Magnetorheological Systems with Optimized Performance

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Doctoral Thesis Summary

Tomas Bata University in Zlín

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Faculty of Technology

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Magnetorheological Systems with Optimized Performance

Magnetoreologické systémy s optimalizovaným výkonem

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ABSTRACT

The field-responsive materials belong among necessary attributes of today´s modern society as they offer a sophisticated solution for many technical needs. From this perspective, the immense potential is assigned to the magnetorheological (MR) systems, which are composed of micron-sized soft ferromagnetic particles dispersed either in non-magnetic dispersing medium or an elastomeric matrix. The feature of these systems known as the MR effect is the ability to rapidly, reversibly and in a controlled manner change their rheological/viscoelastic properties upon the exposure of an external magnetic field.

The presented doctoral thesis is devoted to the development of novel MR systems with controlled performance and enhanced stability properties through the advanced particle-grafting technology. The emphasis is given to the design and synthesis of ferromagnetic core-shell structured particles via atom transfer radical polymerization (ATRP). Using different reaction conditions, this technique allows achieving desired polymer shells with defined structure, molecular weight and thickness. As known, the quality of polymer shell plays a major role in particle stability and also significantly influences the performance of both, the MR suspensions and the MR elastomers. Herein, synthesized particles exhibit remarkably enhanced thermo-oxidation and chemical stability without unsuitably affected magnetization. To this date, this combination of characteristics was challenging to achieve via conventional modification techniques and majority of attempts was not successful. The inventions in Thesis provide significantly enhanced sedimentation stability with negligibly lower MR effect of the MR suspensions. Further, the embedding the ATRP polymer–grafted particles into suitable polymer matrix improves interfacial compatibility and even intensifies the relative MR effect when used in the MR elastomer systems. Moreover, the obtained MR elastomer is characterized by improved magnetostriction and damping capabilities.

As presented in this doctoral thesis, the specially-designed core-shell structures prepared via surface-initiated ATRP may contribute to the development of the next-generation of the MR systems with well-balanced properties tailored towards a specific application, which was not possible to achieve by foregoing conventional methods.

In a view of potential applications, the prospects are expected in the areas ranging from automotive to civil engineering, especially in the development of emission-free MR brakes or semi-active MR bridge bearings preventing the bridge degradation.

ABSTRAKT

Materiály reagující na vnější fyzikální pole nepochybně patří mezi atributy moderní společnosti, jelikož nabízejí sofistikovaná řešení pro mnoho technických potřeb. V tomto ohledu je velký potenciál přisuzován magnetoreologickým (MR) systémům, které jsou složeny z feromagnetických mikročástic dispergovaných v nemagnetické nosné kapalině nebo elastomerní matrici. Hlavním charakteristickým rysem těchto systémů je tzv. MR efekt, což je jejich schopnost rychle, vratně a řízeným způsobem měnit své reologické/viskoelastické vlastnosti v přítomnosti vnějšího magnetického pole.

Předkládaná doktorská práce je věnována vývoji nových MR systémů s regulovatelným výkonem a zvýšenou stabilitou, čehož je dosaženo pomocí pokročilé technologie využívající roubování polymeru na povrch částic. Důraz je kladen na návrh a syntézu feromagnetických částic typu jádro-obal pomocí kontrolované radikálové polymerace s přenosem atomu (ATRP). Použitím různých reakčních podmínek tato technika umožňuje připravit žádaný polymerní obal s definovanou strukturou, molekulovou hmotností a tloušťkou. Jak známo, kvalita polymerního obalu hraje hlavní roli zajišťující stabilitu částic, ale také významně ovlivňuje výkon jak MR suspenzí, tak i MR elastomerů. Částice syntetizované v rámci této doktorské práce vykazují značně zvýšenou termo-oxidační a chemickou stabilitu bez nežádoucího ovlivnění jejich magnetizace. Tuto kombinaci vlastností je v současné době velmi obtížné dosáhnout pomocí konvenčních technik modifikace. Vylepšení prezentovaná v této tezi umožňují významně zvýšit sedimentační stabilitu MR suspenzí s nepatrným snížením MR efektu MR suspenzí. Inkorporace polymerem roubovaných částic připravených pomocí ATRP do vhodně zvolené elastomerní matrice může zlepšit mezifázovou kompatibilitu a dokonce zintenzivnit relativní MR efekt u výsledných MR elastomerních systémů. Získané MR elastomery jsou navíc charakteristické zlepšenou magnetostrikcí a tlumícími schopnostmi.

Jak je prezentováno v této doktorské práci, speciálně navržené struktury typu jádro-obal připravené z povrchu iniciovanou ATRP se mohou uplatnit ve vývoji následující generace MR systémů s vhodně uzpůsobenými vlastnostmi přímo pro konkrétní aplikaci, čehož nebylo možné dosáhnout pomocí konvenčních metod.

S ohledem na potenciální aplikace je očekáváno uplatnění výsledků v řadě odvětví včetně automobilového a stavebního průmyslu, obzvláště při vývoji bezemisních MR brzd nebo semiaktivních MR uložení mostů potlačujících jejich degradaci.

CONTENTS

ACKNOWLEDGEMENTS

LIST OF SYMBOLS AND ABBREVIATIONS

Latin Abbreviations and Acronyms

2-B*i*BB	2-bromoisobutyryl bromide
3-APTES	(3-aminopropyl)triethoxysilane
ATRP	atom transfer radical polymerization
CI	carbonyl iron
CI-*g*-PGMA	CI particles grafted with PGMA
CI-*g*-PHEMATMS	CI particles grafted with PHEMATMS
CSR	controlled shear rate
CSS	controlled shear stress
EB*i*B	ethyl 2-bromoisobutyrate
EDS	energy-dispersive spectroscopy
EMF	external magnetic field
ER	electrorheological
ERS	ER suspension
FTIR	Fourier-transform infrared spectroscopy
GPC	gel permeation chromatography
H–B	Herschel–Bulkley
J–A	Jiles–Atherton
LVR	linear viscoelasticity region
M–B	Mizrahi–Berk
MR	magnetorheological
MRE	MR elastomer
MRS	MR suspension

NLVR		non-linear viscoelasticity region
NMR		nuclear magnetic resonance
PCL		polycaprolactone
PDMS		poly(dimethylsiloxane)
PGMA		poly(glycidyl methacrylate)
PHEMATMS		poly(trimethylsilyloxyethyl methacrylate)
PMDETA		*N,N,N',N'',N''-*pentamethyldiethylenetriamine
RP		radical polymerization
R–S		Robertson–Stiff
TEA		triethyleneamine
TEM		transmission electron microscopy
TGA		thermogravimetric analysis
VSM		vibrating-sample magnetometry

Latin Symbols

A	$(-)$	parameter describing hysteresis-free VSM curve
G^*	(Pa)	complex shear modulus
G'	(Pa)	real part of G^*
G''	(Pa)	imaginary part of G^*
G'_0	(Pa)	off–state storage modulus
G'_H	(Pa)	on–state storage modulus
H	$(A \cdot m^{-1})$	magnetic field strength
H_C	$(A \cdot m^{-1})$	critical H

H_e	$(\text{A}\cdot\text{m}^{-1})$	effective H
K	$(\text{Pa}\cdot\text{s}^n)$	consistency index
M	$(\text{emu}\cdot\text{g}^{-1})$	magnetization (mass)
M_S	$(\text{emu}\cdot\text{g}^{-1})$	saturation M
$\bar{M}_\text{n}$	$(\text{g}\cdot\text{mol}^{-1})$	number–average molecular weight
$\bar{M}_\text{w}$	$(\text{g}\cdot\text{mol}^{-1})$	weight–average molecular weight
n	$(\text{Pa}\cdot\text{s}^n)$	power-law exponent
T_C	$(°\text{C})$	Curie temperature

Greek Symbols

α	$(–)$	coefficient describing domain coupling
γ	$(–)$	strain
$\dot{\gamma}$	(s^{-1})	shear rate
η	$(\text{Pa}\cdot\text{s})$	shear viscosity
η_0	$(\text{Pa}\cdot\text{s})$	off–state η
η_H	$(\text{Pa}\cdot\text{s})$	on–state η
η_pl	$(\text{Pa}\cdot\text{s})$	plastic viscosity
τ_0	(Pa)	yield stress

Special Symbols

$Đ$	$(–)$	dispersity index

LIST OF FRAMING PAPERS

PAPER I

CVEK, M.; MRLIK, M.; ILCIKOVA, M.; PLACHY, T.; SEDLACIK, M.; MOSNACEK, J.; PAVLINEK, V. A facile controllable coating of carbonyl iron particles with poly(glycidyl methacrylate): A tool for adjusting MR response and stability properties. *J. Mater. Chem. C.* **2015**; vol. 3(18), pp. 4646–56, IF 2015 = 5.066.

PAPER II

CVEK, M.; MRLIK, M.; ILCIKOVA, M.; MOSNACEK, J.; BABAYAN, V.; KUCEKOVA, Z.; HUMPOLICEK, P.; PAVLINEK, V. The chemical stability and cytotoxicity of carbonyl iron particles grafted with poly(glycidyl methacrylate) and magnetorheological activity of their suspensions. *RSC Adv.* **2015**; vol. 5(89), pp. 72816–24, IF 2015 = 3.289.

PAPER III

CVEK, M.; MRLIK, M.; ILCIKOVA, M.; MOSNACEK, M.; MUSNTER, L.; PAVLINEK, V. The synthesis of silicone elastomers containing silyl-based polymer-grafted carbonyl iron particles: An efficient way to improve magnetorheological, damping and sensing performances. *Macromolecules*. **2017**; vol. 50(5), pp. 2189–200, IF 2017 = 5.914.

PAPER IV

CVEK, M.; MRLIK, M.; ILCIKOVA, M.; SEDLACIK, M.; MOSNACEK, J. Tailoring performance, damping and stability properties of magnetorheological elastomers via particle-grafting technology. *Submitted manuscript*.

PAPER V

CVEK, M.; MRLIK, M.; MOUCKA, R.; SEDLACIK, M. A systematical study of the overall influence of carbon allotrope additives on performance, stability and redispersibility of magnetorheological fluids. *Colloids Surf., A,* **2018**; vol. 543, pp. 83–92, IF 2017 = 2.829.

1. THEORETICAL BACKGROUND

1.1 Introduction to Magnetorheology

The special-type materials belong among progressively developing areas of materials science and engineering. Particularly, the external stimulus-responsive materials are of interest for many scientists and researchers, due to their ability to rapidly and reversibly change their physical properties upon the application of an external stimulus, which can involve magnetic or electric fields, mechanical stress, UV light, pH change etc.

A concept of the field-responsive materials was introduced in 1947 when Winslow patented a method for translating electrical impulses into mechanical force [1]. He found that certain substances added into non-conducting liquid are able to develop highly-organized internal structures upon the exposure of an external electric field. As the system reacted on the electrical stimulus by changing its rheological behavior, the system was named as the electrorheological suspension (ERS). This finding inspired Rabinow, who experimented with analogous systems consisting of fine magnetic particles dispersed in a liquid and exposing them to magnetic fields [2]. The electromagnetically-controlled behavior – today termed as magnetorheological (MR) effect – was further used in a device that could serve either as a clutch or as a brake.

In the following years, most research activities were devoted to ERSs rather than to MR suspensions (MRSs) probably due to easier technical aspects of creating homogeneous electrical fields. Nevertheless, the ERSs have several drawbacks including relatively low yield stress, high sensitivity to contaminants (e.g. water), demands on high voltage or relatively narrow operating temperature range [3]. On the contrary, the MRSs are capable to develop superior yield stress, they are insensitive to contaminants and have relatively broad operating temperature range. Since the early 1990s the MRSs started commercially dominate over the ERSs and were introduced to many engineering fields including automotive [4-8], civil engineering [5, 8-10], robotics and haptic devices [11-14], exercise equipment [3], or polishing technology [3, 5, 15, 16].

As will be discussed further in text (Chapter 1.5), the sedimentation stability of the MRSs is a limiting factor due to high density of magnetic particles. To restrict the sedimentation problem, a subclass of the MR materials known as MR elastomers (MREs) has been under development since 1983. First report dealing with the MREs is attributed to Rigbi et al. who embedded magnetic particles into an elastomeric matrix [17]. The MREs are stable against sedimentation and they eliminate sealing issues, possible leakage or environmental contamination which may accompany the application of the MRSs [18]. Being the stable solid analogues to MRSs, the particle displacements are controlled by matrix elasticity and the MR effects are generally less pronounced [19]. Despite that, their

properties are suitable for various applications such as fast-response dampers, seismic protection of buildings [9, 20, 21], artificial muscles [22], piezoresistive sensors [23] or even electromagnetic shielding materials [24, 25] and flexible micro-channels for biological fluid transport [26] etc.

1.2 Physical Mechanisms Behind the MR Effect

Before describing physical phenomena occurring in the MR systems after the application of an external magnetic field (EMF), the basic mathematical definition of the MR effect as an important evaluation tool is presented (Eq. 1):

$$MR\ effect = \frac{\eta_H - \eta_0}{\eta_0} \equiv \frac{G_H - G_0}{G_0} \tag{1}$$

Here, the symbols η_H and η_0 denote the viscosity of the MRS in the presence (on–state) and in the absence (off–state) of the EMF, respectively. The relation is analogous for the MRE for which the on–state and off–state quantities are expressed using storage moduli, G_H', G_0', formalism. The MR effect is usually presented as a relative quantity and its typical values for the MRSs are laying in the range from hundreds to thousands of percent [27]. For the MREs, the relative MR effects are much lower most commonly in tens of percent [28, 29], which is caused mainly due to high initial stiffness of the matrix. Nevertheless, the existence of the MREs with a giant response to the EMF was also reported [30], but the G_0 of such MRE was notably diminished due to high plasticizer content.

The fundamental principle behind the MR effect is based on the microstructure change as a reaction on the EMF. The EMF however affects the microstructure of the MRSs and MREs differently due to their different compositions. While the MRSs contain freely-movable particles dispersed in a Newtonian liquid, in the MREs they are locked in their positions upon the completion of the matrix polymerization process [31]. Due to this difference, the MR effect is driven by various mechanisms in both systems.

The conventional MRSs are consisted of micron-sized, soft, ferromagnetic particles dispersed in a Newtonian liquid, which is the most-commonly silicone or mineral oils [5, 27, 32]. In the absence of the EMF, the particles are randomly dispersed and the MRS behaves almost according to the Newton's law or slightly in pseudoplastic manner [33]. When a certain EMF is imposed, the particles become magnetized and build-up the internal chain-like structures aligned in the field direction (Figure 1a) due to mutual field-induced dipolar magnetic interactions [5]. On the micro-level, the structure development process is a complex phenomenon involving the initial aggregation of the particles into single-width chain-like structures that later laterally aggregate to form column-like structures. Besides the inherent properties of the system, the structure formation

is also affected by the rate of field increase [27]. The MRS transition from liquid-like to solid-like state is accompanied by significant (several orders of magnitude) increase of the rheological parameters (viscosity, viscoelastic moduli) [34].

As indicated above, the MREs are fabricated by incorporating the magnetic particles into an elastomeric matrix and locking their position upon final curing. However, depending on the matrix elasticity the applied EMF can shift the magnetic particles from their original positions resulting in highly-anisotropic composite properties including the modulus increase. The MR effect of the MREs is generally characterized by Jolly's dipole-interaction model [34] similarly as for MRSs, however, this model assumes straight particle chains, which have in reality rather wavy character in the MREs and the original model was therefore modified. Moreover, the additional mechanism related to the matrix elasticity is involved as the non-affine matrix deformation and also plays a certain role in the MREs stiffening (Figure 1b) [35].
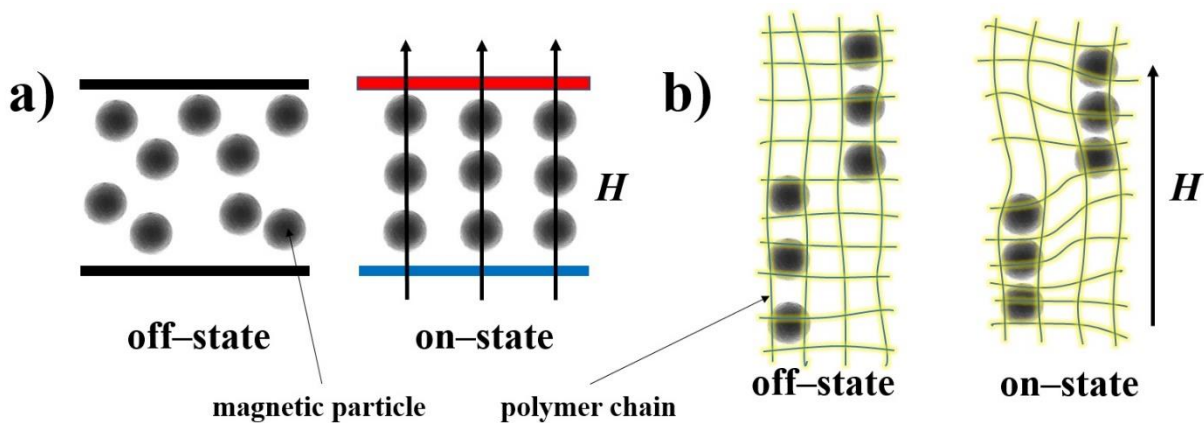
Figure 1. Schematics of the MR effect mechanism in the MRS (a), and non-affine deformation of the polymer matrix as one of the possible mechanisms responsible for the field-stiffening of the MREs (b). Redrawn from [35].

1.3 Rheological Aspects

Due to their unusual composition the MR systems exhibit complicated flow characteristics, which are necessary to be determined in order to assess their suitability in practical applications. Their rheological properties are typically investigated under both situations, the off–state as well as the on–state, while the experimental conditions are preferably chosen with a connection to the potential application. While the rheological behavior of the MRSs can be studied either in (magneto-)shear mode or in (magneto-)oscillatory regime, the MREs can be subjected only to the latter conditions.

1.3.1 Steady-Shear Behavior

In the most devices incorporating the MRSs the operating fluid is subjected to a shear flow (e.g. clutch, brake). In the off–state, the shear stress of the MRSs is almost proportional to the shear rate, which corresponds to nearly Newtonian-like (or slightly pseudoplastic) behavior, which can be approximated by the Newton's model (Eq. 2):

$$\tau = \eta\dot{\gamma} \tag{2}$$

where τ denotes the shear stress, η represents the shear viscosity, and $\dot{\gamma}$ is the shear rate. In the on–state, the field-induced structures represent a resistance against shearing resulting in the τ increase by several orders of magnitude (Figure 2). The on–state τ values are strongly dependent on the applied magnetic field strength, H [5, 27, 32].
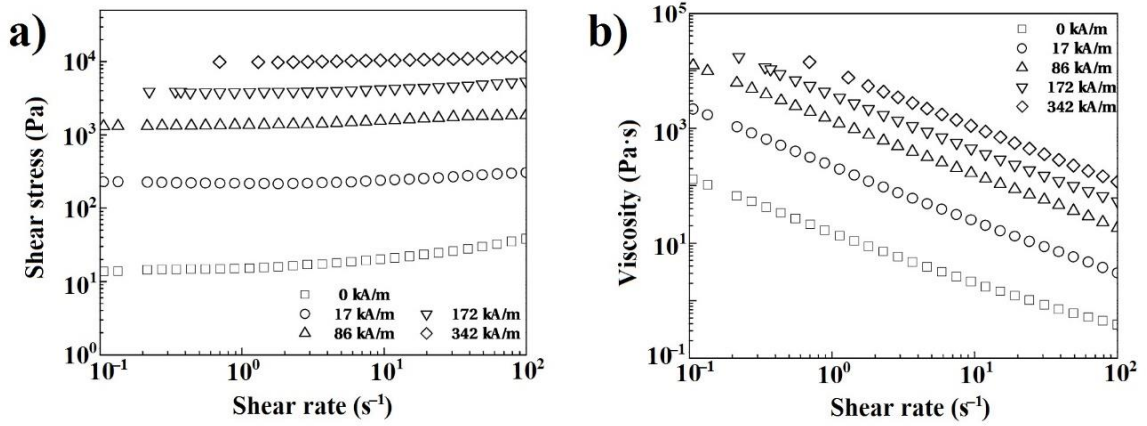

Figure 2. Typical rheological behavior of the MRSs. Shear stress (a) and shear viscosity (b) as a function of shear rate under different magnetic fields strengths. The MRS contained 30 vol.% of the carbonyl iron (CI) particles dispersed in mineral oil at 20 °C. Adopted from Cho et al. [36].

The studies dealing with steady shear magnetorheology tend to classify the MRSs (on–state) as non-Newtonian that behave according to the Bingham plastic (Eq. 3) or the Herschel–Bulkley (H–B) models (Eq. 4) [32, 37-40]. The former is represented by the original viscoplastic equation as follows:

$$\tau = \tau_0 + \eta_{\mathrm{pl}}\dot{\gamma} \tag{3}$$

where τ_0 is the yield stress controlled by H, and the constant of η_{pl} denotes the plastic viscosity of the system. The parameters τ_0 and η_{pl} can be obtained applying the Bingham plastic model to macroscale experimental measurements [41]. The Bingham plastic model has gained popularity mainly because of its simplicity [42]. However, its accuracy is questionable due to its

linear character once τ_0 is exceeded. Recently, it was concluded [33] that the H–B model is more appropriate for the MRSs especially in high $\dot{\gamma}$ region. Replacing constant η_{pl} with the $\dot{\gamma}$-dependent power-law relation the H–B model can be expressed as:

$$\tau = \tau_0 + K\dot{\gamma}^n \tag{4}$$

where K and n are the consistency index and power-law exponent, respectively. The K and n are material parameters related to materials' flow behaviors. Other viscoplastic equation was originally proposed by Casson to describe rheology of printing inks. Later, this empirical model was shown to have the applicability in hemorheology and food technology [42]. Due to its ability to fit behavior of wide variety of viscoplastic materials the Casson model was later used in magnetorheology [16, 27] showing high accuracy for the MR fluids containing the nano-sized iron particles [43].

The Robertson–Stiff (R–S) model (also known as the Vocadlo model) was proposed to describe the rheological behavior with non-linear characteristics of bentonite suspensions, cement slurries, or polymer solutions and gels [42, 44]. Due to the similar flow behavior of these materials with the MRSs, Cvek et al. [33] have employed the R–S model in magnetorheology for the first time. The R–S model was applied in the form [45] (Eq. 5) in order to obtain parameters with a physical meaning applicable for the MRSs:

$$\tau = \left[K^{\frac{1}{n}} |\dot{\gamma}|^{\frac{n-1}{n}} + \left(\frac{\tau_0}{|\dot{\gamma}|} \right)^{\frac{1}{n}} \right]^n \dot{\gamma} \tag{5}$$

with all variables defined similarly as in models above. The application of the R–S model resulted in even better agreement with the experimental data than in the case of the H–B model. Moreover, the Mizrahi–Berk (M–B) model (Eq. 6), which is commonly used in food engineering [46] was recently used in magnetorheology, however, its fitting capability to predict behavior of the MRSs in low $\dot{\gamma}$ steady shear regime was found to be insufficient [33].

$$\tau^{\frac{1}{2}} = \tau_0^{\frac{1}{2}} + K\dot{\gamma}^n \tag{6}$$

Figure 3 compares the fitting/predictive capabilities of 3-parameter viscoplastic models used in magnetorheology. As seen, the H–B and the M–B models tend to under-/overestimate τ values at a lower/higher $\dot{\gamma}$ range, while the best fit was provided with the R–S model.
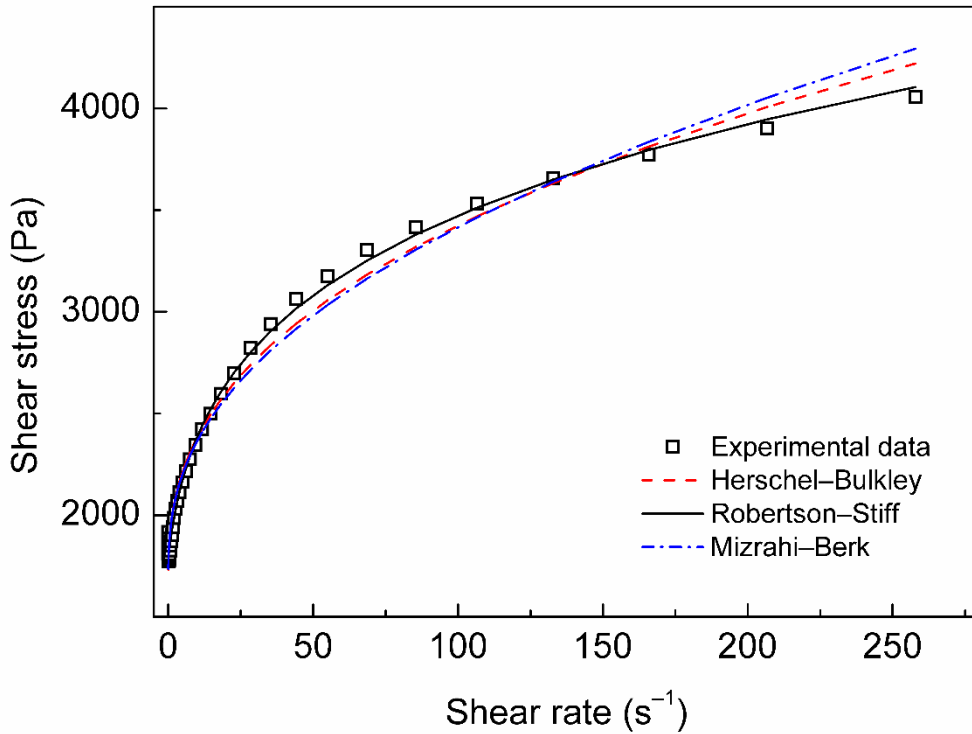
Figure 3. Shear stress vs. shear rate experimental data for the MRS containing 15 vol.% of the CI particles at 216 kA·m^{-1} with the H–B (dashed line), the R–S (solid line), and the M–B (dash/dot line) models applied. Reprinted from Cvek et al. [33].

For the equations above (Eqs. 3–6), the following condition (Eq. 7) can be applied:

$$\dot{\gamma} = 0 \,, |\tau| \, < \tau_0 \tag{7}$$

The expression shows that τ_0 must be overcome to initiate deformation or flow of the material [27]. However, there is still some debate [47] whether a true τ_0 exists or not. Despite the controversy, the engineering reality of τ_0 is a desirable and useful concept in a whole range of applications, once τ_0 is properly defined. There is no standard procedure to measure τ_0 value therefore the common technique is an indirect determination involving appropriate rheological models [48, 49].

1.3.2 Yield Stress

Yield stress is one of the most relevant rheological properties of the MRSs [27]. Basically, there are two approaches that can be utilized to determine τ_0 value. The first approach is based on the measurements in the controlled shear stress (CSS) mode, which can provide information about the static (or frictional) τ_0 (Figure 4a). However, this type of τ_0 is frequently associated with the slipping of

the aggregates on the wall of the geometry used rather than with the structure collapse under an applied shear. Another procedure involves the indirect determination based on the applying appropriate viscoplastic constitutive models (Eqs. 3–6) for the data obtained in the controlled shear rate (CSR) mode. The τ_0 obtained from the CSR mode is referred as dynamic and it is associated with the continuous breaking the aggregates during the magneto-shear. According to the literature [27, 50], the CSR mode is undoubtedly the most widely used τ_0 estimator. The dependence of dynamic τ_0 developed in the MRSs on applied H is basically underlying three regimes (Figure 4b) [51]. At low magnetic fields, τ_0 is quadratically proportional to H owing to the magnetic polarization mechanism. When H overcomes a certain critical value, H_C, the local saturation of the particles becomes more prevailing and the τ_0 further increases sub-quadratically with H. At high magnetic fields, the particles saturate and τ_0 eventually becomes field-independent [27, 33, 51, 52]. From the engineering perspective, it is important to mention that dynamic τ_0 can be also roughly estimated based on the elemental mathematical expressions as reported elsewhere [53]. As it relies from the paragraph above, τ_0 is quantity relevant mainly for MRSs, thus it is not mentioned in connection to MREs.
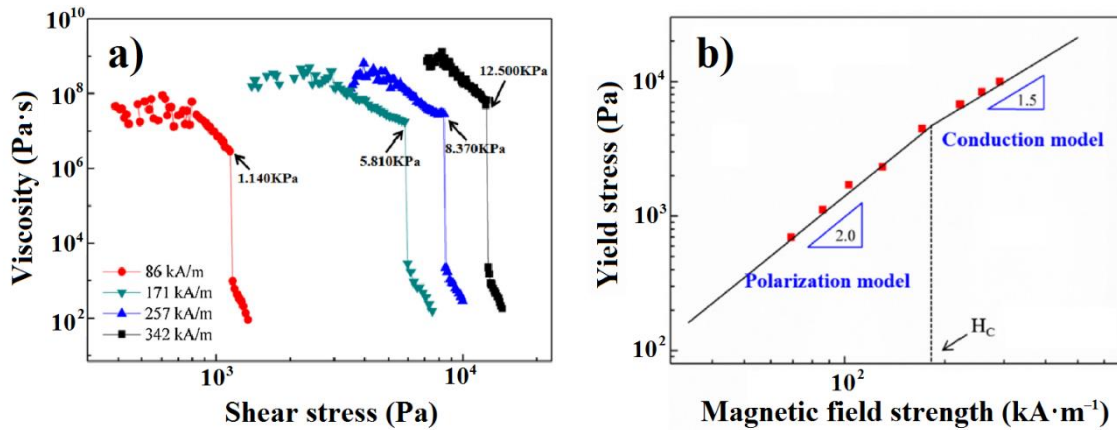

Figure 4. Shear viscosity as a function of applied shear stress obtained from the CSS mode (a) and dynamic yield stress as a function of magnetic field strength obtained from the CSR mode (b) for the MRS containing 20 vol.% of the CI particles coated with polyaniline. Adopted from Moon et al. [52].

1.3.3   Dynamic Behavior

The certain devices incorporating the MR systems such as dampers of seismic vibrations or shock attenuators are designed to operate under dynamic loading conditions [3, 9, 10, 54]. As mentioned above, the steady shear mode is accompanied by breaking and reforming the induced particle structures [55], while the dynamic loading represented as oscillatory shear has rather deformation character. Oscillatory tests at sufficiently low strains, γ, do not destroy the particles internal structures [54], therefore they are established to be a suitable

tool to investigate the viscoelastic properties of the MR systems using the complex shear modulus, G^*, formalism. The both, the real part – storage modulus, G', as well as the imaginary one – loss modulus, G'', of the G^* possess a physical meaning only when the internal structures are not broken [56], thus the investigation of linear viscoelasticity region (LVR) is a basic prerequisite that should be accomplished. The LVR is obtained from γ-sweeps under various H (Figure 5a) as its position is typically shifting to lower γ with increasing H [57]. Then, the linear response of the material is evaluated using the frequency-sweeps. As seen from Figure 5b, in the off–state the G'' dominates over the G' reflecting the liquid-like behavior, however, in the on–state the transition to solid-like behavior occurs and the G' starts to prevail. This situation is typical for the MRSs as the MREs mostly demonstrate dominating G' even in the off–state due to inherent elasticity of the matrix [28].
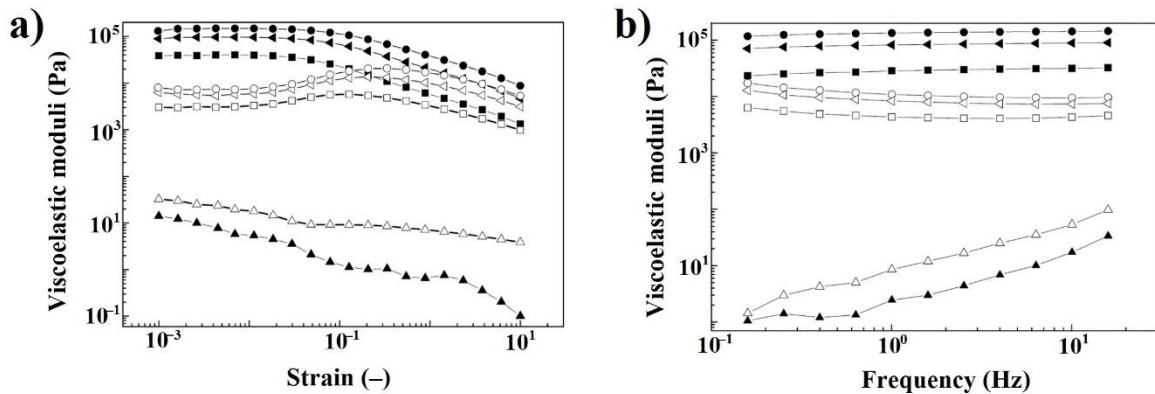

Figure 5. Dependences of the storage (solid symbols) and the loss (open symbols) moduli on applied strain (a) and on applied frequency (b) for the MRS containing 40 wt.% of cholesteryl-coated CI particles in silicone oil at temperature of 25 °C. The measurements were performed in the absence (triangles) as well as in the presence of the EMF with magnetic flux densities of 87 (squares), 178 (left-pointing triangles), and 267 mT (circles). Adopted from Mrlik et al. [58].

The linear response of the MR systems is widely-studied while their behavior in non-linear viscoelastic regime (NLVR) is often ignored although it can be a great source of information related to particles' interactions [27]. The NLVR investigations are better known from rubber industry as the filler-containing rubbers exhibit so-called Payne effect, i.e., rapid G' decrease with increasing γ amplitude due to breakdown of the filler structure [19]. In the case of the MR systems this phenomenon was recently termed as "magnetic Payne effect" by Arief et al. [59] who investigated dynamic breakdown and rearrangement of the particle network in the MRSs under the application of the EMF. Nevertheless, the concept of the Payne effect is more relevant for the MREs due to their inherent elasticity. Sorokin et al. [19] found that the Payne effect is larger in the MREs with softer matrix which is closely connected to the mobility of the particles

within a matrix and consequently to achievable MR effects. Despite the comprehensive research of his group [19, 60], some phenomena remain poorly understood and require further investigations, in particular the effect of polymer-modified particles having enhanced compatibility with the matrix on the Payne effect appearance.

1.4 Composition of MRSs and MREs

In presented MR systems, magnetic particles are dispersed either in a liquid medium or elastomeric matrix. There are several aspects, that should both types of surroundings fulfill to provide stable systems with high performance and durability. Sufficient chemical and thermo-oxidation stability was mentioned in a connection with magnetic particles but these requirements are relevant also for their surroundings. In addition, they should not interact with the particles in any negative way to achieve sufficient stability. The mostly used dispersing medium in the MRSs is silicone oil as it fulfills all the mentioned criteria. Moreover, silicone oils are manufactured with various viscosities, so one can choose the most appropriate type regarding the needs of final application. However, the experiments with mineral oil [61], paraffinic oil [62], kerosene [63], octanol [64], glycol [65] or water-based [66] MRSs have been reported. On the contrary, the majority of the MREs is based on rubber matrices, such as natural rubber [67, 68], or synthetic ones as silicone [24, 28, 69, 70], *cis*-polybutadiene [71, 72], or nitrile rubbers [73]. Also epoxidized-natural rubber [74], waste tire rubbers [75], or even thermosetting polymers e.g. polyurethanes [76, 77] have been recently investigated for this purpose. Important point regarding this topic is connected to the employment of rubber or thermoset matrices, which makes the recycling process very difficult. Due to rising number of devices based on the MREs, the recycling of these components may become a crucial aspect in the next decades. Despite that, the number of papers devoted to development of the MREs based on easily-recyclable elastomeric materials, such as thermo-plastic elastomers matrices is rather limited [25, 78-80].

The materials used as a dispersed phase in the MR systems are limited to ferro- or ferri-magnetic particles. An overwhelming majority of these systems contains the carbonyl iron (CI) particles (references e.g. [25, 81-83]) as a filler due to their suitable size (Figure 6a) and excellent magnetic properties such as high permeability, low remanent magnetization, and high saturation magnetization (Figure 6b). Also cobalt-ferrite ($CoFe_2O_4$) [57], magnetite (Fe_3O_4) [84], maghemite (γ-Fe_2O_3) [85] or neodymium-iron-boron alloy (NdFeB) [61] particles have been recently used as a magnetic filler, however, their magnetization is considerably lower in comparison with the CI particles which reduces the MR effect.
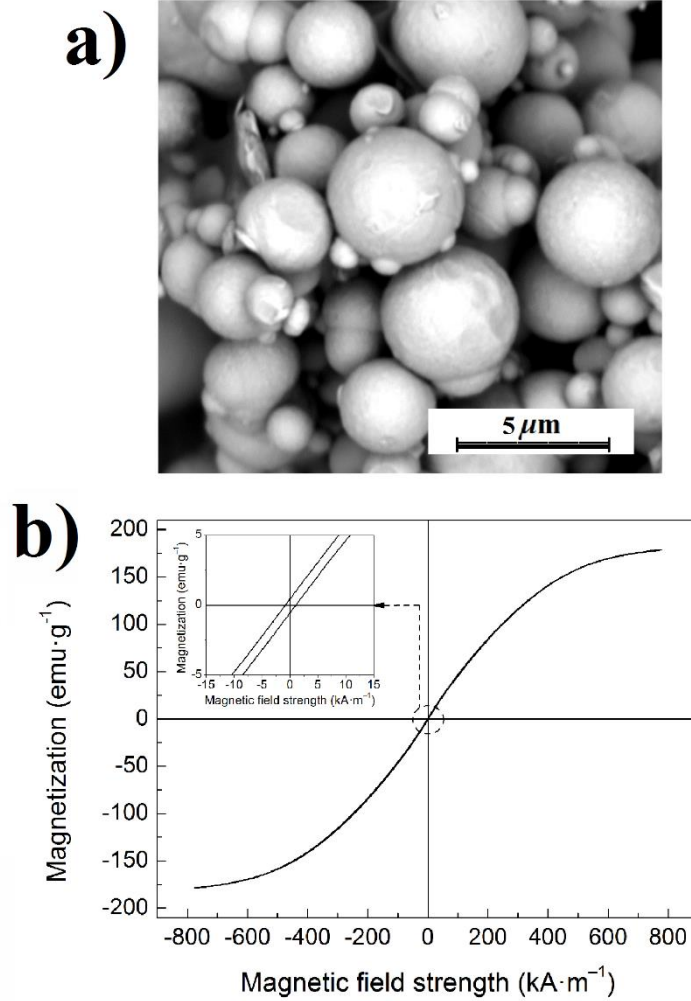
Figure 6. Scanning electron microscopy micrograph of the CI particles (a), together with their magnetization curve (b). The inset figure displays particles' magnetic hysteresis. Adopted from Cvek et al. [33].

To numerically-assess the suitability of the particles to meet the requirements of the MR systems, the Jiles–Atherton (J–A) model can be used. This mathematical formula sufficiently describes the non-linear magnetization curves typical for ferromagnetic particles. Despite that, the J–A magnetic model is still being improved to eliminate, or at least, minimize its drawbacks, which mainly include insufficient reproducibility of data fitting near the saturation state [86]. In the J–A model, the hysteresis-free curve is generally described by the modified Langevin equation (Eq. 8) having the following form:

$$M(H) = M_S \left[\coth\left(\frac{H_e}{A}\right) - \left(\frac{A}{H_e}\right) \right] \tag{8}$$

21

Here, M denotes the magnetization of material, M_S is the saturation magnetization and H is the magnetic field strength. The magnetization curve shape without hysteresis is given by the parameter A, whereas the effective magnetic field strength is denoted with H_e, which is calculated according to the expression (Eq. 9):

$$H_e = H + \alpha M \qquad (9)$$

where α represents the coefficient describing coupling between domains [87]. Recently, this model was successfully used by Cvek et al. [88] in magnetorheology to correlate the thickness of polymer coating with the reduction of magnetic properties of modified CI particles.

1.5 Common Drawbacks of the MR Systems

Despite their singularity, the MRSs suffer from several drawbacks which hinder their potential. Previously addressed issue of the MRSs is their poor sedimentation stability, which occurs due to the density mismatch between the magnetic particles and the dispersing medium. Once the particles settle down, severe redispersibility problems may arise due to their inter-particle aggregation [27]. Such situation can cause performance decrease and malfunctioning of the MR device.

The main advantage of the MREs over MRSs is that the particle sedimentation is eliminated [28]. However, the interface between the particles and the matrix plays a key role in performance and durability of the MREs. As the magnetic particles are mostly hydrophilic and the elastomeric matrix hydrophobic, the particle/matrix compatibility and matrix properties must be taken into consideration when designing the effective MREs.

The mutual issues of the MRSs and the MREs are connected to their poor thermo-oxidation and insufficient chemical stability [27, 58, 88-93]. The MR devices can be exposed to demanding operating conditions, e.g. they can operate during subterranean gas and oil exploration works, where the temperature is relatively high (~150 °C) due to geothermal gradient [94]. Moreover, they can be exposed to the presence of reactive species such as acid rains, sea humidity etc. [25]. High chemical resistance of magnetic particles is essential in final polishing of high precision optics, because this technology requires a decrease in pH to improve the MR finishing of certain polycrystalline materials [95].

1.6 State-of-the-Art

In order to reduce the above-mentioned drawbacks of the MR systems, several methods and approaches have been developed over the years and these will be further discussed in more details. As indicated in the title of the treatise, this work was devoted to MR fundamentals along with the performance enhancements of the MR systems through particle's modifications. Admittedly, a great portion of MR research has been performed via the employment of additives therefore a brief insight into the topic of additives is included in the first part of this chapter. In the second part, a special emphasis is devoted to the preparation of the core-shell structures as a mutual denominator connecting the areas of the MRSs and the MREs.

1.6.1　Additives

Facile but effective approach to enhance poor sedimentation stability and difficult redispersibility of the MRSs is the addition of stabilizing agents such as nanofillers. Using this method, no special or toxic chemicals are needed thus it is preferable way from the environmental point of view. The incorporated additives are mostly submicron-sized low-density gap fillers that occupy the interspaces between the magnetic particles (Figure 7) reducing their sedimentation rate while increasing the dispersion stability and enhancing rigidity of the internal structures in the on–state [96]. The diverse materials including fumed silica [97], organoclays [98], γ-Fe_2O_3 nanoparticles [99], graphene oxide [96] *etc*. were utilized in the MRSs for the mentioned purpose. The preparation of dimorphic [100] or bidispersed [101, 102] MRSs was also found to be beneficial. Recent study [103] compares the effect of non-magnetic rod-like ferrous oxalate dihydrate particles and their magnetic iron oxide rod-like analogues on the MR effect and stability properties. Both variants effectively enhanced magneto-induced shear stress and sedimentation stability of the MRSs; moreover the addition of magnetic rods was superior at low shear rates due to their contribution to magneto-static forces. Since the above-mentioned additives were of different sizes, shapes (spherical, rod-like/fibrous, plate-like), and were tested under various conditions (particle/additive ratio, dispersion medium, applied EMF) the overall efficiency is still unclear. Due to high interest in comparative studies and precise evaluation techniques, Cvek et al. [104] recently presented a systematical study evaluating the role of different carbon nano-additives on complex behavior of the MRSs. Based on their findings, the optimization of the MRSs for commercial applications could be based on combining the additives varying in the mechanism of their action to ensure both, rigidity of the internal structures as well as the sufficient sedimentation stability and redispersibility.

The additives are relevant due to their enhancing effects to be used mainly in the MRSs, however it should be mentioned that some work regarding this topic

was also applied on the MREs. Recently, Wang et al. [105] found that the addition of carbon black into ethylene propylene diene rubber-based MREs during the processing lead to in-situ formation of complex particle structures, which ultimately resulted in increased tensile strength, and damping of as-designed composite. Although the addition of nano-fillers is efficient regarding the sedimentation and redispersibility phenomena in the MRSs, it is ineffective for enhancements of thermo-oxidation or chemical stability of the particles. Therefore, more advanced approaches such as synthesis of the core-shell structures have been thoroughly investigated.
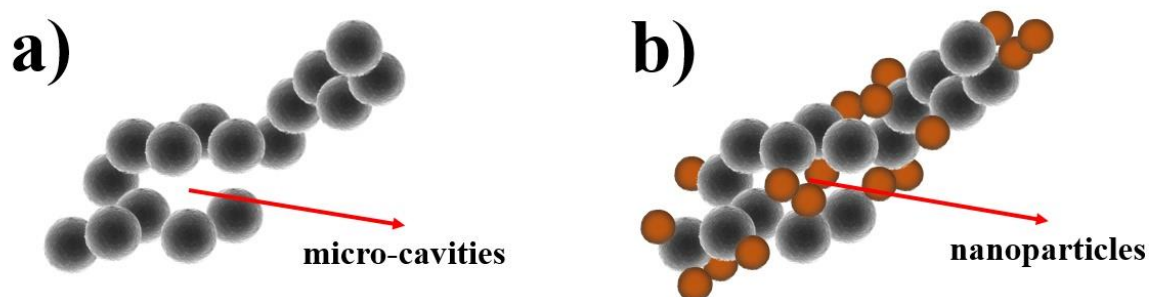

Figure 7. Magnetically-induced particle chain-like structure formation in conventional MRS with observable micro-cavities (a) and after stabilization with magnetic nanoparticles filling the micro-cavities (b). Redrawn from Ashtiani et al. [106].

1.6.2 Core-Shell Structures

The fabrication of complex particles such as core-shell-type structures has gained significant attention in the last two decades. In the early work, Shchukin et al. [107] have designed a novel photocatalytic systems based on magnetic cores coated with silica and titania shells and studied their rheological properties. Nevertheless, the first core-shell particles directly intended to enhance properties of the MRSs were prepared few years later [36]. Since that time many different types of the core-shell particles with either organic or inorganic shells have been fabricated. Prior reviewing the recent developments in the area, the basic functions of the shell material are outlined.

As concluded in numerous studies (references e.g. [5, 27, 89, 93]), the shell material generally has a lower density in comparison with metallic magnetic cores and therefore its presence contributes to lower bulk density of the core-shell structure reducing the sedimentation rate of the MRSs. High temperature or the presence of reactive species result in a degradation of magnetic particles and a formation of less magnetic products, which leads to lower response to EMFs. Therefore, the shell basically serves as a protection layer of magnetic particles prolonging the durability of the MR systems [25, 93]. In the MREs, the particles incorporated in the matrix are not directly exposed to air, but the moisture and

oxygen can diffuse through the matrix. This process is further accelerated at higher temperatures and finally results in a reduction of particle/matrix strength and lower the MR performance [81]. In the case of MRSs, the abrasiveness of the magnetic particles must be taken into consideration as well. Depending on the final application the abrasiveness can be favored or not. Jacobs [95] has shown that inorganic shell such as zirconia applied on magnetic particles enhances aqueous corrosion stability and efficiency in polishing a variety of optical glasses or crystalline ceramics due to high abrasiveness. On the other hand, organic shell generally reduces the abrasion of device surfaces extending the service life of the MR device itself [108], which is important for the majority of applications. Therefore, the attention in Thesis will be paid mainly to organics shells while omitting their inorganic analogues.

Organic shells represent the largest group of materials used in magnetorheology, which include low-molecular weight substances or different polymer coatings. Among the most investigated organic layers applied on the magnetic particles belong silane-based coupling agents [25, 78, 106, 109-112]. These substances bear hydrolysable groups (methoxy, ethoxy, acetoxy) at one end on the molecule, which enable covalent bonding to the inorganic surface, i.e., particles. The other end the molecule can contain organofunctional group (amino, vinyl, sulphide) allowing the reaction with the elastomeric matrix in the case of the MREs. Thus, silica coupling agents are used mainly to improve interfacial adhesion as they can act as a bridge between inorganic and organic materials [112]. Particle treatment with these substances is relatively straightforward, and economical, however, the modifications performed via more advanced techniques bring other advantages into the MR systems as will be explained further.

Besides low-molecular compounds such as silanes, also large spectrum of polymers has been applied as a shell material. Poly(methyl methacrylate) [36, 113, 114], poly(vinylbutyral) [108], polyaniline [115], polypyrrole [116], polydopamine [117], etc. represent only a small fraction of polymer shells examined in magnetorheology. The conventional encapsulation techniques included an in-situ dispersion polymerization [36, 115], suspension polymerization [118], or solvent evaporation [119], but these do not allow precise control of shell thickness, which results in significantly reduced particle magnetization. Moreover, such polymer shells are frequently attached via non-covalent interaction therefore the durability of prepared systems is rather questionable.

All the prepared material combinations enhanced utility properties of the MR systems to a certain degree, however, for further enhancements the researchers experimented with several other innovative methods to suitably modify the surface of magnetic particles. Fang et al. [120] employed dual-step functionality

coatings by applying the polyaniline layer and subsequent multi-walled carbon nanotube layer. This combination of materials was cautiously chosen due to their properties and intended contribution to the overall performance. For more details please see the original paper [120]. Other material combinations of sequential coatings were also investigated [120-122]. The remarkable approach introduced by Sedlacik et al. [123] involved vacuum plasma deposition of fluorinated substances onto the magnetic particle surface. With this method, it was possible to tune the particle properties to a certain degree by changing the modification times. Recently, Chuah et al. [124] showed a conceptually interesting method in which the applied polystyrene layer on the CI particles was further foamed using a supercritical carbon dioxide, which resulted in significantly enhanced sedimentation stability of the MRS.

To the author's knowledge, the literature dealing with special type coatings applied on the magnetic particles embedded in the MREs is rather limited. One example noted is the preparation of the flower-like CI particles by an in-situ reduction method [125]. Such structures were successfully used to prepare polyurethane-based MREs with significantly enhanced microwave-absorbing performance due to improved electric impedance-matching characteristics.

In general, the fabrication of core-shell particles is recognized as an effective approach to enhance performance and stability properties of the MRSs. However, there is a strong correlation among the properties which are affected by the particle modification. It is therefore a necessity to employ a strategy that allows precise tuning of modifying layers (on a molecular level) and consequently the overall behavior of the MR systems. To this date, no comprehensive research focused on this topic has been performed. Therefore, the main objective of work in Thesis will be addressed to this need. As a suitable synthesis tool the atom transfer radical polymerization (ATRP) has been chosen.

1.7 Atom Transfer Radical Polymerization

Conventional radical polymerization (RP) has a great significance in preparation of large amount of various polymers. The architectural control of resulting polymers is however limited due to very fast termination rate of the radicals. This obstacle can be avoided by applying an advanced concept known as controlled or "living" RP. A major difference between conventional RP and controlled RP is the lifetime of propagating radicals during the reaction. In the former, radical generated by decomposition of the initiator undergoes propagation and termination within a second, while in the controlled RP the lifetime of the living radical can be extended to several hours [126]. With this concept, it is possible to tailor the polymers with precisely controlled molecular weight, relatively low polydispersity, diverse composition or functionality [127, 128].

There are several types of controlled RPs among which metal-catalyzed ATRP plays the important role. This ATRP-type can be further modified to be initiated directly from the substrate which is called a surface-initiated ATRP. Assuming the suitable treatment of the substrate, this technique allows covalent grafting of the polymer chains from an inorganic surface leading to the creation of polymer brushes, which in a connection to the MR systems can positively affect the interactions with the surrounding medium or the matrix [28, 83, 89, 126]. To this date, the surface-initiated ATRP is considered as one of the most suitable fabrication techniques to gently modify the magnetic substrates and to prepare core-shell structures intended to magnetorheology.

1.8 Further Factors Influencing the MR Effect

Although the conventional MR systems are only two-phase entities, there is large variability of the individual components, what makes the understanding of these systems relatively complex. In the following part, the major factors influencing the MR effect are presented, and some of them were considered when designing novel MR systems in the experimental part of this Thesis. The next goal of this chapter is to point out some aspects which can be considered as important for further practical applications and are not covered in the current literature.

1.8.1 Particle-Related Factors

As the particles are the only magnetically-active component in the conventional MR systems, their properties such as size, shape, porosity etc. are of high importance. But the first basic assumption is related to the inherent magnetic properties of the particles as described in Chapter 1.4. Additionally, it is important to mention that the vast majority of the MR systems contains magnetically-soft materials, which allow fast demagnetization process [129] necessary for practical applications. From obvious reasons, magnetically-hard materials are not used for the preparation of the MRSs, but they were already applied in the MREs [130]. According to the authors [130], such hybrid material cannot be demagnetized by the application of reverse field, which indicates a potential to be used as active and simultaneously passive damping element.

Regarding the particle concentration; there exists a minimal threshold concentration below which no obvious field-induced response of the MR system is observed. In practice, the effective particle volume fraction for the MRSs ranges from 20 to 40 vol.%, which corresponds with a typical iron content in weight between 75 and 85 wt.% [131] as documented by the composition of commercially-available MRSs. However, the maximum yield stress (~210 kPa) can be obtained by using 50 vol.% of the particles as estimated via finite element analysis by Ginder et al. [53]. The situation is different in the MREs as in their

case the certain mechanical properties are expected even in the off–state. Thus, to ensure both, the maximal MR effect and retained mechanical properties the optimum particle fraction should not exceed 30 vol% [73].

As known, the dimensions of the particles correlate with their magnetic properties. Generally, magnetic microparticles exhibit higher M_S values when compared with nano-sized ones [132] leading to higher MR effect in such systems. Nevertheless, the sedimentation stability of the MRS should be still taken into the consideration because according to the Stokes' law, the sedimentation rate increases with the squared spherical particle radius (assuming fulfilled Stokes' law conditions). In this sense, the design of the MRS is always a compromise between high MR performance and sufficient sedimentation stability, which is usually ensured by applying the particles with the average diameter around 1–10 μm [27]. On the contrary, the MREs are stable against sedimentation which allows the incorporation of much larger particles [133], typically with the average diameter around 10–100 μm [110]. However, in some fabrication techniques of the MREs such as casting the particle sedimentation may play also a major role. To investigate the number of factors affecting the behavior of the MREs, Khimi et al. [110] designed the Taguchi method (a statistical method identifying the performance trends among multiple factors and determining their combination that yields the optimum results). This approach seems to be an effective tool in optimization of the MR systems for practical applications.

Undoubtedly, the shape of magnetic particles can severely affect the behavior of the MR systems. In principle, the particles having their major axis aligned with the direction of the EMF, i.e. rod-like particles, will have a higher induced moment and thus, the stiffer internal particles structures are formed when compared with their spherical analogues [134]. However, this aspect was found to be negligible at large particle contents and/or high magnetic field strengths [37]. The MRSs containing the rod-like particles moreover exhibited better sedimentation stability and structuration at lower magnetic fields [135], but their maximum volume fraction was less than desired 20–40 vol.% [134]. Eventually, the majority of the MRSs' research was performed on spherical particles (references e.g. [51, 53, 64, 104, 124]) probably due to their better availability from the commercial sources. In the MREs, the particles are locked in their positions in the matrix, thus their field-induced reorientation is practically impossible, although in soft matrices a certain particle rotation has been observed [136]. Therefore, the application of rod-like particles will not significantly increase magnetic permeability of the MREs when compared to their spherical analogues. To maximize magnetic permeability, the development of so called anisotropic MREs was proposed [32]. Such kind of the MREs is fabricated by particle alignment using the EMF during the curing process [69]. The anisotropic structures were found to increase the MR efficiency [137], electric conductivity [23], electromagnetic shielding capability and heat transport

properties [24] in the direction of the particle structures. These recent findings may significantly influence the development of the MREs in the near future as this approach basically allows achieving the desired utility property at lower particle volume fraction, which decreases weight of the MR devices.

Finally, it was found that also particle porosity can affect flow behavior of the MRSs. In the research performed by Vereda et al. [138] the porous iron suspensions exhibited atypical thickening behavior, which was not observed in their solid counterparts even if the particle size and magnetization were similar. The effect of particle porosity on the behavior of the MREs has not been studied yet, probably due to problematic embedding the particles into usually high-viscosity polymer matrices.

1.8.2 Dispersing Phase-Related Factors

The behavior of both MR systems is undoubtedly affected by the type of liquid medium or elastomeric matrix, respectively. Various surroundings utilized to design the MRSs or the MREs were briefly mentioned in Chapter 1.4. Here, more in-depth insights into the technology of the MR systems are given. Dealing with the MRSs, one must pay attention to the viscosity of carrier liquid with respect to the final application. Whereas the utilization of low viscosity liquid can lead to serious sedimentation instability, high viscosity inevitably increases the off–state MRS viscosity and eventually reduces the MR effect (Eq. 1) [106]. In the commercial MRSs it is also necessary to incorporate the thixotropic agents (e.g. metallic soaps as lithium and/or sodium stearate), dispersants (e.g. iron napthanate or iron oleate), anti-friction and anti-abrasion compounds. These additives are necessary to control not only the viscosity of the MRSs, but also sedimentation of the particles, the inter-particle friction, and they prevent fluid-thickening after several cycles of use [139].

As indicated further in Chapter 1.4, there is more variability in the continuous phase, when designing the MREs. Considering the wide range of elastomers and the possibility of their mutual miscibility [140] the number of possible matrices is impressively high. However, as recently published by eminent prof. Choi [141] the burdensome issue of the current MREs is their relatively high initial stiffness, thus the progress in the matrix-softening methods is expected.

Currently, there are several ways to control the matrix stiffness in the technology of MREs. The first approach is based on a reduction of the matrix cross-link density, which can be achieved by reducing the amounts of the cross-link agents [28, 29], or by tuning the ratio between the vulcanizing agent and the plasticizer such as sulfur and naphthenic oil, respectively [71]. Also, the incorporation of the particles treated with various surfactants such as fatty acids or calcium and aluminum soaps was shown to be an effective approach [70].

However, the use of low-molecular weight substances can cause durability problems as they have a tendency to migrate through the polymer matrix [142]. Other approach in controlling the MREs properties introduced by Gong et al. [72] involves the addition of polycaprolactone (PCL) as a temperature-controlled component. The PCL can transform from a semi-crystalline solid to a liquidated soft material once the temperature is increased above the PCL melting point. Thus, the MREs' stiffness properties can be controlled by varying the PCL content and temperature. Further, the matrix properties can be affected by the exposure to γ radiation [143], which is relevant mainly in aerospace and nuclear power station applications. As observed recently, the natural rubber-based MREs increased/decreased their off–state modulus depending on γ radiation dose, which was explained as a competition between cross-linking and degradation processes in the matrix. Although these approaches are effective in tuning the MREs properties, they rather omit the protection of the incorporated particles against high temperatures or acidic environment, which is essential in some practical applications [94]. Thus, there is a need to address the demands related to both, the performance and stability properties of the MREs, ideally in a single-step way. This topic was very recently solved by Cvek et al. [144], who applied the ATRP polymer-grafted particles into the MREs. Their particles were thermally and chemically resistant and at the same time, they ensured the appropriate matrix stiffness by modulating the matrix cross-link density in the vicinity of the particles.

1.8.3 Temperature

Temperature is considered as an important factor affecting the physical and mechanical properties of all the materials, which is in the case of the MR systems manifested in their MR efficiency changes. As presented above, some industrial or military applications require operations at elevated temperatures as a result of surrounding conditions or intense viscous heating [94]. This topic is unfortunately rather omitted and only sparse literature sources can be found. Aggravating fact is that the experiments in these studies were performed solely in the temperature range conditions close to ambient temperature [145]. However, these experimental studies unambiguously describe the "significant" reduction of the on-state τ in the MRSs with increasing temperature. The results of temperature sensitivity change slightly depending on the MRS used during the experiments, but generally an average normalized sensitivity of the MR dynamic yield stress is in order of $-$units $\times$ 10^{-3} °C^{-1} [145, 146]. This parameter describes the ratio between the change in the measured yield stress and the yield stress value at the reference temperature. The explanation of this phenomenon is not straightforward and one has to consider different functional relations that evolve with increasing temperature, such as decreasing particle magnetization (ferromagnetic material), intensified Brownian motion, decreased fluid viscosity, changes in expansion

coefficients of the particles and the fluid etc. These changes were mathematically described and correlated by Ocalan et al. [94]. In the practice, the reduction of the τ at high temperatures can be compensated by increasing the applied H. Interestingly, in less concentrated MRSs the opposite trend was observed, and the toughness of the induced internal structures generated in the MRSs increased with the temperature [147]. In any case, the design of the MRSs that withstand such demanding conditions has to be considered. The water-based MRSs are essentially excluded [66], while theoretically having more freedom in the choice of magnetic particles as they are generally characterized by the Curie temperature, T_C, well-above the tested temperatures (e.g. for iron, $T_C = 770$ °C) [94].

In the case of the MREs, the effect of temperature on their properties is probably even more relevant. The fact stems predominantly from their polymer basis, which is in some cases prone to thermo-oxidation. The undesirable aging and durability phenomena of the MREs were recently investigated through both theoretical and experimental approaches by Zhang et al. [140]. As known, the MR effect of the MREs tends to increase in a normal temperature range (20–90 °C) when the samples are exposed to the desired temperatures for only a limited period of time [148]. However, in the aging tests the MR effect of the *cis*-polybutadiene rubber-based MREs notably decreased after their several-hour aging at the temperature of 70 °C. The results showed that decrease of the MR effect was dependent on the type of rubber matrix, and aging conditions such as time and temperature [140].

In theory, the MR systems can be used in the middle latitudes or polar region, where the surrounding temperature can drop below water freezing-point due to seasonal changes. To the authors' knowledge, the literature dealing with the behavior of the MR systems in low temperatures is less than scarce, and the studies dealing with this topic would probably deserve more attention.

2. MOTIVATION AND AIMS OF THE DOCTORAL STUDY

2.1 Motivation

The MR systems belong among advanced materials that can change their physical properties upon the external magnetic stimulus. These materials play an important role in many disciplines ranging from automotive to civil or even biomedical engineering. In the first-mentioned field, they belong among safety systems (e.g. brakes, clutches, fast-response dampers) that can improve the comfort of passengers and contribute to lower risk of vehicle crash and consequently minimize the number of casualties and injuries. Further, the consequences of wind gusts and seismic vibrations in high-rise buildings can be efficiently reduced with the use of the vibration absorption components based on the MR systems. Their employment in medicine is in a stage of testing as drug delivery systems, or local embolization agents. The special-type particle modification may serve as a versatile tool to reduce specific as well as general drawbacks of presented MR systems across the utilization fields.

Despite the significant improvements and broad industrial applications, there are still certain physical phenomena that need to be clarified to understand and precisely predict the behavior of MR systems. Nowadays, the tailoring of the properties is of particular interest, which helps to design a material towards a specific application. The MR systems are among rapidly developing areas of materials science due to their unique properties allowing their versatile use. Figure 8 presents the number of articles published during the past 25 years devoted to the MR systems. The growth in the number of publications reflects the increasing interest in this field.
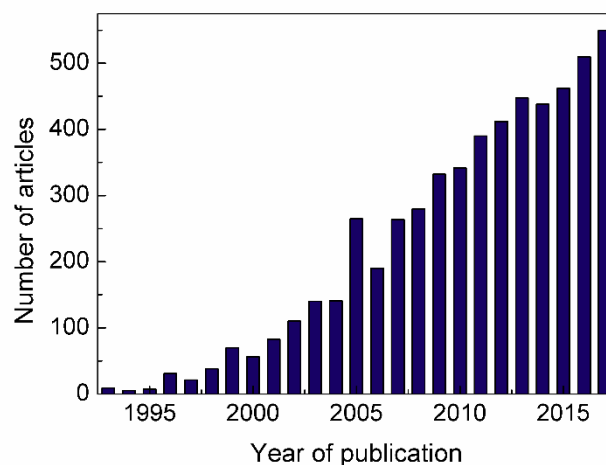

Figure 8. Number of articles published on "Web of Science" database during past 25 years (using the term magnetorheol as a topic item).*

2.2 Aims of the Doctoral Study

The main aim of the doctoral study was to eliminate common drawbacks of the conventional MR systems. The experimental work stems from the previously-published scientific papers describing the fundamental phenomena occurring in the MR systems, however the large portion of the results was not predicable and could not be obtained by any kind of simulation prior the laboratory experiments. Thus, the data was analyzed by comparing the performance and stability properties of the MR systems containing bare magnetic particles and their specifically-designed ATRP-grafted analogues. The key tasks were defined as follows:

a) Complex study of physical and chemical aspects affecting the performance of the conventional as well as recent MR systems.

b) Design and preparation of novel magnetic core-shell particles with tailored properties via surface-initiated ATRP, which could be successfully used as a dispersed phase in the MR systems in order to enhance their performance and stability properties.

c) Analysis of the shell thickness and graft molecular weight influences on magnetic properties, thermo-oxidation and chemical stability of as-designed particles.

d) Fabrication of the MR systems (suspensions or elastomers) containing the ATRP-modified particles and the study of their interactions with the dispersing medium and the elastomeric matrix, respectively.

e) The incorporation of submicron-sized additives into the MRSs and the investigation of their influence on complex behavior of such systems with the help of mathematical modeling and complementary experimental techniques.

f) Analysis of the effects of the particle modifications and additive incorporation on the MR performance, and other relevant utility properties (cytotoxicity, magnetostriction, redispersibility etc.) of as-designed MR systems. Evaluation of the systems' efficiency by comparing their properties with a reference based on bare magnetic particles.

3. OBJECTIVES OF THE WORK AND FINDINGS SYNOPSIS

The Thesis predominantly deals with the synthesis of precisely-defined core-shell particles and their possible use in the MR systems in order to reduce the outlined drawbacks of their conventional analogues. As the most of the framing papers is topically inter-connected via ATRP technique, the summary of performed syntheses and particles' general characterization are firstly given. Further, the most significant results of the individual papers are specified with respect to their primary emphasis.

3.1 Syntheses of Core-Shell Particles via ATRP

The advantages of core-shell structures in the MR systems are indisputable (Chapter 1.6.2), and shell characteristics play an important role in the overall performance and achieving high MR response. In this work, precisely-defined core-shell particles were prepared using the surface-initiated ATRP as a versatile synthesis tool. The CI particles were used as a substrate due to their suitable magnetic properties (Chapter 1.4). Under normal conditions the surface of the CI particles is covered with a thin oxide layer, therefore they were treated in acidic solution in order to increase their reactivity according to the procedure presented elsewhere [149]. Then, the activated CI particles were functionalized with (3-aminopropyl) triethoxysilane (3-APTES) and subsequently modified via amidation reaction with 2-bromoisobutyryl bromide (2-B*i*BB). The modified material was thoroughly washed and dried [89]. Finally, the surface-initiated ATRP of the desired monomer from the 2-B*i*BB-treated particles was performed and the core-shell structures grafted with either poly(glycidyl methacrylate) (PGMA) or poly(trimethylsilyloxyethyl methacrylate) (PHEMATMS) as different shells materials were synthesized (Figure 9).

The former shell variant was chosen due to suitable polymerization kinetics of its monomer during the ATRP and the presence of the oxyrane groups preserving the possibility to bond other substances [150]. The CI particles grafted with PGMA (CI-*g*-PGMA) were utilized to prepare novel MRSs (Papers I, II). The PHEMATMS was used as a shell material modifying the CI particles resulting in CI-*g*-PHEMATMS intended for use as the filler in the MREs (Papers III, IV). The PHEMATMS is hydrophobic, thus it was assumed that this silyl-based polymer will enhance the interaction of the CI particles with poly(dimethylsiloxane) (PDMS) representing the matrix material in newly-designed MREs. The shell thicknesses expressed as molar mass of PGMA and PHEMATMS were controlled by tailoring the monomer : initiator ratio, reaction time and temperature.
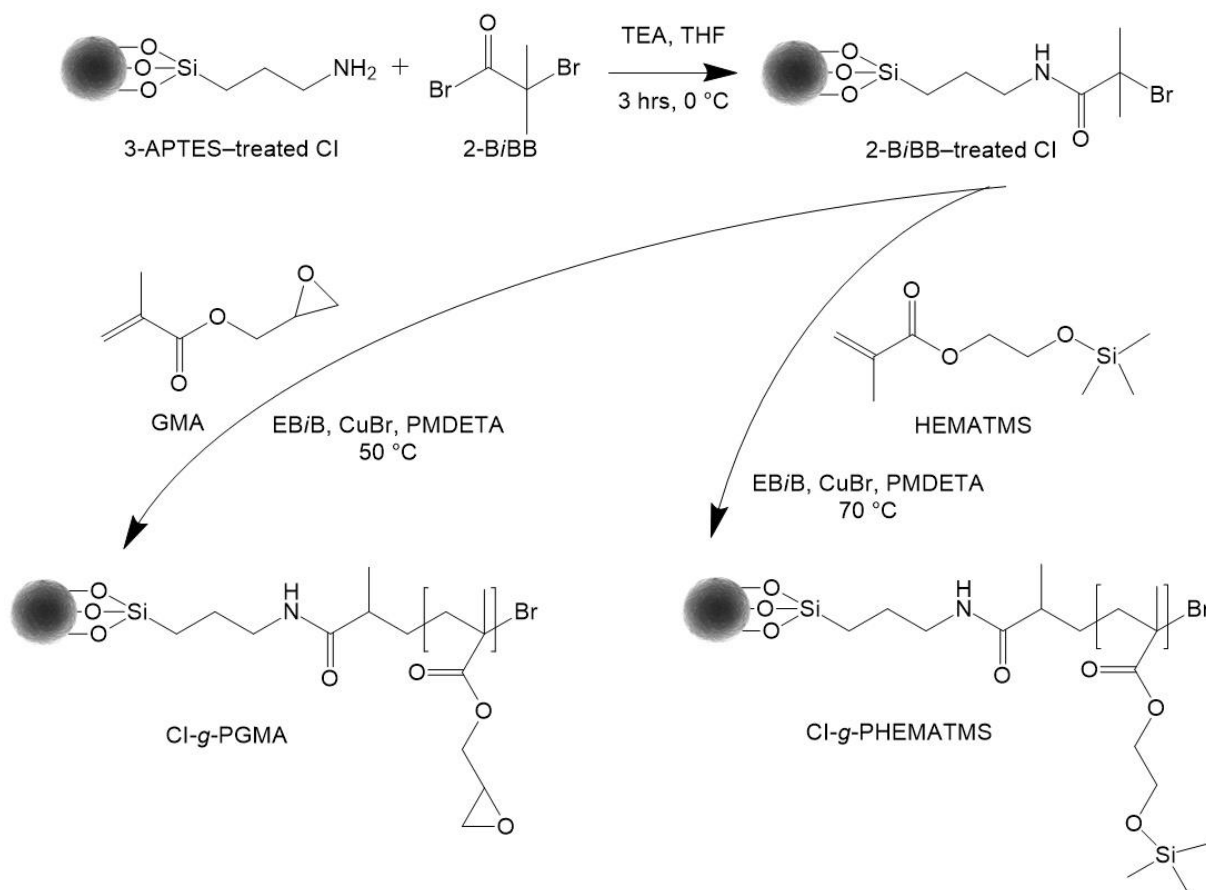
Figure 9. Immobilization of 2-BiBB initiator on functionalized CI particles with their subsequent grafting with PGMA and PHEMATMS via surface-initiated ATRP. The names of the chemicals are explained below.*

For each reaction, the feed ratio between the monomer and the initiator was tailored in order to obtain the desired shell thickness, and its maximal value was theoretically determined before the experiment. Thus, two different molecular weights of each polymer shell (sample code 1 and 2), consisting of either PGMA or PHEMATMS, respectively, were prepared and characterized. The determination of their molecular weight was possible due to the presence of a sacrificial initiator, namely ethyl 2-bromoisobutyrate (EB*i*B) in the reaction system. Molecular weight of grafted polymers and monomer conversions were obtained via gel permeation chromatography (GPC) and nuclear magnetic resonance (NMR) spectroscopy, respectively. The GPC results were evaluated considering the assumption of Goncalves et al. [151] and together with NMR ones they are summarized in Table 1. A relatively low dispersity index (*Đ*) reflects a high polymerization control via ATRP and uniformity of grafted polymer layers.

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* *TEA – Triethyleneamine, THF – Tetrahydrofuran, EBiB – Ethyl 2-bromoisobutyrate, CuBr – Copper bromide, PMDETA – N,N,N',N'',N''-Pentamethyldiethylenetriamine*

Table 1. Conversion and molecular characteristics of prepared core-shell particles.

sample code	conversion[a] (%)	$\overline{M}_W$ (g·mol^{-1})	$\overline{M}_n$ (g·mol^{-1})	$\DJ$ (–)
CI-*g*-PGMA-1	87.0	6 600	5 000	1.32
CI-*g*-PGMA-2	88.5	12 500	9 700	1.29
CI-*g*-PHEMATMS-1	90.0	11 800	9 200	1.28
CI-*g*-PHEMATMS-2	75.0	23 500	17 900	1.31

[a]Based on ^{1}H NMR spectra

3.2 Characterization of Prepared Particles

To limit the size of this Thesis summary, only some results are presented. For more data, the reader will be referred to the corresponding articles. The transmission electron microscopy (TEM) images were acquired to observe the thicknesses and uniformity of grafted layers. Figures 10b and 10c show the presence of a lower contrast polymer shell grafted onto the darker CI core. As can be seen, the grafted layers were generally uniform in both cases with the thicknesses of around ~15 nm and ~35 nm. The increasing molar mass of the grafted polymer thus led to increasing layer thickness.
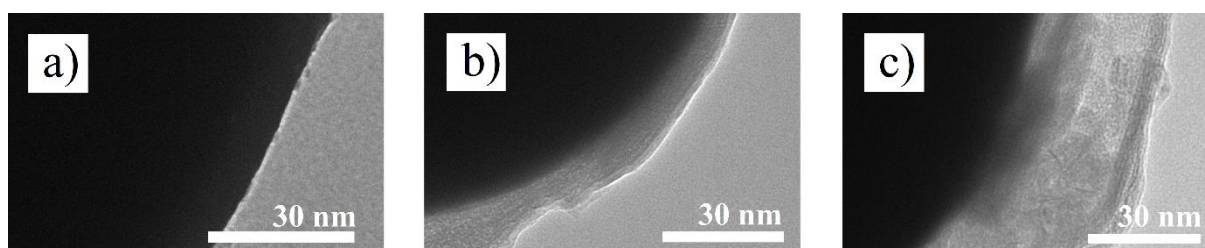
Figure 10. TEM images of bare CI (a), CI-g-PHEMATMS-1 (b), and CI-g-PHEMATMS-2 (c) showing a part of the corresponding single particle.

The energy-dispersive spectroscopy (EDS) and Fourier-transform infrared (FTIR) spectroscopy were used in order to verify the presence of polymer brushes grafted onto the CI substrate. Besides strong peaks representing iron, the EDS spectra generally confirmed the presence of expected elements coming from APTES and 2-B*i*BB residues, and the presence of elements constituting the

PGMA or PHEMATMS polymer grafts. The core-shell particles with higher molecular weight contained slightly higher percentage of carbon when compared to their thinner-shell analogues. Additionally, the peaks occurring in FTIR spectra confirmed the presence of expected functional groups in all samples, thus the ATRPs performed on the CI particles were proved to be successful. More detailed data descriptions are shown in already published papers by Cvek et al. [28, 89].

The magnetic properties of bare CI particles as well as their polymer-grafted analogues were investigated using a vibrating-sample magnetometry (VSM). The magnetization of the CI particles decreased due to the presence of both non-magnetic coatings – PGMA (please see full paper by Cvek et al. [89]) and PHEMATMS (Figure 11) – however, the decrease was negligible as the shell thicknesses were controlled via ATRP means to be in nanometer scale. Admittedly, the presence of thicker shells further reduced particle magnetization, but this difference can be interpreted as rather marginal. At the maximum employed field ($780 \, \text{kA} \cdot \text{m}^{-1}$), the PHEMATMS coatings decreased magnetization by less than ~10 % when compared to the magnetization of bare CI (Figure 11, inset). The reported magnetization decrease of core-shell particles prepared via conventional methods was more than ~50 % [152]. Furthermore, at the examined temperature the particles preserved their almost hysteresis-free character, small coercivities, and remanent magnetizations suggesting fast demagnetization processes important for the implementation to the MR applications.
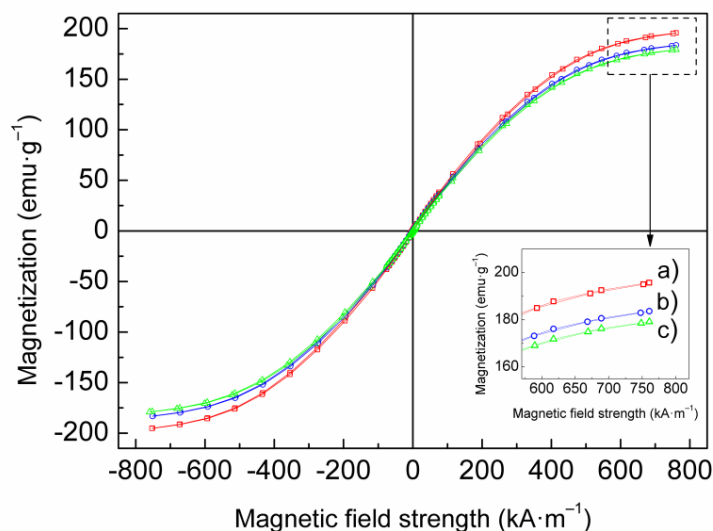

Figure 11. Magnetization curves of bare CI (a), CI-g-PHEMATMS-1 (b), and CI-g-PHEMATMS-2 (c) particles obtained using VSM. The inset figure shows the data differences near the saturation magnetization.

In service life, the MR systems can be exposed to demanding operating conditions such as high temperatures or acidic reactive species (e.g. acid rains, sea humidity, operating fluid leakage), which are key factors affecting their long-

term stability and durability [69]. Therefore, the effects of grafted polymer layers and their thicknesses on thermo-oxidation and chemical stability of the particles were also thoroughly investigated. The thermo-oxidative stability of the particles was analyzed using a thermogravimetric analysis (TGA). Figure 12a summarizes the results obtained by Cvek et al. [89] showing the TGA curves of all the studied particles. Comparing the thermo-oxidation process of bare CI particles and their PGMA- as well as PHEMATMS-grafted analogues a significant difference is notable. The presence of polymeric layers generally shifted the beginning of the particle thermo-degradation to higher temperatures, as these layers effectively shielded the iron core, which is susceptible to oxidize.
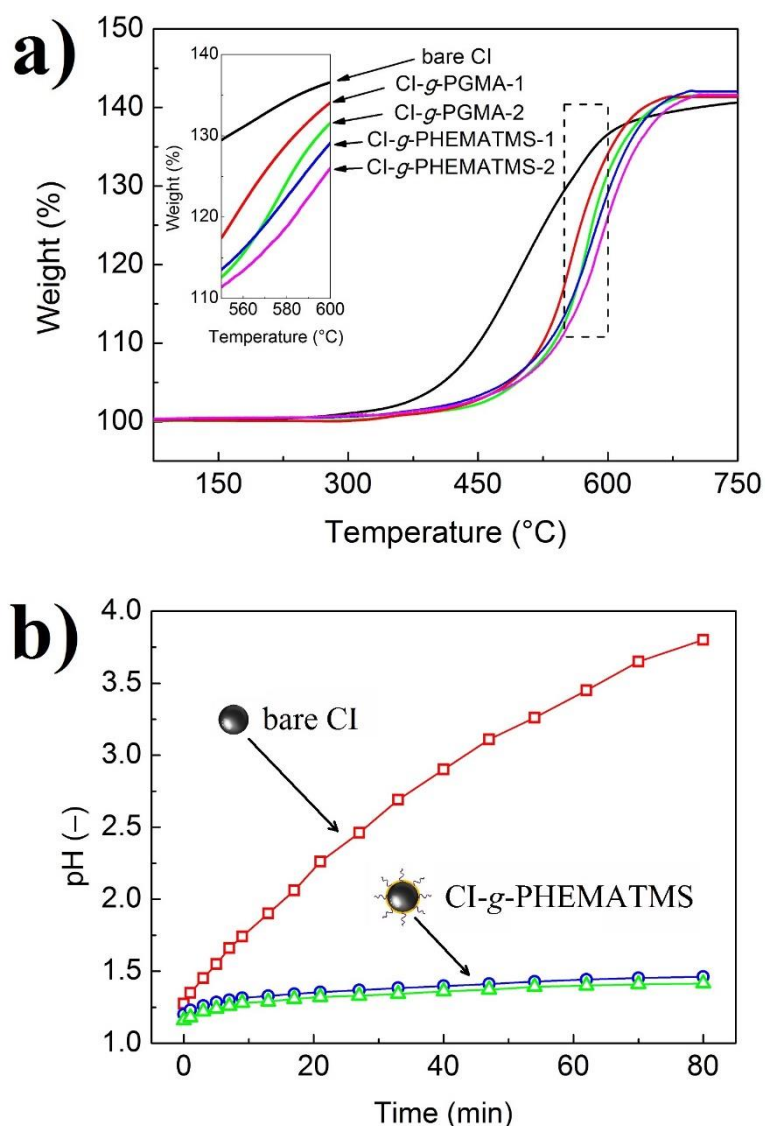
Figure 12. TGA curves of bare CI particles and their PGMA- or PHEMATMS-grafted analogues (a), and the comparison between the chemical stability of bare CI particles and their PHEMATMS-grafted analogues (b).

The chemical stability of the particles was examined by a facile corrosion test [25, 88, 93]. Here, the particles were dispersed in acidic solution (0.1 M HCl), while pH value was recorded as a function of time. As seen in Figure 12b, bare CI particles were relatively unstable in acidic environment, which caused the decrease of their magnetic properties [25, 88]. Both variants of CI-*g*-PHEMATMS particles were however extremely stable indicating that the grafted layers were compact without any defects and the CI core was efficiently protected. The similar results were obtained also for the particles' variant with PGMA shell [88]. To conclude, the presence of polymer shell is necessary to enhance stability of the CI particles (assuming compact shell without defects) but its thickness plays rather a minor role for further stability enhancement.

3.3 Enhancements of the MRSs

Having thermally- and chemically-stable magnetic particles allowed their further implementation into the MR systems. First, the appropriate amounts of either bare CI particles or their PGMA-grafted analogues were dispersed in silicone oil to prepare novel stable MRSs. The effects of the PGMA layers and their thicknesses on the MR performance and sedimentation stability were investigated and the results were introduced in *Paper I – A facile controllable coating of carbonyl iron particles with poly(glycidyl methacrylate): A tool for adjusting MR response and stability properties*. The MR behavior of the representative MRSs was obtained using a rotational rheometer equipped with a magneto-cell device and a parallel plate geometry. Uniformity of the EMF, suspension sedimentation and the wall slip phenomenon as major possible issues related to magnetorheometry were considered [153]. The repeatability and accuracy of the data was ensured by following the protocol [33]. Figure 13 illustrates the rheological data obtained from the CSR mode for the examined MRSs. Generally, the obtained flow curves exhibited typical characteristics for the MRSs. In the off–state, almost Newtonian-like behavior corresponding to the linear increase of the τ with $\dot{\gamma}$ was observed, while in the on–state the yield stress appeared and the τ values of all MRSs dramatically increased and the flow became pseudoplastic [5, 27, 32]. Admittedly, the presence of PGMA grafts slightly increased off–state τ (and consequently η), while decreasing the on–state τ values resulting in a weaker response to the EMF. Nevertheless, these changes were much smaller than those in the conventional core-shell systems [112]. From obvious reasons, the MR behavior was more affected when CI-*g*-PGMA-2 particles were used as a dispersed. In order to exclude possible data distortion, the off–state flow curves are not shown within the whole $\dot{\gamma}$ range due to the inappropriateness of measuring MR geometry for low $\dot{\gamma}$ as was also presented by authors [154]. The results suggest that the MRSs containing PGMA-grafted

particles synthesized via ATRP are able to develop considerable τ sufficient for practical applications.
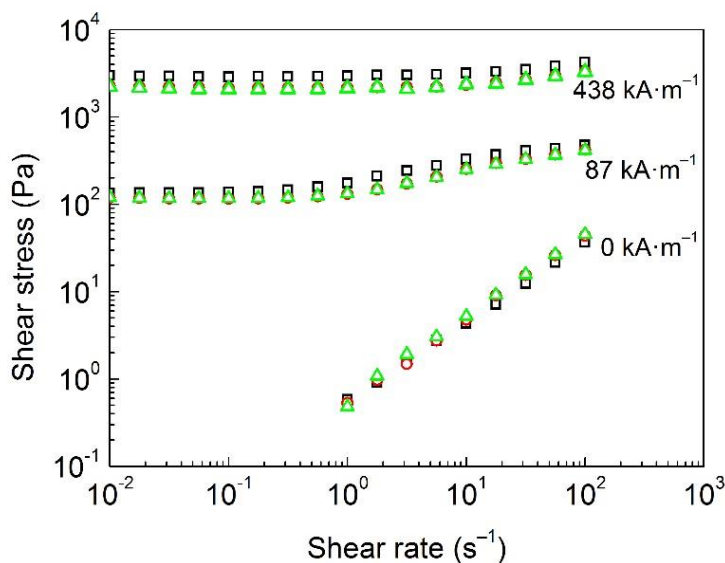

Figure 13. The dependences of the shear stress on the shear rate for the MRSs containing 40 wt.% of bare CI particles (open squares), CI-g-PGMA-1 particles (open circles), or CI-g-PGMA-2 particles (open triangles) under various magnetic field strengths.

Further, the sedimentation stability of tested MRSs was determined via tensiometric method which was recently proposed by Sedlacik et al. [93]. In this method, the funnel-shaped probe hanging up on scales is immersed into the tested suspension, and the weight of settling particles is measured as a function of time (Figure 14). In order to avoid the overfilling of the probe during the experiment, the particle concentration of the MRSs was set to 10 wt.%. Based on the results, it is evident, that the use of CI-*g*-PGMA particles as dispersed phase considerably enhanced sedimentation stability of the MRSs when compared to their analogue containing the same amount of bare CI particles. Improved sedimentation stability is explained as a consequence of PGMA grafts which contributed to reduced bulk density of composite particles and better compatibility with silicone oil [5, 27]. The MRSs containing both variants of PGMA-grafted CI particles exhibited similar sedimentation stability, but the molecular weight of PGMA did not play that important role as expected.
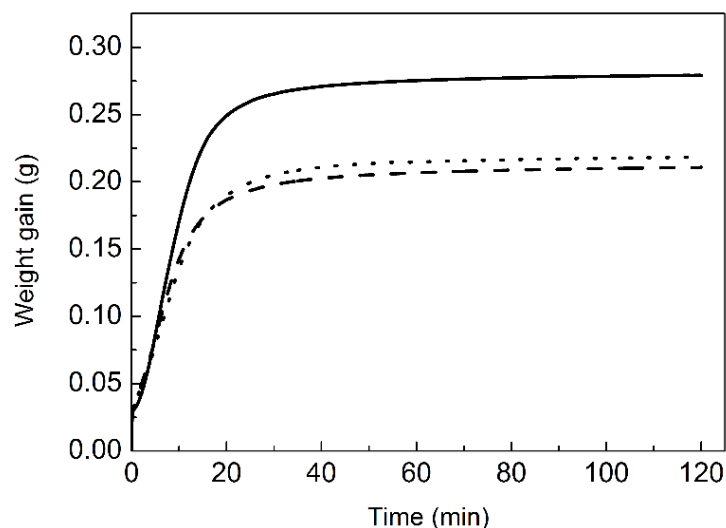
Figure 14. Time dependences of the weight gain representing settled particles of the MRSs containing 10 wt.% of bare CI particles (solid line), CI-g-PGMA-1 particles (dotted line), or CI-g-PGMA-2 particles (dashed line).

Apart from the industrial applications, the magnetic particles have also found promising applicability in biomedical field, e.g. in cancer therapy through embolization of blood veins [155], local drug delivery [156], or cell therapy [157]. According to Silva et al. [156] the use of magnetic microparticles (0.5–5 μm) is necessary in order to target organs that lie deeply in the body cavity (8–12 cm from the body surface). Besides that, the PGMA contains the oxyrane group preserving the possibility to bond other substances [150] and recently this polymer has gained interest in drug and biomolecule binding. Therefore, the special combination of materials such as CI-*g*-PGMA particles was investigated as a versatile platform for possible biomedical applications in ***Paper II – The chemical stability and cytotoxicity of carbonyl iron particles grafted with poly(glycidyl methacrylate) and the magnetorheological activity of their suspensions***. It was found, that CI-*g*-PGMA exhibited extremely enhanced anti-acid-corrosion stability enabling their potential application in medicine. Further, their cytotoxicity was tested according to the international standard EN ISO 10993-5 using NIH/3T3 mouse embryonic fibroblast cell line (ATCC, CRL-1658). Subsequently, the particle extracts were prepared according to ISO 10993-12 and diluted to desired concentrations. The cytotoxicity was evaluated using MTT assay, which was analyzed using microscopic observations. Figure 15a–c shows the effect of extracts on the cell morphologies. Based on the results it was stated that bare CI particles as well as their PGMA-grafted analogues belong to the category with the absence of cytotoxicity within the whole tested concentration range (please see full paper [88]). The absence of cytotoxicity,

gained functionality, and preserved magnetization make the CI-*g*-PGMA particles very interesting from a further research point of view. Moreover, the rheological data of the investigated MRSs was analyzed using the H–B model (Eq. 4) in order to predict possible τ_0 decrease caused by the polymer grafts. Due to controllable coating via ATRP the decreases in magnetization were considered to be insignificant, which was also reflected in slightly decreased MR efficiency (Figure 15). Assuming these findings CI-*g*-PGMA particles prepared via ATRP were found to be a versatile material for miscellaneous applications.
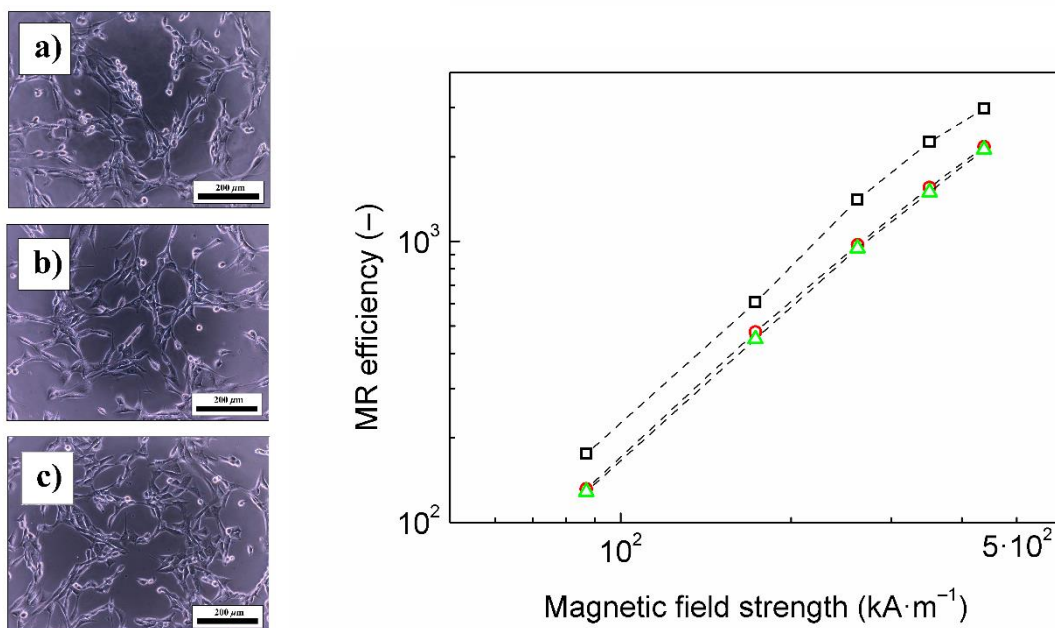
Figure 15. Micrographs of the NIH/3T3 mouse embryonic fibroblast cell line treated with 50% CI extract (a), 50% CI-g-PGMA extract (b), and the reference (c). On the right-hand side the dependences of the MR efficiency on the applied magnetic field strength for the MRSs containing 40 wt.% of bare CI particles (open squares), CI-g-PGMA-1 particles (open circles) or CI-g-PGMA-2 particles (open triangles) are presented.

3.4 Enhancements of the MREs

To fabricate the MREs with enhanced thermo-oxidation stability, chemical stability and improved particle/matrix compatibility the CI-*g*-PHEMATMS particles were used as a suitable filler. This topic was the main objective of ***Paper III – Synthesis of silicone elastomers containing silyl-based polymer-grafted carbonyl iron particles: An efficient way to improve magnetorheological, damping and sensing performances.*** In this research, the desired amounts of either bare CI particles or their PHEMATMS-grafted analogues were mixed with the silicone elastomer/curing agent components and

the homogeneous mixture was casted into the molds. The curing process was accelerated by elevated temperature (100 °C) to obtain the isotropic MREs (concentration of 60 wt.%). The representative samples were tested using a rotational rheometer under dynamic conditions and the applied EMF. Firstly, the γ-sweeps under various H were performed to ensure that all the data fall into the LVR. Besides the MR effect, the MREs were shown to exhibit magnetostriction and hysteresis behavior [60] when exposed to the ascending/descending EMFs. The shear viscoelastic moduli, and damping factor were recorded as a function of H (Figure 16), while the ascending/descending character of H was denoted by the arrows.

As clearly seen, the fabricated MREs exhibited characteristics typical for magnetically-active materials. Expectedly, the G' of neat matrix (as a reference) was independent on the applied H due to the absence of magnetic particles, however the G' distinctly increased by the addition of the CI particles, since rigid inorganic particles have a much higher stiffness than PDMS matrix [158]. The increase of G' with the H was explained by gradual particle magnetization and magnetic interactions resulting in particle network formation, which reinforced the MRE structure. For more details, please see the full article by Cvek et al. [28].
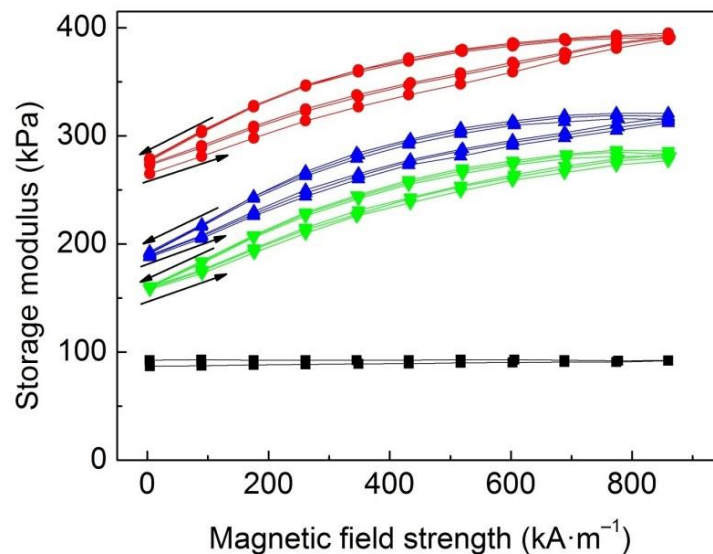

Figure 16. The storage modulus of neat matrix (black squares), and the MREs containing bare CI (red circles), CI-g-PHEMATMS-1 (blue up–triangles), and CI-g-PHEMATMS-2 (green down–triangles) particles as a function of applied magnetic field strength.

In particular, the effect of PHEMATMS coating on viscoelastic properties of the MREs was evaluated. Surprisingly, the highest G' possessed the MRE containing bare CI, which was explained as a consequence of possible particle/matrix chemical bonding as the hydroxyl groups on the surface of bare CI particles can form covalent bonds with the silane-groups of PDMS curing agent [70]. Nevertheless, it is believed that the bonding was limited due to poor adhesion between these two materials. As it was shown via microstructure analysis (Figure 17), bare CI particles were surrounded by the cavities as the result of particle/matrix incompatibility, which can consequently lead to decreased mechanical properties over time. However, the presence of PHEMATMS grafts increased the particle mobility within the matrix most likely due to loosen PDMS cross-link density in the vicinity of the particles. This phenomenon was more pronounced in the MRE containing CI-*g*-PHEMATMS-2 due to the presence of longer grafts, which enabled higher relative particle motions as a reaction on the applied EMF. As a result, the MREs containing the PHEMATMS-grafted particles ultimately exhibited increased relative MR effects (Eq. 1), which suggests their versatility and better practical applicability.
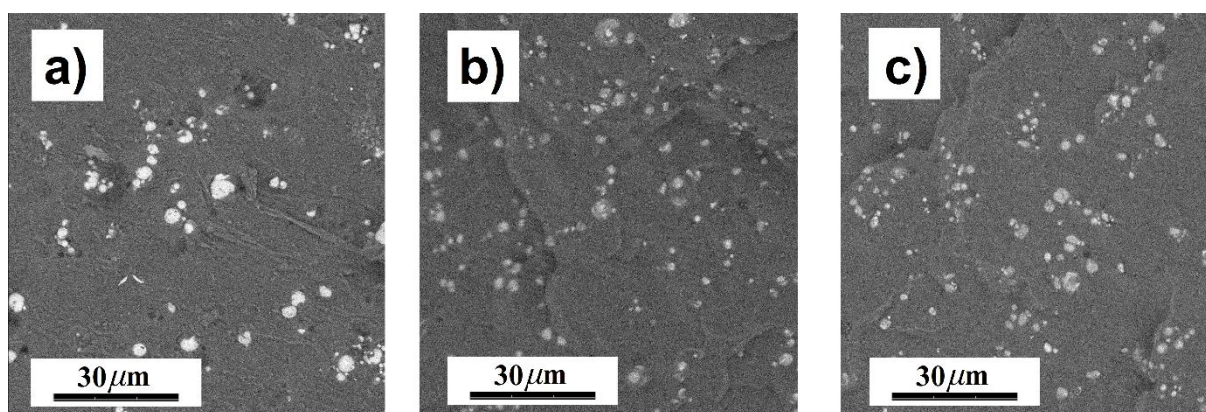

Figure 17. SEM images of the isotropic MREs containing bare CI (a), CI-g-PHEMATMS-1 (b) and CI-g-PHEMATMS-2 (c) particles.

The principal function of the MREs is their damping capability. As indicated above (Chapter 1.8.2), the damping control of the MREs can be executed via the incorporation of temperature-sensitive components [72] or by the addition of plasticizers [71]. These approaches may fail when the MREs are exposed to demanding operating conditions such as extreme temperatures or chemically-contaminated environment inducing the particle degradation and the loss of the MR performance. Therefore, the main idea of ***Paper IV – Tailoring performance, damping and stability properties of magnetorheological elastomers via particle-grafting technology*** was to develop a new concept, which allows enhancing the both, performance and stability properties of the MREs preferably in a single-step procedure. The stability properties of modified particles were assessed above (Figure 12). However, the PHEMATMS-grafted particles were found to be multi-

functional because due to the different molecular weight of PHEMATMS they modulated particle/matrix interface to a different degree. Therefore, the effect of the PHEMATMS molecular weight on the damping capability was investigated. As known [159], total damping capability of the MREs includes several damping mechanisms, namely damping by the viscous flow of the rubber matrix, interfacial damping and magnetism-induced damping. As demonstrated in Figure 18, the viscous flow mechanism appeared to be the main contribution for damping as the neat PDMS exhibited the average damping factor of ~0.155. The inclusion of particles generally enabled additional damping mechanisms. The incorporation of bare CI particles, however decreased the damping capability, which straighten the theory that bare CI particles rather acted as micro-cavities in the body of the matrix. The presence of CI-*g*-PHEMATMS particles increased particle mobility and energy dissipation (associated with interfacial slipping between particles and the matrix) and impressive enhancements of damping properties occurred. The MRE containing the CI-*g*-PHEMATMS-2 particles achieved the average damping factor of ~0.234, which was explained as a consequence of the additional interfacial friction as a result of higher molecular weight PHEMATMS grafts occurrence. Finally, herein fabricated MREs exhibited similar damping capability as the recently-reported ones [31], however at lower particle fractions, which offers not only high MR performance, but also light weight and enhanced stability properties.
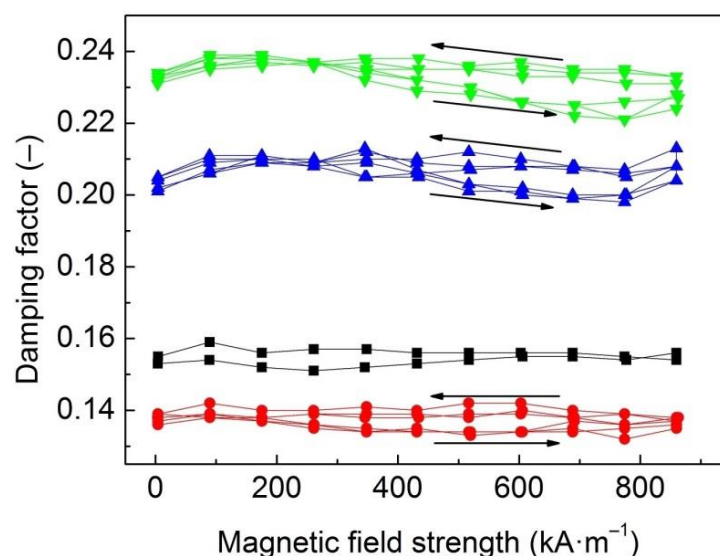

Figure 18. The damping factor of neat matrix (black squares), and the MREs containing bare CI (red circles), CI-g-PHEMATMS-1 (blue up–triangles), and CI-g-PHEMATMS-2 (green down–triangles) particles as a function of applied magnetic field strength.

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3.5 Stabilization of MRSs using additives

The incorporation of submicron-sized additives into the MRSs is known as a facile approach to enhance their sedimentation stability [96-98], redispersibility [104] and in some cases also the MR performance [160]. Using the additives, no special or toxic chemicals are needed making this method feasible and preferable for large-scale applications. As reviewed above (Chapter 1.6.3), many different materials have been used as possible additives to stabilize the MRSs. It is important to mention that there is no standardized procedure to evaluate the effects of the individual additives, thus their unbiased efficiency is still unclear. Therefore, the motivation of ***Paper V – A systematical study of the overall influence of carbon allotrope additives on performance, stability and redispersibility of magnetorheological fluids*** was to introduce different shaped carbon additives (fullerene powder, carbon nanotubes, graphene nanoplatelets) into the MRSs and to methodically investigate their effects under defined conditions with the help of mathematical modelling and complementary experimental techniques. The MR performance of the MRSs was analyzed using the R–S model (Eq. 5) providing reliable data fits. It was found, that even 1 wt.% of the additives inevitably increased the off–state shear stress values of the MRSs. At the on–state, two different manifestations of the additives were distinguished. While the presence of fullerene powder anchored the field-induced CI particle structures possessing the "gap-filling" effect, the other employed additives rather disrupted the particle structures decreasing the obtained shear stress values. The differences were studied in the CSR mode, but they were also apparent under periodically changing magnetic field (Figure 19).
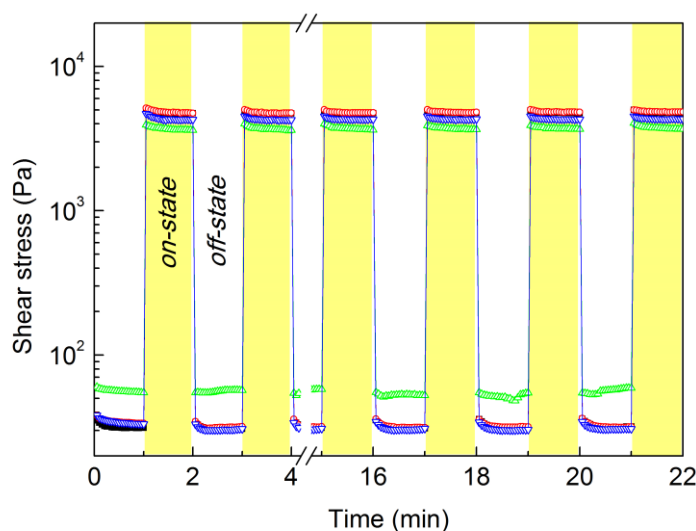

Figure 19. The shear stress vs. time dependences during periodically switching off/on magnetic field (~288 kA·m^{-1}) at a shear rate of 50 s^{-1} for the reference sample (black), and the MRSs containing 1 wt.% of fine fullerene powder (red), the carbon nanotubes (green), or the graphene nanoplateles (blue).

On the contrary, the carbon nanotubes and graphene nanoplateles as the additives decreasing the MR performance were able to enhance the sedimentation stability of the MRSs. The most significant stabilization effect was achieved using the carbon nanotubes, which were able to create a 3D network preventing the CI particle sedimentation. This result was complementarily proved by both, the conventional long-term direct observation and the advanced optical analyzer, i.e. Turbiscan. Finally, the recommendations to design the MRSs for practical applications were given. Based on the results, the optimization of the MRSs behavior could be based on combining the additives varying in the mechanism of their action (gap-filling, sedimentation enhancing effect) to ensure the both, rigidity of the internal structures as well as the sufficient sedimentation stability.

4. Contributions to the Science and Practice

The results obtained within this doctoral study will be beneficial to scientific community especially due to presented approach in particle's modifications using surface-initiated ATRP from the magnetic substrate leading to a design of particle's properties towards a specific application.

The achieved results were already presented at several international conferences and published in high-quality international peer-reviewed journals registered in *Scopus* and *Web of Science* databases. Some of the author's publications have already been numerously cited by foreign scientists demonstrating the interest in the subject and confirming its scientific relevance.

From the practical perspectives, the long-term stability enhancement of newly-designed MR systems can be pointed out, which is currently a very desirable aspect. The novel idea also covers the reliability and durability of the MR systems, which can prolong their service life and, thus, safe costs which were accompanied with their short-term replacements in the past.

The significance of the research outputs is expected to increase in following years due to increasing amount of the commercially available MR devices utilized in real-life applications.

5. CONCLUSIONS

Some of the common drawbacks of the conventional MRSs and MREs were successfully eliminated using a novel approach based on the fabrication of the precisely-defined core-shell structures via surface-initiated ATRP. The particle's modifications were performed in a controlled way, thus the potential of prepared materials exceeded the properties of the existing systems. The physical phenomena related to particle/matrix interaction were clarified, which may affect the development of the next-generation MREs. Although the approach involving ATRP is not straightforward, it offers a great possibility to enhance properties of the MR systems in a controlled way, which was not possible to achieve using the conventional methods. Besides the presented applications, the ATRP may found utilization in other research areas such as biology and medicine in which well-defined modification of magnetic substrate is necessary. Finally, the introduced methodological approach to evaluate the performance and stability of the MRSs may serve as a suitable tool to test the specific effect of the individual additives incorporated in the MRSs and to optimize these for practical applications.

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7. LIST OF FIGURES

CURRICULUM VITAE

Personal Information

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Address	Mistřice 404, 687 12 Bílovice, Czech Republic
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Nationality	Czech
Date of Birth	21/10/1989

Work Experience

Dates	01/2015–present
Occupation or Position Held	Junior Researcher
Name of Employer	Tomas Bata University (TBU) in Zlín, Centre of Polymer Systems
Dates	09/2014–12/2014
Occupation or Position Held	Technical Staff
Name of Employer	TBU in Zlín, University Institute

Education

Dates	2014–present
Title of Qualification Awarded	Doctoral Study – Defended in 09/2018
Principal Branch	Technology of Macromol. Compds. – P2808/2808V006
Organization Providing Education	TBU in Zlín, Faculty of Technology
Dates	2012–2014
Title of Qualification Awarded	Ing. (equivalent to M.Sc.)
Principal Branch	Materials Engineering – N2808/3911T011
Organization Providing Education	TBU in Zlín, Faculty of Technology
Dates	2009–2012
Title of Qualification Awarded	Bc.
Principal Branch	Chemistry and Materials Technology – B2808/2808R009
Organization Providing Education	TBU in Zlín, Faculty of Technology

Training Abroad

Dates	May–July 2018 (3 months)
	Scientific Mobility at University of Granada, Department of Applied Physics (Spain)
Field of Study	Advanced Phenomena Occurring in the MR Systems

Dates	March–April 2017 (2 months)
	Student Intership at Johannes Kepler University Linz, Institute of Polymer Science (Austria)
Field of Study	Processing of Polymer Materials
Dates	October–November 2015 (1 month)
	Research Period at Slovak Academy of Sciences, Polymer Institute (Slovakia)
Field of Study	Modification of Substrate via Controlled Radical Polymerization
Dates	June 2013 (2 weeks)
	Short-Term Study Stay at Clermont-Ferrand Superior National School of Chemistry (France)
Field of Study	Polymer Processing and Functionalization
Expertise Fields	Magnetorheology, Rheology, Particulate Systems, Polymer Processing, Mechanical Properties of Polymers
Other Languages	**Level**
English	C1
Russian	A2
Awards 2017	Best Poster Award (co-author) – Miskolc (Hungary), Conference Paper "*Ferrous Oxalate Micro-Rods as a Promising Material for Electrorheology*"
2017	Aktion Österreich-Tschechien, AÖCZ-Semesterstipendien
2016	First-place Award at the Scientific conference held at the Faculty of Technology of TBU in Zlín
2014	Award by the Head of the Department of Physics and Materials Engineering for excellent Diploma thesis
2014	Best Poster Award – Granada (Spain), Conference paper "*Magnetorheology of Suspensions Based on Carbonyl Iron Particles Coated with Poly(Glycidyl Methacrylate)*"
2012	Award by the Dean of the Faculty of Technology of TBU in Zlín for excellent academic performance during the entire period of studies
Professional Service	Reviewer: *Journal of Industrial and Engineering Chemistry* *Journal of the Taiwan Institute of Chemical Engineers*

LIST OF PUBLICATIONS

Publications in the Journals with Impact Factor:

1. CVEK, M.; MRLIK, M.; ILCIKOVA, M.; SEDLACIK, M.; MOSNACEK, J. Tailoring performance, damping and stability properties of magnetorheological elastomers via particle-grafting technology. *Submitted manuscript.*

2. KUTALKOVA, E.; PLACHY, T.; OSICKA, J.; CVEK, M.; MRLIK, M.; SEDLACIK, M. Electrorheological behavior of fluids containing iron(II) oxalate micro-rods. *RSC Adv.* **2018**; vol. 8, pp. 24773–9, IF 2017 = 2.936.

3. MOUCKA, R.; SEDLACIK, M.; CVEK, M. Dielectric properties of magnetorheological elastomers with different microstructure. *Appl. Phys. Lett.* **2018**; vol. 112, 122901, IF 2017 = 3.495.

4. CVEK, M.; MRLIK, M.; MOUCKA, R.; SEDLACIK, M. A systematical study of the overall influence of carbon allotrope additives on performance, stability and redispersibility of magnetorheological fluids. *Colloids Surf., A*, **2018**; vol. 543, pp. 83–92, IF 2017 = 2.829.

5. MRLIK, M.; CVEK, M.; OSICKA, J.; MOUCKA, R.; SEDLACIK, M.; PAVLINEK, V. Surface-initiated atom transfer radical polymerization from graphene oxide: A way towards fine tuning of electrical conductivity and electro-responsive capabilities. *Mater. Lett.* **2018**; vol. 211, pp. 138–41, IF 2017 = 2.687.

6. CVEK, M.; MOUCKA, R.; SEDLACIK, M.; PAVLINEK, V. Electromagnetic, magnetorheological and stability properties of polysiloxane elastomers based on silane-modified carbonyl iron particles with enhanced wettability. *Smart Mater. Struct.* **2017**; vol. 26, 105003 (10pp), IF 2017 = 2.963.

7. CVEK, M.; MOUCKA, R.; SEDLACIK, M.; BABAYAN, V.; PAVLINEK, V. Enhancement of radio-absorbing properties and thermal conductivity of polysiloxane-based magnetorheological elastomers by the alignment of filler particles. *Smart Mater. Struct.* **2017**; vol. 26, 095005 (7pp), IF 2017 = 2.963.

8. CVEK, M.; MRLIK, M.; ILCIKOVA, M.; MOSNACEK, M.; MUNSTER, L.; PAVLINEK, V. The synthesis of silicone elastomers containing silyl-based polymer-grafted carbonyl iron particles: An efficient way to improve magnetorheological, damping and sensing performances. *Macromolecules.* **2017**; vol. 50(5), pp. 2189–200, IF 2017 = 5.914.

9. PLACHY, T.; CVEK, M.; KOZAKOVA, Z.; SEDLACIK, M.; MOUCKA, R. The enhanced MR performance of dimorphic MR suspensions containing either magnetic rods or their non-magnetic analogs. *Smart Mater. Struct.* **2017**; vol. 26, 025026 (8pp), IF 2017 = 2.963.

10. CVEK, M.; MRLIK, M.; PAVLINEK, V. A rheological evaluation of steady shear magnetorheological flow behavior using three-parameter viscoplastic models. *J. Rheol.* **2016**; vol. 60(4), pp. 687–94, IF 2016 = 3.136.

11. MRLIK, M.; ILCIKOVA, M.; CVEK, M.; PAVLINEK, V.; ZAHORANOVA, A.; KRONEKOVA, Z.; KASAK, P. Carbonyl iron coated with a sulfobetaine moiety as a biocompatible system and the magnetorheological performance of its silicone oil suspensions. *RSC Adv.* **2016**; vol. 6(39), pp. 32823–30, IF 2016 = 3.108.

12. CVEK, M.; MRLIK, M.; ILCIKOVA, M.; MOSNACEK, J.; BABAYAN, V.; KUCEKOVA, Z.; HUMPOLICEK, P.; PAVLINEK, V. The chemical stability and cytotoxicity of carbonyl iron particles grafted with poly(glycidyl methacrylate) and the magnetorheological activity of their suspensions. *RSC Adv.* **2015**; vol. 5(89), pp. 72816–24, IF 2015 = 3.289.

13. CVEK, M.; MRLIK, M.; ILCIKOVA, M.; PLACHY, T.; SEDLACIK, M.; MOSNACEK, J.; PAVLINEK, V. A facile controllable coating of carbonyl iron particles with poly(glycidyl methacrylate): A tool for adjusting MR response and stability properties. *J. Mater. Chem. C.* **2015**; vol. 3(18), pp. 4646–56, IF 2015 = 5.066.

Original Full Papers in Conference Proceedings (Indexed on Web of Science):

1. CVEK, M.; MRLIK, M.; ILCIKOVA, M.; MOSNACEK, J.; SEDLACIK, M. Electrorheological fluids containing graphene oxide sheets grafted with poly(methyl methacrylate). In Nanocon **2017** – Conference Proceedings – *9th International Conference on Nanomaterials – Research and Application*, p. 87, ISBN: 978-80-87294-78-9.

2. CVEK, M.; MRLIK, M.; ILCIKOVA, M.; MOSNACEK, J.; KRONEK, J.; SEDLACIK, M. Grafting of magnetic particles with poly(2-isopropenyl-2-oxazoline). In Nanocon **2017** – Conference Proceedings – *9th International Conference on Nanomaterials – Research and Application*, p. 123, ISBN: 978-80-87294-78-9.

3. OSICKA, J.; CVEK, M.; MRLIK, M.; ILCIKOVA, M.; PAVLINEK, V.; MOSNACEK, M. Light-induced and sensing capabilities of SI-ATRP modified graphene oxide particles in elastomeric matrix. In: *Proc. SPIE 10164, Active and Passive Smart Structures and Integrated Systems*, **2017**, 1016434.

4. CVEK, M.; MRLIK, M.; ILCIKOVA, M.; MOSNACEK, J.; PAVLINEK, V. Preparation and characterization of graphene oxide sheets controllably grafted with PMMA brushes via surface–initiated ATRP. In Nanocon **2016** – Conference Proceedings – *8th International Conference on Nanomaterials – Research and Application*, p. 116–21, ISBN: 978-808729471-0.

5. MRLIK, M.; ILCIKOVA, M.; CVEK, M.; URBANEK, P.; PAVLINEK, V.; MOSNACEK, J. Controllable reduction of graphene oxide/poly(butyl acrylate) hybrids under ATRP conditions. In Nanocon **2016** – Conference Proceedings – *8th International Conference on Nanomaterials – Research and Application*, p. 104–9, ISBN: 978-808729471-0.

Conference Contributions:

1. CVEK, M.; MRLIK, M.; ZAHORANOVA, A.; SRAMKOVA, P.; SEDLACIK, M.; KRONEK, J. The preparation of stable magnetorheological gels based on the poly(2-oxazoline) matrix (**2018**) – *16th International Conference on Electrorheological Fluids and Magnetorheological Suspensions*, Maryland, USA.

2. SEDLACIK, M.; CVEK, M.; KRACALIK, M.; MRLIK, M. The recyclable magnetorheological elastomers based on thermoplastic matrix (**2018**) – *16th International Conference on Electrorheological Fluids and Magnetorheological Suspensions*, Maryland, USA.

3. TORRES-MENDIETA, R.; URBANEK, M.; CVEK, M.; HAVELKA, O.; WACLAWEK, S.; PADIL, V.V.T.; JAKSIKOVA, D.; KOTEK, M.; CERNIK, M. Laser-mediated synthesis of Cu by photochemical reduction of metal salts in a continuous flow jet (**2018**) – *XII Reunion Nacional de Optica*, Castellon, Spain.

4. KUTALKOVA, E.; PLACHY, T.; OSICKA, J.; CVEK, M.; SEDLACIK, M. Iron(II) oxalate micro-rods as a promising material for electrorheology (**2018**) – *Polymers: Site of Advanced Horizons and Ambits*, Zlín, Czech Republic.

5. CVEK, M.; MRLIK, M.; SEDLACIK, M. Magnetorheological behavior of poly(2-isopropenyl-2-oxazoline)-grafted magnetic particles in physiological solution (**2018**) – *Polymers: Site of Advanced Horizons and Ambits*, Zlín, Czech Republic.

6. SEDLACIK, M.; CVEK, M.; MOUCKA, R.; SYKOROVA, L.; BILEK, O.; PAVLINEK, V.; SAHA, P. Electromagnetic shielding of elastomers containing particles with enhanced wettability (**2018**) – *Polymers: Site of Advanced Horizons and Ambits*, Zlín, Czech Republic.

7. CVEK, M.; PLACHY, T.; OSICKA, J.; KUTALKOVA, E.; MRLIK, M.; SEDLACIK, M. Ferrous oxalate micro-rods as a promising material for electrorheology (**2017**) – *3rd International Conference on Rheology and Modeling of Materials*, Miskolc-Lillafured, Hungary.

8. CVEK, M.; MRLIK, M.; SEDLACIK, M. Effect of carbon nano-additives on performance and stability properties of magnetorheological fluids (**2017**) – *3rd International Conference on Rheology and Modeling of Materials*, Miskolc-Lillafured, Hungary.

9. CVEK, M.; MRLIK, M.; ILCIKOVA, M.; SEDLACIK, M.; MOSNACEK, J.; PAVLINEK, V. Magnetorheological elastomers with tailored performance and stability properties based on multifunctional core-shell particles with various shell thickness (**2017**) – *3rd International Conference on Mechanics of Composites*, Bologna, Italy.

10. MOUCKA, R.; SEDLACIK, M.; CVEK, M.; PAVLINEK, V. Dielectric behavior of structured magnetorheological elastomers (**2017**) – *3rd International Conference on Mechanics of Composites*, Bologna, Italy.

11. CVEK, M.; MRLIK, M.; ILCIKOVA, M.; MOSNACEK, J.; PAVLINEK, V. Tunable damping of magnetorheological elastomers based on particles controllably grafted with polymer chains (**2016**) – *XXII. Bratislava International Conference on Macromolecules*, Bratislava, Slovakia.

12. CVEK, M.; MRLIK, M.; ILCIKOVA, M.; MOSNACEK, J.; PAVLINEK, V. Magnetorheological elastomers based on carbonyl iron particles grafted with trimethylsilyloxyethyl methacrylate (**2016**) – *15th International Conference on Electrorheological Fluids and Magnetorheological Suspensions*, Incheon, Korea.

13. CVEK, M.; MOUCKA, R.; SEDLACIK, M.; BABAYAN, V; PAVLINEK, V. Shielding and magnetorheological performance of elastomers containing silane–modified carbonyl iron particles (**2016**) – *15th International Conference on Electrorheological Fluids and Magnetorheological Suspensions*, Incheon, Korea.

14. CVEK, M.; MRLIK, M.; PAVLINEK, V. On the modelling of rheological behavior of MRFs as Robertson–Stiff fluids (**2015**) – *2nd International Conference on Rheology and Modeling of Materials*, Miskolc-Lillafured, Hungary.

15. CVEK, M.; MRLIK, M.; ILCIKOVA, M.; PLACHY, T.; SEDLACIK, M.; MOSNACEK, J.; PAVLINEK, V. Magnetorheology of suspensions based on carbonyl iron particles coated with poly(glycidyl methacrylate) (**2014**) – *14th International Conference on Electrorheological Fluids and Magnetorheological Suspensions*, Granada, Spain.

Work on Projects:

Project	Period	Position
EU Funds – OP Research, Development and Education in cooperation with Ministry of Education, Youth and Sports, *International Mobility of TBU in Zlín Researchers*, CZ.02.2.69/0.0/0.0/16_027/0008464	2018	Member of Research Team
Internal Grant Agency of TBU in Zlín, *Preparation and Characterization of Electrorheological Suspensions Based on Ferrous Oxalates and Clays*, IGA/CPS/2018/004	2018	Member of Research Team
Czech Science Foundation (GACR), *Novel Magnetorheological Elastomers Based on Modified Magnetic Fillers*, 17-24730S	2017–2019	Member of Research Team
Internal Grant Agency of TBU in Zlín, *Evaluation of Carbon-Based Nano-Additives' Influence on Magnetorheological Performance and Preparation of Electrorheological Suspension with Enhanced Sedimentation Stability*, IGA/CPS/2017/004	2017	Principal Investigator
Czech Science Foundation (GACR), *Smart systems based on modified graphene oxide particles*, GA0/GJ, GJ16-20361Y	2016–2018	Member of Research Team
Internal Grant Agency of TBU in Zlín, *Magnetorheological Elastomers Based on Modified Filler and Preparation of Advanced Suspensions of Magnetic Particles*, IGA/CPS/2016/008	2016	Principal Investigator
Ministry of Education Youth and Sports (MSMT), *Centre of Polymer Systems plus*, LO1504	2015–2020	Member of Research Team
Technology Agency of the Czech Republic (TACR), *Development of Polyurethane Matrixes for Composite Production*, PURKOMP, TH01011438	2015–2017	Member of Research Team
Ministry of Education Youth and Sports (MSMT), *Centre of Polymer Systems*, CZ.1.05/2.1.00/03.0111	2011–2015	Member of Research Team
Internal Grant Agency of TBU in Zlín, *Electrorheological Fluids with Enhanced Sedimentation Stability and Dimorphic Magnetorheological Suspensions*, IGA/CPS/2015/007	2015	Member of Research Team
Internal Grant Agency of TBU in Zlín, *Modification of Particles for Intelligent Systems with Higher Efficiency and Enhanced Stability*, IGA/FT/2014/017	2014	Member of Research Team

Martin Cvek

Magnetorheological Systems with Optimized Performance

Magnetoreologické systémy s optimalizovaným výkonem

Doctoral Thesis Summary