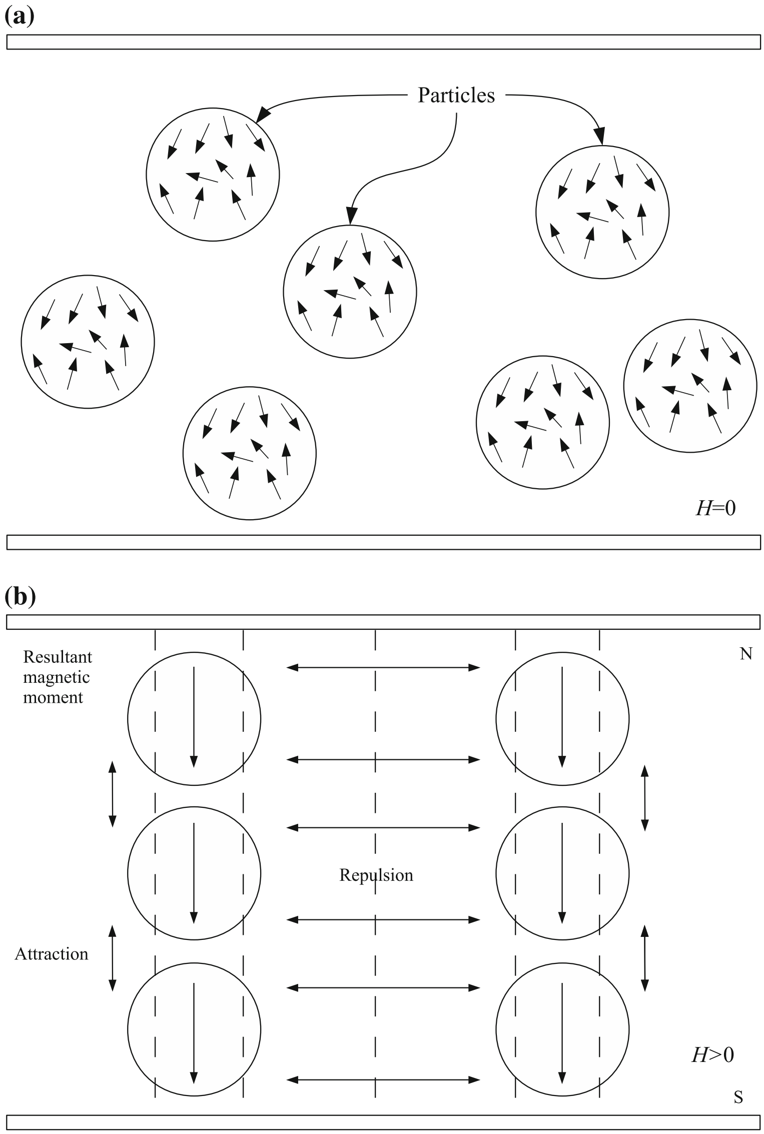

Fig. 2.1 MR fluid particles (Agraval et al. 2001). **a** Off-state, **b** On-state

absence of magnetic stimuli (see Fig. 2.1a). Once they become ordered in the presence of the magnetic field, all the sub-domains in a particle align in one direction, and the particle is subjected to magnetic forces. It can be seen that the forces between two particles interact with each other and the particles attract (or repel) each other. Attraction occurs in the direction parallel to the magnetic field lines, and repulsion in the direction perpendicular to them. That leads to the formation of chain-like structures in the direction of the magnetic field lines as illustrated in Fig. 2.1b.

The MR response results from the polarization induced in the suspension particles upon the application of the external stimuli (Jolly and Nakano 2013; Jolly et al. 1999) which induces a dipole moment in each of them. As the dipole-dipole interaction increases, the particles align to form chains along the flux lines (Felt et al. 1996). Increasing the magnetic field strength causes the aggregation of these chains into columnar structures parallel to the field lines. In this condition the fluid exhibits yield stress that is magnetic field dependent. The yield stress is the minimum stress that needs to be overcome by external forces to initiate the flow. The manner and rate at which the particles form the chain-like structures depend mainly on the rate of magnetic field strength increase and particle ordering (Mohebi et al. 1999).

Several models have been proposed so far to predict the chain formation process and to evaluate the yield stress in monodisperse as well as bidisperse MR fluids (Bossis and Lemaire 1991; Ginder et al. 1996a; Ginder 1998b; Kittipoomwong and Klingenberg 2005; Li and Peng 2012; Shulman et al. 1986; Si et al. 2008). The researchers have attempted to model magnetic dipole interactions between adjacent particles within a particle chain. One of the earliest attempts to quantify the magnetorheological effect through using mathematical models came from Shulman et al. (1986) who developed a micromechanical model of the MR suspension on the basis of the statistical theory of diluted suspensions of the following form

$$\tau = \mu \frac{du}{dz} + \mu_0 H^2 \phi_v \alpha_0^{-1} \frac{\kappa_v}{2} + \kappa_v \tag{2.1}$$

where τ—shear stress, μ—viscosity, H—magnetic field strength, ϕ_v—particle volume fraction, μ_0—vacuum permeability, κ_v—susceptibility and α_0 is a constant. Although novel and useful at that time, the model was quite limited, and other efforts followed soon. For example, Ginder et al. (1996a) and Ginder and Davis (1994) carried out a numerical and analytical analysis of the magnetic phenomena by means of the finite-element method. The researchers identified three regimes. At low applied fields, the stress would increase quadratically, i.e. proportionally to the magnetic field strength squared. In the second (intermediate region), the change rate of the stress would be reduced. That is followed by a complete saturation at high fields. Ly et al. (1999) simulated the process of chain formation using the Fast Multipole Method. The authors performed the analysis for fluids with different iron content. They concluded that the time to form the aggregates is inversely proportional to the iron volume fraction. One recent model by Si et al. (2008) is a further attempt to characterize the yield stress in terms of the magnetic field strength, particle size, iron particle volume fraction. The model is illustrated in Fig. 2.2, where h denotes the gap height, and F_a is the external force applied to the upper plate. The bottom plate is held stationary. The τ_0 is the shear yield stress per unit area. It is assumed that $\tau_0 = F_a \sin\theta_a$, where θ_a is the angle between the centerline of the chain and the magnetic field direction. The analysis showed that the yield stress of MR fluid under these conditions can be given as

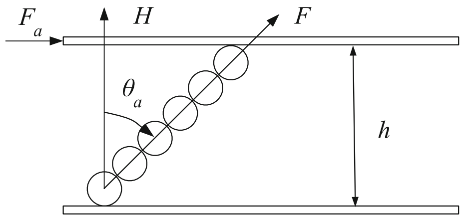

Fig. 2.2 Shear yield stress (Si et al. 2008)

$$\tau_0(H) = \sum_{n=1}^{k_p} \frac{\mu_0}{12n^2} \frac{r\phi_v(\mu_{MR} - 1)^2 H^2}{(2r + \delta_p)} \sin\theta_a \cos\theta_a \tag{2.2}$$

where μ_{MR}—relative permeability of the MR fluid, $\mu_{MR} = 1 + \kappa_v$, κ_v—susceptibility, k_p—average number of particles in each chain, and $k_p = A_f h / V_s N_s$, A_f—flat plate area, V_s—average volume of solid particles, N_s—number of chains in unit area. All models predict similar relationships as long as low magnetic field strengths as well as low solid phase volume fractions are involved. However, Ginder and Davis (1994) and (Phule and Ginder 1999) showed that as the field increased, other expressions would yield more satisfactory results. At intermediate magnetic field strengths the yield stress can be predicted according to

$$\tau_0 = \sqrt{6}\phi_v M_s^{-1/2} H^{3/2} \tag{2.3}$$

where M_s—saturation magnetization, and the saturation yield stress can be calculated as

$$\tau_0 = 0.086\phi_v \mu_0 M_s^2 \tag{2.4}$$

Finally, it should be noted that the above expressions express the yield stress as a linear function of the iron volume fraction ϕ. de Vincente et al. (2011), however, argued that the conclusion would be primarily valid for diluted MR suspensions only.

2.3 MR Materials

An MR fluid is a suspension of magnetizable particles in a carrier fluid. Various additives are present in the composition to enhance the yield stress, improve the settling rate, reduce oxidation, etc. (Phule 2001).

In the context of the automotive industry that by far has been the biggest beneficiary of the Rabinov's discovery, MR material compositions should be optimized for maximum yield stress magnitude changes, temperature operating range, durability, minimum settling, oxidation and paste formation (in-use thickening—IUT). MR fluids should have an acceptable low viscosity in the absence of magnetic forces, yet at the same time they should be capable of exhibiting large yield stresses ($\tau_0 \gg$ 20 kPa) when subjected to a magnetic stimuli of flux densities within the range from 0.6 to 1.0 T (tesla) (Carlson and Chrzan 1994)—high turn-up ratio. The temperature range that commercial MR fluids are claimed to operate within varies from −40 to 120 °C in the conditions of continuous exposure and with excursions up to 150 °C (Alexandridis 2007).

Automotive vehicle dampers impose the most demanding conditions for MR materials. The shear rate well exceeds 10^5 s^{-1} at the piston velocity of 1 m/s, and the temperature range within which the fluid should operate approaches 140 °C. Rotary MR dampers in which the fluid is generally subjected to shear rates of 10,000 s^{-1} provide a far more benign environment for exploiting the material's controllable characteristics. It is also clear that easy-to-remix MR suspensions would be preferred over thixotropic gels, for example. Arguably, the latter would be more suitable for seismic damper applications, whereas the former would perform best in the automotive environment.

2.3.1 Liquid Phase

Examples of typical carrier fluids (constituting the continuous phase of MR fluids) are silicon oils, mineral oils, paraffin oils, silicone copolymers, white oils, hydraulic oils, synthetic hydrocarbon oil, water, esterified fatty acid, ferrofluid (Carlson and Weiss 1995; Foister 1997; Iyengar and Foister 2003a; Iyengar et al. 2004b; Lopez-Lopez et al. 2006; Weiss et al. 1997a, b, 2000). One particularly preferred carrier fluid material is polyalphaolefin (PAO). The material is well known for its wide temperature operating range. Again, in the context of automotive suspension applications, the viscosity of the carrier fluid should vary between 0.001 Pa·s and 0.1 Pa·s when measured at ambient temperature. According to Weiss et al. (1997a), carrier fluids should be chemically compatible with both the material the particles are made of and device materials. Moreover, they should be capable of functioning over a broad temperature range (−40 . . . 120 °C in the case of vehicle dampers), exhibit low thermal expansion, and ensure excellent lubricity in addition to presenting no hazard to the surrounding environment.

2.3.2 Solid Phase

A magnetorheological fluid is a suspension of fine, non-colloidal, low-coercivity ferromagnetic particles in a carrier fluid that responds to the magnetic field. Based on the analysis of available patent literature it seems the range of suitable solid phase soft-magnetic, low coercivity materials includes pure iron, iron alloys (incl. cobalt, vanadium manganese, molybdenum, silicon, nickel), carbonyl iron, atomized iron, water-atomized iron, iron oxides (incl. Fe_2O_3, Fe_3O_4), low carbon steel grades, silicon steel, nickel, cobalt, ferritic stainless steel, atomized stainless steel, and the like (Bombard et al. 2011; Foister et al. 2003, 2004; Forehand and Barber 2010; Iyengar and Foister 2003a, b; Iyengar et al. 2004b; Margida et al. 1996; Munoz et al. 1998). In general, the solid phase should exhibit high saturation magnetisation (1.6 . . . 2.1 T) and low remanence (coercivity). The saturation limits the magnetic field induced yield stress variation range, and low remanence delays long-term particle agglomeration and improves redispersibility (de Vincente et al. 2011; Phule et al. 1999). Other limiting factors are cost and durability.

The solid phase material of choice seems reduced carbonyl iron powder (CIP)—the thermal decomposition product of iron pentacarbonyl. The material exhibits good magnetisation properties and low remanence. Also, polymer coated carbonyl iron powder has been known to improve dispersion stability (Choi et al. 2006). Indeed a vast majority of commercially available MR fluids are carbonyl-iron based. The material's manufacturing process, however, is relatively expensive compared to other methods of producing iron powders. On-going efforts by the industry to lower the cost of the MR fluid have resulted, e.g. in the application of water-atomized iron powder in a high-durability MR fluid (Forehand and Barber 2010).

A brief review of physical properties of available off-the-shelf commercial MR fluids (e.g. Basonetic 2040, Basonetic 4035, Basonetic 5030, MRF-122EG, MRF-132DG, MRF-140CG) indicates that the solid (Fe) phase content by volume is typically within the range from 20–22 to 40–48 %—the density of those fluids varies from appr. 2,300 to 4,120 kg/m^3. Again, the former is best used with rotary brakes and clutches, whereas the latter with linear dampers. Again, low iron content fluids are likely to suffer from sedimentation, and those with high iron content accelerate wear with devices they are used in.

The particle size is typically from 1 to 100 μ in diameter, preferably in the range from 1 to 10 μ. Particles larger than 100 μ are known to cause irreversible jamming in MR devices, increased friction and accelerated wear, and those smaller than 1 micron (as in ferrofluids) have been confirmed to generate insufficient yield stress changes that have rendered them useless for use in controlled damping applications and devices, However, Carlson et al. (2008) claims to have eliminated this inefficiency with the recent discovery of an MR valve operating in the so-called jamming mode. Apparently, small particles would not be subject to sedimentation but Brownian motion would prevent them from developing the yield stress. In fact, MR fluids using small size particles were reported to suffer from long-term particle agglomeration and separation due to surfactant breakdown. Although, decreasing the particle size reduces the sedimentation rate, the field-induced yield stress is negatively affected by the changes. Specifically, Lopez-Lopez et al. (2010) examined monodisperse spherical cobalt particles with the average diameter across the range from 60 to 800 nm and found a significant degradation of the MR yield stress when the average particle size was below 100 nm.

Unlike monodisperse formulations, bidisperse suspensions involve a mixture of particles of two significantly different sizes. For example, Rosenfeld andWereley (2002) and Wereley et al. (2006) as well as Trendler and Bose (2005) and Lopez-Lopez et al. (2013) examined formulations incorporating micron-sized particles and nanometer-sized particles. For example, the mixture examined by Wereley et al. (2006) with 20 wt% of the microparticles replaced by nanoparticles led to a substantial reduction in the settling rate (by an order of magnitude), and an increase in the dynamic yield stress of over 15 % at high magnetic field. For comparison, Foister (1997) developed a micron-size bimodal suspension involving a mixture of large-size particles ($<10\,\mu m$) and smaller particles. The large-to-small particle size ratio was 5:1. The mixture provided a significant enhancement in the yield stress with no increase in the off-state viscosity—the reported improvement was 2.7 times over the monosized suspension. Also, the study of Bombard et al. (2005) confirms that mixing two CIP materials with different particle improves the rheology of MR fluids by decreasing the off-state viscosity and increasing the yield stress.

In general, particles are spherical in shape. This geometry is preferred for low magnetic anisotropy, lubricity and durability. Recent studies have explored the possibility of using nano-wires and micro-wires (fibers) in MR fluids for improved settling and yield stress enhancement (Bell et al. 2008; de Vicente et al. 2010; Gomez-Ramirez et al. 2011; Jiang et al. 2011; Ngatu et al. 2008; Starkovich and Shtarkman 2002). It seems the fiber-based MR fluids offer a potential for a larger yield stress than a suspension of spherical particles of the same concentration.

Some interesting results were reported by Ulicny et al. (2010), who performed studies on MR fluid compositions containing a portion of non-magnetizable spheres in an attempt to increase the yield stress and decrease the fluid's weight as well as cost. Others, e.g. Ohori et al. (2013), claimed an improvement in the storage modulus in bimodal gels consisting of carbonyl iron particles and non-magnetic aluminum hydroxide particles. Finally, mixing micron-size carbonyl iron particles with nano-size particles allowed for the fourfold increase in the yield stress as well (Ginder et al. 1996b).

Polymer coating of particles improves their surface properties (reduces oxidation and abrasion) and improves dispersion stability (Choi et al. 2006; Sedlacik et al. 2011).

2.3.3 Additives

As MR fluids are suspensions of solid particles in a liquid phase, surfactants are used to delay particle settling as well as to prevent redispersion difficulties (Bombard et al. 2009; Bossis et al. 2008). The rate at which settling occurs is one of the most important design criteria when developing the fluid compositions for a specific application. Typically, thickeners or thixotropic agents are used to prevent separation of the particles and the liquid phase (Weiss et al. 1997b). Here, the range of suitable materials includes polymeric thickeners (hydrocarbons) or fumed silica (Iyengar and Foister 2002), colloidal clays (organoclays) (Foister et al. 2003; Hato et al. 2011; Munoz et al. 2001), fluorocarbon grease (Iyengar et al. 2010). For example, adding a low volume fraction (2–3 %) of silica particles produces a low off-state yield stress fluid with low settling rates. On the other hand, even such low volume fractions of fumed silica have been known to be abrasive towards damper seals and coatings (Foister et al. 2003). When used in MR fluid compositions, surface treated colloidal clays develop a low yield stress in the material that is sufficient to prevent the particles from settling. Rich et al. (2012) examined MR suspensions with the particle volume fraction up to 30 %. The suspensions composed of a synthetic clay showed a practically zero settling rate.

Although the settling rate is an important parameter, tests with MR fluids have shown that MR dampers return to nominal forces after one stroking cycle even after one year of storage (Burson 2006; Carlson 2002).

Additives are also used to cope with another in-use failure mode—in-use-thickening or paste formation. Carlson (2002, 2003) described the increase in the off-state viscosity of the fluid by a factor of 3 after 600,000 cycles when used in a damper and under the influence of magnetic field. According to Foister et al. (2003), the key factor in the process is the use of fumed silica. Surface protection agents and anti-oxidants can be added to the formulation in order to reduce or eliminate the process. Implementation of various passenger car platforms with MR fluids based shock absorbers indicates at successful solutions of the failure mode; typical life cycle requirements of automotive OEMs vary between one million and two million cycles.

Other additives are also used to reduce friction, wear, and to improve durability, as well as to ensure compatibility with device materials. For example, Foister et al. (2003) describe an exemplary durable MR fluid formulation which comprised about 50–95 % by weight magnetizable particles, about 5–50 % by weight liquid carrier, about 0.025–10 % by weight of one or more thickeners such as organoclays, fumed silicas, precipitated silicas, polyureas, alkali soaps, and an additive package. The package (organomolybdenum ditiocarbamate, an ashless ditiocarbamate and a tolutriazole compound) may further incorporate a total of at least 0.05–5 % by weight of the formulation. This formulation was optimized for use in automotive applications (controlled MR dampers) requiring fluids to pass durability (life) tests and exhibit acceptable variation of off-state forces.

In another exemplary application, Iyengar et al. (2006) describe a formulation for use with devices containing natural rubber, e.g. powertrain MR mounts. The composition incorporates a silicone fluid with a treated fumed metal oxide thickener and several hydro-bonding chemicals such as propylene glycol and a bifunctional ethoxylated amine. Other additives in the form of anti-wear agents and anti-friction agents include zinc dialkyl dithiophosphate (ZDDP), organomolybdenums (Iyengar and Foister 2003a).

2.4 Rheology of MR Fluids

Among other factors, the rheology of MR suspensions relies on particle concentration, particle shape distribution, properties of the carrier fluid, additional additives, applied magnetic field, temperature (Jolly et al. 1999). Specifically, the off-state behaviour of MR fluids depends on carrier fluid properties, additives, particle volume fraction, etc., whereas the on-state (energized) behaviour depends on the solid phase properties and the volume fraction of the solid phase.

Rheology of MR fluids is characterized in terms of pre-yield as well as post-yield conditions

$$\tau = \begin{cases} G^{*}\gamma_e & \dot{\gamma}_e = 0, \ \tau < \tau_0 \\ \mu\dot{\gamma}_e + \tau_0, \ \tau \geq \tau_0 \end{cases} \tag{2.5}$$

where γ_e denotes deformation. The post-yield behaviour of MR fluids has been observed experimentally and described mathematically on numerous occasions—see de Vincente et al. (2011). The Bingham plastic model has now become the tool of choice in characterizing the properties of MR fluids and the performance of MR fluid based devices. Phillips (1969) first used the rheological model in explaining the behaviour of variable yield stress fluids. In the pre-yield regime (below the yield stress) the behaviour of the material is viscoelastic, and the complex modulus G^{*} was found to be magnetic field dependent, too (Weiss et al. 1994). However, the Bingham model is insufficient in characterizing the behaviour of MR fluids at high shear rates, and cannot account for the pre-yield characteristics. Shear-thinning as well as shear-thickening effects appear to dominate at high shear rates and they were included in the visco-plastic Herschel-Bulkley model

$$\tau = \tau_0 + \mu\dot{\gamma}_e^{\frac{1}{m}} \tag{2.6}$$

Note that the above model is reduced to that of a Bingham plastic for $m = 1$. The Bingham model as well as the Herschel-Bulkley model are analysed in detail in Sect. 4.2. Most MR exhibit shear-thinning behaviour (Ginder 1998a), i.e. when apparent viscosity decreases with an increasing shear rate.

In general, the viscosity increases with the particle concentration. For example, Felt et al. (1996) investigated an aqueous suspension for a range of particle concentrations from 0.014 to 0.12. In the study the viscosity μ was found to vary in accordance with

$$\mu = \mu_b(1 + 2.5\phi_v) \tag{2.7}$$

where μ_b is the carrier oil viscosity. Also, particle size was found to have a significant effect on viscosity in fluids with high concentration of particles.

The yield stress $\tau_0(H)$ varies with the magnetic field applied. Therefore, the magnetization characteristics (B–H curves) are among the most important material properties when drawing requirements for a specific application. At the same time, most often it is the MR fluid that represents the biggest reluctance within the magnetic circuit of a device it is used in. Jolly et al. (1996) demonstrated that commercial MR fluids exhibited approximately linear magnetic properties up to a field of about $0.02/\mu$ A/m ($\mu = \mu_0\mu_r$). As such, studies up to this magnetic field strength assume the fluid's magnetic permeability (the slope of the fluid's B–H curve) to be constant within that range. Above the magnetic field strength the materials revealed a gradual saturation. The point at which the material saturation occurs, i.e. saturation flux density, is the product of the particle volume fraction, and particle saturation magnetization. Factors that influence the material's magnetic saturation are related to the solid phase properties and include the solid phase material, particle size, iron volume fraction, and certain additives (e.g. ferrofluids). In contrast to ER fluids, the material's yield stress is insensitive to operating temperature. Finally, due to the very good magnetic properties of the soft iron material used for the solid phase, MR fluids show little or no hysteresis.

2.5 Figures of Merit

Several performance measures have been defined by various authors to be able to predict the expected life of an MR fluid in a specific application, define the turn-up ratio, the required MR fluid volume and power consumption (Carlson 2003, 2002). For example, Jolly et al. (1999) defined the mechanical power density W_m^* and the electrical power density W_e^*

$$\begin{cases} W_m^* = \tau\dot{\gamma}_e \\ W_e^* = \dfrac{BH}{2t_c} \end{cases} \tag{2.8}$$

where t_c refers to the establishment of a magnetic field in the MR fluid, τ is the shear stress, whereas B and H are magnetic field density and field strength, respectively. Combining W_m^* and W_e^* yields the expression for calculating the efficiency of an MR fluid as

$$\alpha_m = 2\tau\dot{\gamma}_e\frac{t_c}{BH} \tag{2.9}$$

The figure of merit F_1 is based on the active volume of an MR fluid and is given as

$$F_1 = \frac{\tau}{\mu} \tag{2.10}$$

F_1 is inversely proportional to the minimum active MR fluid volume V_{min} and reflects the turn-up ratio as well as the required MR fluid volume and power consumption. Maximizing F_1 makes an MR device smaller and more energy-efficient.

Modifying F_1 to include the density ρ allows for incorporating weight. Thus,

$$F_2 = \frac{\tau}{\mu\rho} \tag{2.11}$$

The remaining figure of merit F_3 reflects the power efficiency of MR fluids

$$F_3 = \frac{\tau}{BH} \tag{2.12}$$

Maximizing F_3 relaxes the power consumption requirements in a given application device.

The life of a device (*LDE*) is given by

$$LDE = \frac{1}{V}\int_0^{TR} P_m \cdot dt \tag{2.13}$$

where V denotes the volume of MR fluid and P_m is the mechanical power converted into heat during the operation of an MR device. *LDE* determines the life time dissipated energy (*TR*) or the total mechanical energy converted into heat per unit volume of MR fluid. For example, F_1 usually varies from 10^{10} to 10^{11} for many commercial fluids (Agraval et al. 2001; Jolly et al. 1999), whereas *LDE* of a good MR fluid is on the order of 10^7 J/cm^3 or higher (Carlson 2002).

2.6 Summary

In general, MR fluids reveal a fascinating complex behaviour that can be tuned upon the application of an external magnetic field. The material has matured as several barriers have been overcome over the period of last twenty years. It has evolved from being a scientific curiosity to a well-established and proven technology. The fluid is a rare example of a smart material that paved its way into a relatively high-volume product in the automotive market. The automotive environment in which fluids are subjected to high shear rates, changing road conditions, temperature variation, heating, etc. imposes challenging and diverse life and performance requirements. That is a strong evidence that many numerous obstacles and challenges, e.g. improving the sedimentation stability of MR fluids, enhancement of their rheological properties and durability, operating temperature range, in-use thickening in MR fluid based systems have been overcome and resolved. Regrettably, published studies on fluid durability are rare and have received relatively little attention (Carlson 2003, 2002; Forehand and Barber 2010; Iyengar et al. 2004a). Efforts toward new formulations of MR fluids seem to be directed towards low cost and durability as well as settling.

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