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Research Article

Reinforced, Extruded, Isotropic Magnetic Elastomer Composites: Fabrication and Properties

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The study involves the development of isotropic magnetorheological elastomer composites (i-MREs) with improved mechanical properties, their preparations, and properties characterizations. The novelty of the research is the use of extrusion process in the preparation of composites containing two different fillers: carbonyl iron powder (CIP) as magnetic filler and carbon black (FEF, N550) as reinforcing one. So far, the process of extrusion has been used to prepare magnetic composites without filler that improve mechanical properties. It is worth mentioning that the polymer matrix (ethylene-octene copolymer, EOR) offers excellent performance in extrusion applications. In this research, two methods of magnetic composites preparation were reported: traditional, during two-roll mill, and a new one using extrusion process. It was found that the usage of new processing technology allowed forming more homogenous dispersion of particles in elastomer matrix and oriented polymer chains, resulting in an improved rheometric characteristic, increased crosslink density, and decrease of the storage modulus (Payne effect). Based on both static/dynamic mechanical properties and damping properties under the influence of compression stress, the positive influence of extrusion process on material characteristics was confirmed. Moreover, all composites proved very good magnetic properties and resistance to thermooxidative ageing.

1. Introduction

Magnetorheological elastomers (MREs) belong to a class of smart materials in which rheological properties can rapidly change under the influence of a magnetic field. The interest in this type of materials is constantly increasing due to the inexhaustible possibilities of their applications. In addition, compared to the well-known magnetorheological fluids [MRFs] there is still a lot to improve in terms of both material and processing issues [1–4].

MREs should be classified into two categories: anisotropic and isotropic type. The anisotropic MRE has aligned chain-like structure of the magnetic particles as the magnetic field is induced during vulcanization [5]. Meanwhile, the magnetic particles in the isotropic MRE are uniformly dispersed in the matrix [6]. Change of material stiffness in various operating conditions is the basis for many MRE applications; for example, in variable-stiffness devices, tunable vibration absorbers and damps the components [7, 8]. The capability of fast, reversible, and large deformation has made these materials promising candidates for next generation of remotely controlled actuators or sensors [9, 10]. The large field-induced volumetric strain of a porous magneto-active polymer may also be used in drug delivery systems [11]. However, there are still some problems, especially with mechanical and damping properties, which limit their industrial application. Generally, MREs are fabricated with magnetic particles and elastomers; thus most works on improving the properties of MREs focus on these two aspects [12].

The vast majority of MREs described in the literature contain carbonyl iron powder (CIP) particles added into the elastomer matrix [13–15]. There is also a group of magnetorheological elastomers produced with the usage of $COFe_2O_4$ [16], SiC [17], Ni [18], $FeCo_3$ [19], and Fe [20]. Aside from the aspects related to the magnetic particles, the selection of matrix type also plays an important role in the MRE properties. Various types of elastomer matrices have been explored by many researchers, including synthetic [21–24] and natural rubber [25] and/or mixtures of both [26]. However, each of these polymers has its limitations including poor aging resistance, weathering, resilience to wide temperature range, relatively low resistance to oil, and weak mechanical properties, which cannot satisfy the requirements of applications subjected to high impact loading. That is why, the search for new polymer matrices with peculiar properties is essential.

The mechanical and damping properties of MREs depend not only on the types of rubber matrix and magnetic particles, but also on the presence of different additives polymer mixtures. They can serve as reinforcing particles, affecting adhesion between fillers and the rubber matrix [27, 28]. Carbon black is selected as an additive to modify and improve MRE mechanical properties, since it is an important reinforcing filler in polymer engineering, especially in rubber technology [29].

Based on the survey of the subject literature [1, 2, 4, 5] it can be concluded that it is easier and cheaper to manufacture isotropic than anisotropic elastomers. The mechanical properties of the former offer prospects of wide industrial application. Therefore the aim of this research was to fabricate and determine the mechanical properties of selected isotropic magnetic elastomer composites. The literature clearly stresses the importance of particle dispersion in isotropic composites, which significantly affects properties of smart materials, especially their mechanical properties. However, the problem of improving the mechanical characteristics of MREs is still unsolved. Moreover, there is lack of information about the influence of different processing techniques and their parameters on the properties of magnetorheological composites, hence, the idea of using the extrusion process as an alternative method of preparing MRE composites. There are only a few works in the literature containing a description of this technique, e.g., concerning the manufacture of magnetic filaments for 3D printing [30].

From the authors previous study it follows that the application of extrusion for ethylene-octene rubber composites with the addition of various magnetic fillers can help achieve an appropriate degree of particle dispersion influencing mechanical properties of composites [31]. Based on their experience, the authors decided to choose one of the previously characterized systems (ENGAGE/CIP 60 phr) and apply it to reinforce carbon black. Moreover a new test method for MRE characterization, determining the relative damping coefficient under the shear stress conditions, has also been added. Most of MRE research works present in the literature concentrate on obtaining materials with improved magnetic characteristics and mechanical or damping properties being often overlooked. It indicates that this topic may prove to be extremely valuable in the context of later application of extruded i-MRE.

In this work the authors focused on obtaining magnetorheological elastomer composites with improved mechanical and magnetic properties as well as the degree of fillers dispersion. The study involves the development of MREs based on carbonyl iron powder (CIP) as magnetoactive filler in a new, nonpolar, ethylene-octene elastomer, which would also present good ageing resistance. To achieve better mechanical characteristic of MREs, FEF (Fast Extrusion Furnace) N550 was added during the composites preparation.

Table 1: Composition of elastomer mixtures.

Sample	EOR	DCP	CIP	FEF N550
		phr		
0	100	2	-	-
1	100	2	60	-
2	100	2	60	5
3	100	2	60	10
4	100	2	60	15
5	100	2	60	20

phr: per hundred rubbers

Application of extrusion process was used as an additional processing method before typical vulcanization of rubber. Firstly, base compositions were mixed using two-roll mill; then one part of the mixture was extruded and next vulcanized. The second part of previously prepared mixture was only vulcanized (without extrusion). The characteristic feature of this processing method is the enhanced chain orientation and improved dispersion of particles, what should impact positively on the properties of the composite.

2. Experimental

2.1. Materials

(1) Elastomer. ENGAGE™ polyolefin elastomer: ethylene-octene copolymer (EOR) containing 25 wt% comonomer octene was obtained from DOW Chemical Company USA. The Mooney viscosity was (ML (1+4) at 121°C: 35).

(2) Cross-Linking Agent. Rubber mixtures were vulcanized with dicumyl peroxide DCP (purity: 98%) produced by Sigma Aldrich. (Aldrich).

(3) Fillers. (i) Carbonyl iron powder (CIP) – av. particle size of 3100 nm was purchased from BASF.

(ii) Carbon Black N550, Fast Extrusion Furnace Black, (FEF, N550), av. particle size of 39-55 nm was obtained from Evonik Degussa GmbH.

The compositions of elastomer mixtures are given in Table 1.

2.2. Methods. Elastomer mixtures, based on ethylene-octene copolymer filled with carbonyl iron powder, carbon black, and cross-linking agent (dicumyl peroxide), were prepared using a laboratory two–roll mill, with roll dimensions of D = 140 mm and L = 300 mm. The rotational speed of the front roll was $V_p = 16\ min^{-1}$; the friction and the width of the gap between rollers were 1-1.2, 1.5-3 mm. The average temperature of the rolls was of about 30°C.

Then, to obtain the oriented structure of polymer chain, one part of each of the elastomer mixtures was subjected to extrusion (in accordance with ASTM D 2233 standard) using a Brabender single screw measuring extruder 19/10 DW (Ribbon die head). Extrusion speed: 30 rpm, residence time in the extruder: 1 min. Extrusion is a widely used method of processing industry, which is important from a technological and practical point of view. One of the characteristic features of this processing method is the enhanced macromolecule chain and/or filler structure orientation and improved particle dispersion, which should have a positive effect on the properties of the composite. The second part of previously prepared mixture was only vulcanized (without extrusion).

The kinetics of rubber vulcanization as well as rheometric properties of compounds were studied using moving die rheometer (Model - MDR) from Alpha Technologies (ISO 6502: Rubber - Guide to the use of curemeters) at 160°C. Determination of minimum torque (M_L), maximum torque (M_H), the time required for the torque to reach 90% of the maximum achievable torque (t90), which is used as an indicator of optimum time cure (tw), were taken from vulcanization curve. The values of torque increase (ΔM) of the composites have been calculated based on the equation (1) [32]:

$$\Delta M = M_H - M_L \tag{1}$$

The vulcanization of the rubber mixtures was performed using steel vulcanization molds placed between the shelves of an electrically heated hydraulic press. The samples were cured at 160°C, at 15 MPa pressure for curing time, which was measured by a rheometer.

The mechanical properties of the vulcanizates were examined by universal testing machine Zwick (RoellGroup, Ulm, Germany), at room temperature with the crosshead speed of 500 mm/min for five dumbbell samples w-3 for each composite (in extrusion direction for extruded samples). The measurements of the vulcanizates were tested according to the standard procedures in ISO 37.

Dynamic mechanical analysis (DMA) was studied with the application of ARES Rheometer (plate-plate system, plate diameter: 20 mm; gap 2 mm). Dynamic mechanical measurements parameters: temperature: 25°C, sample deformation rate: 10 rad/s, stress: from 0.1% to 150%, test force: 5 N. The Payne effect ($\Delta G'$) values of the composites have been calculated based on the equations (2) [33].

$$\Delta G' = G'_{min}\left(\lim 10^{-2}\right) - G_{max}(\infty) \tag{2}$$

$G'_{min}(\lim 10^{-2})$—a composite storage modulus determined under the deformation of 10^{-2}%; $G'_{max}(\infty)$— a composite storage modulus determined under the max deformation.

The crosslink density of the vulcanizates was determined by equilibrium swelling in toluene, based on the Flory-Rehne [34] (equation (3)):

$$\gamma_e = \frac{\ln\left(-1 - V_r\right) + V_r + \mu V_r^2}{V_0\left(V_r^{1/3} - V_r/2\right)} \tag{3}$$

γ_e—the crosslink density (mol/cm^3), V_0—the molecular volume of solvent (106.7 cm^3/mol) μ—the Huggins parameter of the EOR-solvent interaction was calculated from the equation (4) [35]:

$$\mu = \mu_0 + \beta \bullet V_r \tag{4}$$

μ0—the parameter determine of non-cross-linked polymer/solvent relations, β—the parameter determine of cross-linked polymer/solvent relations (μ_0 = 0.478, β =0.404),and Vr—the volume fraction of elastomer in the swollen gel (equation (5)) [34].

$$V_r = \frac{1}{1 + Q_w\left(\rho_k/\rho_r\right)} \tag{5}$$

Q_w—weight of equilibrium swelling, ρ_k— density of rubber (0,868 g/cm^3), and ρ_r— density of solvent (0,86 g/cm^3).

The thermooxidative ageing characteristics were determined according to the PN-82/C-04216 standard. Samples were exposed to air at an elevated temperature (70°C) for 14 days in a dryer FD (Binder), with thermocirculation. To estimate the resistance of the samples to aging, their mechanical properties after aging were determined and compared with the values obtained for vulcanizates before the aging process.

The ageing coefficient (K) was calculated according to the relationship (equation (6)) [36]:

$$K = \frac{(TS \bullet EB)_{after\ aging}}{(TS \bullet EB)_{before\ aging}} \tag{6}$$

where TS is the tensile strength of the vulcanizates and EB is the elongation at break.

Damping properties of composites were determined under the influence of compressive stress using a ZWICK 1435 (Zwick/Roell) [37]. The relative damping value was calculated with the equation (7) (according PN-C-04289:1987) [38]:

$$T_{\tau w} = \frac{\Delta W_i}{W_{ibel}} \bullet 100\% \tag{7}$$

where

$T_{\tau w}$—relative damping;

ΔW_i—the difference between the compression work and the work during reducing the compressive stresses;

W_{ibel}—compression work.

The degree of dispersion of the filler in the elastomer matrix was elevated using a DisperTester 3000 microscope. The measurements of the vulcanizates were tested according to the standard procedures in ISO 11345: 2006 [39]. The instruments precision telecentric optical system uses the reflected light method for obtaining high-resolution reflective images from the sample surface.

The morphology of composites samples (5,20 phr carbon black) were analyzed by means of the SEM, a scanning electron microscopy JEOL 35C (Japan). Before measuring with the SEM, the samples were placed on carbon plasters and coated with the use of carbon target by the Cressington 208 HR system.

Magnetic properties of the samples were measured with vibrating sample magnetometer VSM Lakeshore 7410, in the range of the field (H) up to 960 kA/m.

3. Results and Discussion

3.1. Rheometric Properties of Ethylene-Octene Copolymer Mixtures. The aim of rheometric (vulcametric) measurements is prompt determination of cross-linking (vulcanization)

Table 2: Rheological properties of magnetorheological rubber mixtures filled with carbon black in different contents (0-20 phr) prepared by traditional rubber processing and extrusion process.

Sample	Carbonyl iron powder content [phr]	Carbon black content [phr]	M_L [dNm]	ΔM [dNm]	t_{90} [min]
Composites prepared using traditional rubber processing					
Reference sample	-	-	0.72±0.02	8.6 ±0.2	16.7±0.3
	60	-	0.81±0.05	10.4±0.1	16.0±0.2
Carbon black filled sample	60	5	0.92±0.04	10.6±0.2	16.0±0.3
		10	0.95±0.05	10.8±0.1	16.2±0.4
		15	0.98±0.07	12.0±0.3	16.2±0.4
		20	1.02±0.06	13.8±0.1	16.4±0.2
Composites prepared using extrusion process					
Reference sample	-	-	0.63±0.03	8.6±0.2	16.7±0.4
	60	-	0.81±0.05	10.2±0.2	15.9±0.5
Carbon black filled sample	60	5	0.71±0.05	11.3±0.3	15.8±0.4
		10	0.69±0.06	11.9±0.1	15.9±0.3
		15	0,98±0,05	12,9±0,1	16,0±0,3
		20	1,00±0,04	13,3±0,2	16,3±0,4

M_L: minimum torque;
ΔM: torque increase;
t_{90}: optimum curing time.

properties of rubber compounds. The measurements are intended to control and develop compounds possessing, characterize rheological properties of polymer mixtures, and determine the optimal time of vulcanization.

Table 2 shows rheological properties of ethylene-octene copolymer mixtures (increase in torque during cross-linking, minimum torque, and curing time) filled with magnetoactive filler and carbon black.

Rheometric properties analysis showed that the introduction of carbon black into the elastomer induced an increase in torque increment compared with ΔM values obtained for both reference samples: filled with pure CIP and absolutely unfilled mixture. Furthermore, the higher the amount of carbon black in the composition was, the higher the increase in torque was observed. Such effect is possibly a consequence of higher (more expanded), structure formation of the filler complex in polymer matrix, which also results in an increase in stiffness of the system. Addition of the fillers increases the viscosity of rubber mixtures via a hydrodynamic effect—the rigid (inextensible) particles cannot deform, so their strain is transferred into the polymer chains.

The addition of different type of filler had various effects on kinetic characteristic of composites. The use of CIP as a filler of ethylene-octene rubber reduced the optimum curing time in comparison with the unfilled system. However, the rate of the vulcanization process of mixtures containing carbon black was slightly smaller; thus the curing time of reference sample with magnetoactive filler was longer. The value of t(90) gradually increased with the increasing amount of reinforcing filler. The composite containing maximum amount - 20 phr of carbon black characterized the optimum time of cross-linking that was approximated to the unfilled sample. The differences between rheometric properties of ethylene-octene copolymer mixtures obtained by different processing methods were negligible.

3.2. Static Mechanical Properties and Cross-Linking Density. The influence of different processing methods on the ethylene-octene rubber was estimated based on the mechanical properties: tensile strength (Figure 1), elongation at break, and stress at 100% elongation or cross-linking density of the vulcanizates (Table 3) and stress-strain curves (Figure 2).

As predicted with higher content of carbon black, the mechanical properties of the magnetic composites were improved. Both stress at 100% elongation and tensile strength of the vulcanizates increased with the increased content of the filler. The mechanical properties of the composites obtained by the extrusion process were significantly better than the conventionally produced materials in terms of strength. Additional processing stresses can lead to a better dispersion of the filler, which consists of even distribution and limited agglomeration, thus enhancing the filler-polymer interactions. When molding the pressurized mixture through the extrusion nozzle, the orientation of the polymer chain and the spatial network of the filler in the direction of flow may occur. As a result, in the elastomeric medium an oriented and more developed filler structure was formed, generating a greater reinforcing potential. Strong interactions between the filler and polymer exhibited behaviour similar to physical nodes of the network and act as additional elements in the network of the system. The results of cross-linking density obtained by the equilibrium swelling method supported this thesis. The addition of carbon black resulted in an increase in γe values of composites with respect to the results obtained for the vulcanizate containing only the addition of the magnetic filler and nonfilled composite. Extruded and filled magnetoactive composites exhibited nearly two times higher values of cross-linking density as compared to the vulcanizates prepared by conventional method. In addition, there was a certain amount of soot between the spatially developed carbon black structure called “bound

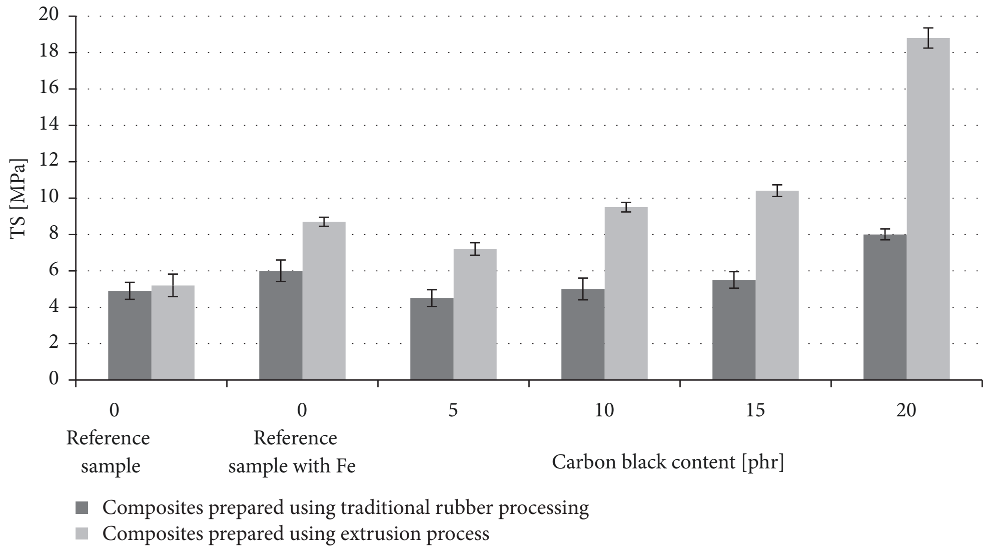

Figure 1: Tensile strength of ethylene-octene composites prepared using extrusion and traditional rubber processing methods.

Table 3: Mechanical properties and cross-linking density of magnetorheological composites prepared by different producing methods.

Sample	Carbonyl iron powder content [phr]	Carbon black content [phr]	SE_{100} [MPa]	EB [%]	TS [MPa]	$\gamma_e 10^{-5}$ [mol/cm^3]
Composites prepared using traditional rubber processing						
Reference sample	-	-	2.9±0.1	482±5	4.9±0.5	3.0±0.2
	60	-	3.2±0.1	294±4	6.0±0.6	2.9±0.3
Carbon black filled sample	60	5	3.4±0.2	177±2	4.5±0.5	3.1±0.1
		10	4.0±0.1	160±7	5.0±0.6	3.7±0.2
		15	4.6±0.1	140±6	5.5±0.5	4.0±0.3
		20	5.6±0.2	137±9	8.0±0.3	5.0±0.2
Composites prepared using extrusion process						
Reference sample	-	-	2.9±0.2	492±11	5.2±0.6	3.0±0.2
	60	-	3.5±0.1	373±5	8.7±0.2	6.2±0.1
Carbon black filled sample	60	5	4.3±0.1	232±4	7.2±0.3	6.8±0.2
		10	4.9±0.1	246±9	9.5±0.3	7.6±0.3
		15	5.4±0.1	227±8	10.4±0.3	8.7±0.4
		20	5.9±0.1	218±4	18.8±0.5	9.8±0.2

SE_{100}: stress at 100% elongation;
EB: elongation at break;
TS: tensile strength;
γe: the crosslinking density of the vulcanizates in toluene.

rubber," which occurred as part of the whole elastomer network and thus increased its volume. As a result of the increased total cross-linking density, the strength of the extruded composites increased more than 100% as the filler-polymer interaction increased. Strong interactions could contribute directly to the loss of elasticity of the polymer at the interface, resulting in a reduced elongation at break of composites.

3.3. Thermooxidative and Ultraviolet Aging Processes. Polymer composites lose their mechanical properties during aging. Small changes in the physical or chemical structures can cause intense variations in the future usability of the composites. The results of mechanical properties of composites subjected to the process of aging are given in Figure 3.

The thermooxidative aging coefficients [K] obtained for pure EOR vulcanizates and the vulcanizate containing only the magnetic filler itself (sample 1) were 0.74 and 0.71, respectively. Therefore it can be stated that the mechanical properties of these samples slightly deteriorated as the result of the thermooxidative aging process. The addition of carbonyl iron did not accelerate the aging process, although there is a widespread belief that metals with varying degrees of oxidation (iron in this case) accelerate aging processes. This is probably due to the absence of Fe^{2+} and Fe^{3+} iron on the

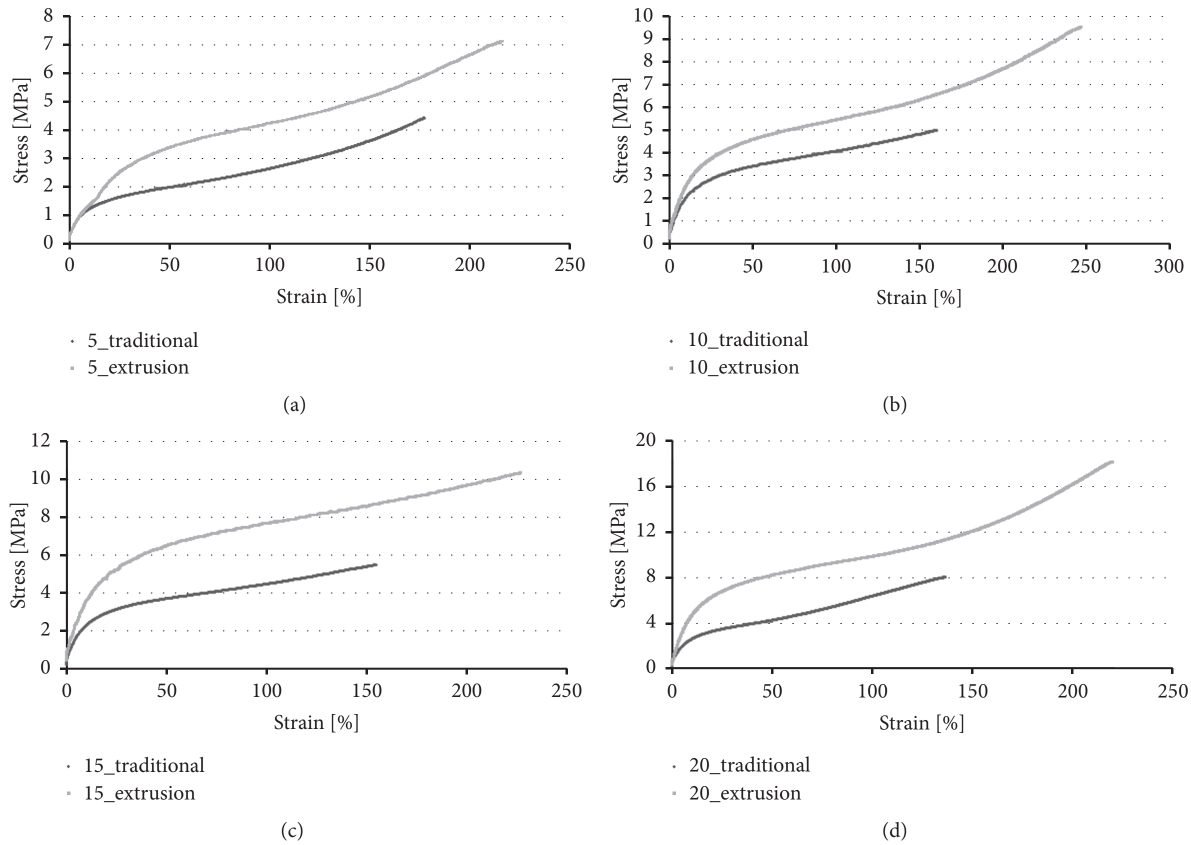

Figure 2: Stress-strain plots of composites filled with 5 phr (a), 10 phr (b), 15 phr (c), 20 phr, and (d) carbon black prepared using extrusion and traditional rubber processing methods.

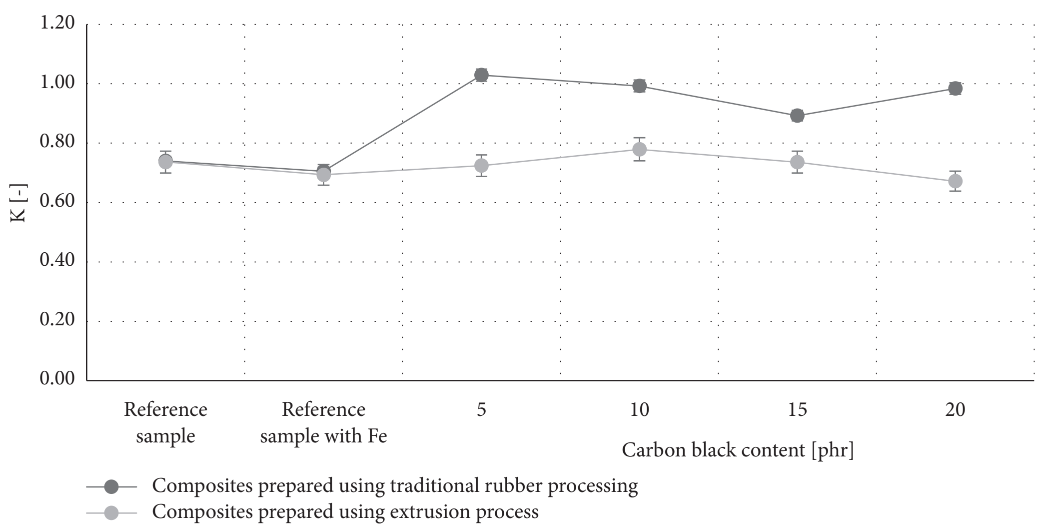

Figure 3: Thermooxidative aging factors of ethylene-octene composites prepared by different producing methods.

surface of vulcanizates, which initiate the degradation of the elastomeric composites [40].

The obtained results show that even the smallest addition of carbon black significantly improves the resistance of composites to thermooxidative aging. The aging coefficients obtained for the filled composites were close to 1, which shows that their mechanical properties did not deteriorate as a result of the aging process. Such samples can thus be considered as resistant to the process of aging. Composites made by extrusion exhibited slightly lower K-values compared to composites prepared using traditional rubber processing method. Their values remained at the level of 0.7-0.8.

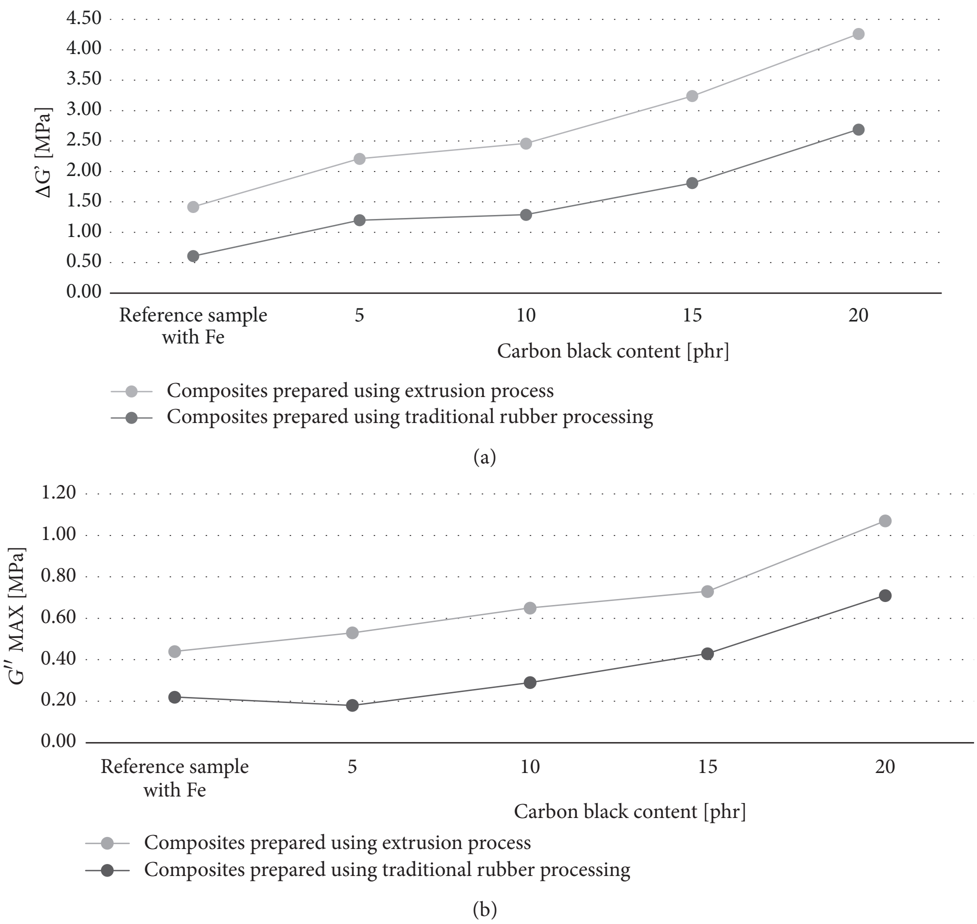

Figure 4: Influence of different processing methods on dynamic mechanical properties of magnetorheological composites: (a) $\Delta G'$: the increase the elastic modulus (Payne effect); (b) G''_{MAX}: the loss modulus determined under the max deformation.

3.4. Dynamic Mechanical Properties. The elastic module (G') of all examined vulcanizates decreased as the amplitude of the deformation increased, which is a confirmation of the Payne effect occurrence. This dependence was especially pronounced in the case of composites filled with carbon black (much stronger than for the reference sample, filled with CIP), which confirmed the fact that this effect is characteristic especially for reinforcing fillers. The results of dynamic mechanical properties of composites are given in Figure 4.

The higher the carbon black content of the examined vulcanizate, the stronger the Payne's effect. As the filler amount increased, the difference of the storage modulus value ($\Delta G'$) increased with the function of deformation. The higher the content of reinforcing filler in the elastomeric medium is, the more extensive spatial network was created by the particles in matrix. Thus, the effect of the filler-filler interactions that were damaged during the deformation of the sample was greater (influencing the Payne effect). Moreover, it can be stated that also G''max values depend strongly on the amount of carbon black contents in the vulcanizates. As the FEF N550 increased, the value of the maximum loss module also increased which is connected with high ability to dissipate energy during deformation.

Furthermore, the values of $\Delta G'$ for extruded composites were smaller than those made in a conventional manner. The obtained results confirm that additional stresses influencing the rubber compound during extrusion can cause disintegration of filler aggregates and orient the polymer chains in the direction of extrusion. Simultaneously improving the dispersion of the filler resulted in a reduction of the dynamic module.

3.5. Damping Properties under the Influence of Compression Stress. The damping properties of the studied magnetic composites are closely related to their ability to dissipate energy during deformation. Therefore, the relative attenuation values obtained were higher, so the composite dissipated the energy more strongly and therefore showed better performance. The influence of different processing method and fillers content on damping properties of the vulcanizates are given in Table 4.

The magnetoactive filler additive adversely affected the damping properties of EOR rubber composites. On the other hand, the introduction of an active filler resulted in a slight increase in the $T_{\tau w}$ value compared to the composite containing only the CIP additive and the reference sample. The amount of carbon black also had a negligible effect on

Table 4: Relative damping values of EOR vulcanizates obtained by various processing methods.

Sample	Carbonyl iron powder content [phr]	Carbon black content [phr]	Tτw [%]
Composites prepared using traditional rubber processing			
Reference sample	60	-	24.8±0.3
Carbon black filled sample		5	20.7±0.1
		10	21.2±0.2
		15	21.9±0.1
		20	21.6±0.1
Composites prepared using extrusion process			
Reference sample	60	-	24.8±0.5
Carbon black filled sample		5	26.2±0.2
		10	26.8±0.3
		15	26.2±0.1
		20	27.2±0.4

the attenuation coefficient. However, the improvement of the damping properties under compressive stress conditions of reinforced composites positively influenced the extrusion process. In each case, an improvement in the damping properties was observed compared to the conventionally produced composites. Once the rut is confirmed, it is possible to use this processing method to improve the degree of dispersion of the filler particles in the polymer matrix and thus improve the utility properties of the MRE composites.

3.6. Particles Fillers Dispersion Analysis of Composites. The aim of the extrusion process at the rubber compounding stage was to improve the degree of the dispersion of the filler particles and the cross-linker in the elastomer and to create oriented polymer chain structures. The degree of dispersion of the filler particles has a significant effect on its reinforcing effect and thus the mechanical properties of the vulcanizates. Morphology of the fillers in MRE composites was assessed on the basis of SEM images (Figure 5) and images from DisperTester (Figure 6) of the selected vulcanizates.

The images of vulcanizates prepared with the conventional method show that the filler particles characterize a high tendency to agglomerate. There are visible agglomerates of the size of several μm. Higher filler content contributed to this effect. This phenomenon leads to stress concentration and less mechanical strength. In addition, the increased tendency to agglomerate limited the activity of the cross-linking agent in the vulcanization process.

The confirmation of the positive impact of the extrusion process on the degree of dispersion of the filler particles is given in Figures 5 and 6. Particles agglomerates were still present, but they were smaller in size than in the vulcanizates produced conventionally. In both smaller and larger filler contents, more uniform filler network and a higher degree of dispersion were observed. As a result, vulcanizates with higher tensile strength and cross-linking density were obtained.

3.7. Magnetic Properties of Ethylene-Octene Composites. In order to confirm the magnetic properties of the composites, the vulcanizates were tested on vibration sample magnetometer. The results are given in Figure 7.

High magnetic susceptibility of magnetic filler (carbonyl iron powder, 200 Am^2/kg) caused all vulcanizates to exhibit magnetic properties. All obtained composites were characterized by magnetization (M_s) of 70 to 75 Am/kg. This is undeniably due to the strong magnetic character of ferromagnetic particles. Remanence for all prepared composites filled with magnetic powder varied from 1.0 till 1.1 emu/g. Coercivity values obtained for the MREs fluctuated from 0.6 to 2 kA/m indicating magnetically soft materials.

As expected, with the addition of reinforcing filler and its increased amount in the composites, magnetic properties of the samples were nearly at the same reference level. The highest values of M_s were designated for ethylene-octene/carbonyl iron without carbon black. With the increase content of the reinforcing filler addition, the value of saturation magnetization slightly decreased; however, these changes were not relevant from the material point of view and do not limit the potential usage of these materials.

4. Conclusions

Although designing materials based on magnetic fillers have already been applied successfully in various fields, more work still needs to be done to make them more multifunctional. The mechanical characteristic of MRE composites is one of the most important factors with perspectives to offer wide industrial applications. Therefore, the aim of this research was to fabricate selected isotropic magnetic elastomer composites and determine their mechanical properties.

(1) Carbon black (FEF N550) was selected as an additive to modify and improve iMRE mechanical properties, as it is an important reinforcing filler in polymer engineering, especially in rubber technology. From the results obtained in the research, it seems that extrusion processing can be used as a method to improve mechanical and damping properties of new, nonpolar, ethylene-octane rubber composites. This method allows creating oriented polymer chains and improving dispersion of filler particles.

(2) Based on the data, it follows that tensile strength of composites prepared by extrusion process increased more

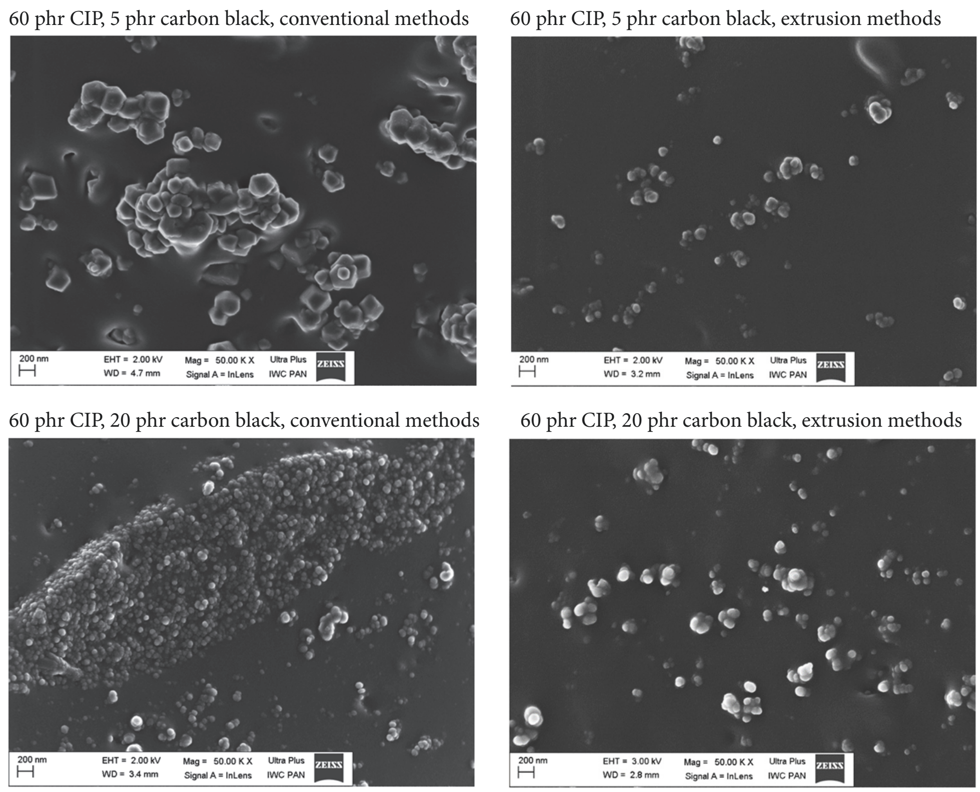

Figure 5: Morphological characteristics SEM image of composite filled with 60 phr CIP and 5 phr (up) and 20 phr (down) carbon black.

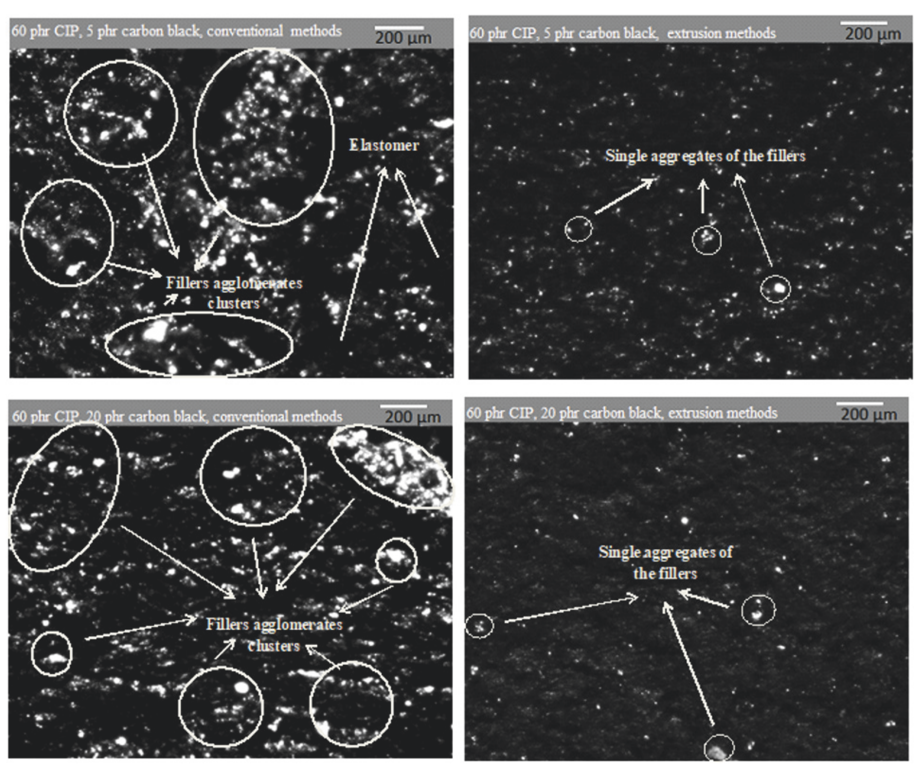

Figure 6: Morphological characteristics DisperTester images of composite filled with 60 phr CIP and 5 phr (up) and 20 phr (down) carbon black.

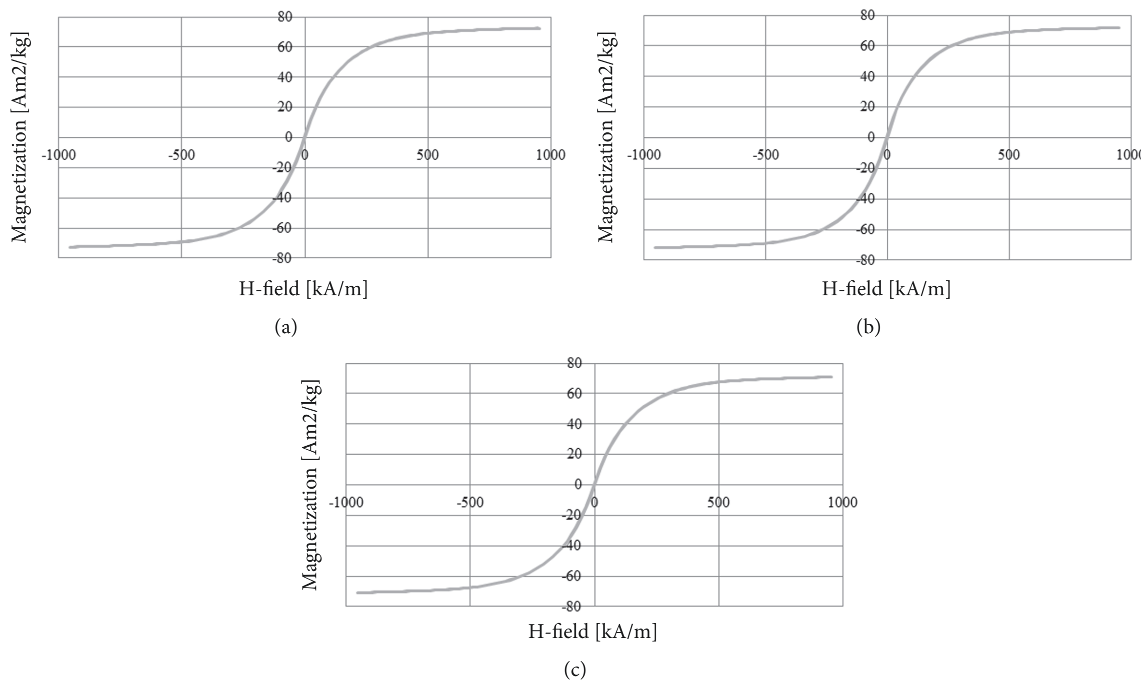

Figure 7: Magnetic properties–hysteresis loops of MRE samples filled with 60 phr carbonyl iron powder and 5 phr (a), 10 phr (b), 15 phr, and (c) carbon black cured under magnetic field (H) of 1,2T applied perpendicular (L) to the sample.

than twice compared to the vulcanizates obtained by commonly used methods (mixing and rolling).

(3) The extrusion process positively influenced the relative damping value of vulcanizates compared to conventionally produced composites. New application of extrusion process for reinforced i-MRE composites preparation was confirmed.

(4) Dynamic-mechanical analysis indicated the presence of strongly developed secondary structure of the fillers in MRE composites.

(5) The differences between rheometric properties and kinetic characteristic of ethylene-octene copolymer mixtures obtained with different processing methods were insignificant.

(6) Thermooxidative aging process did not influence mechanical properties of the filled composites. The resistance of vulcanizates to degradation processes was satisfactory, which can have a positive effect on the case of industrial application of manufactured material.

Data Availability

The data used to support the findings of this study are included within the article.

Ethical Approval

This work complies with ethical standards.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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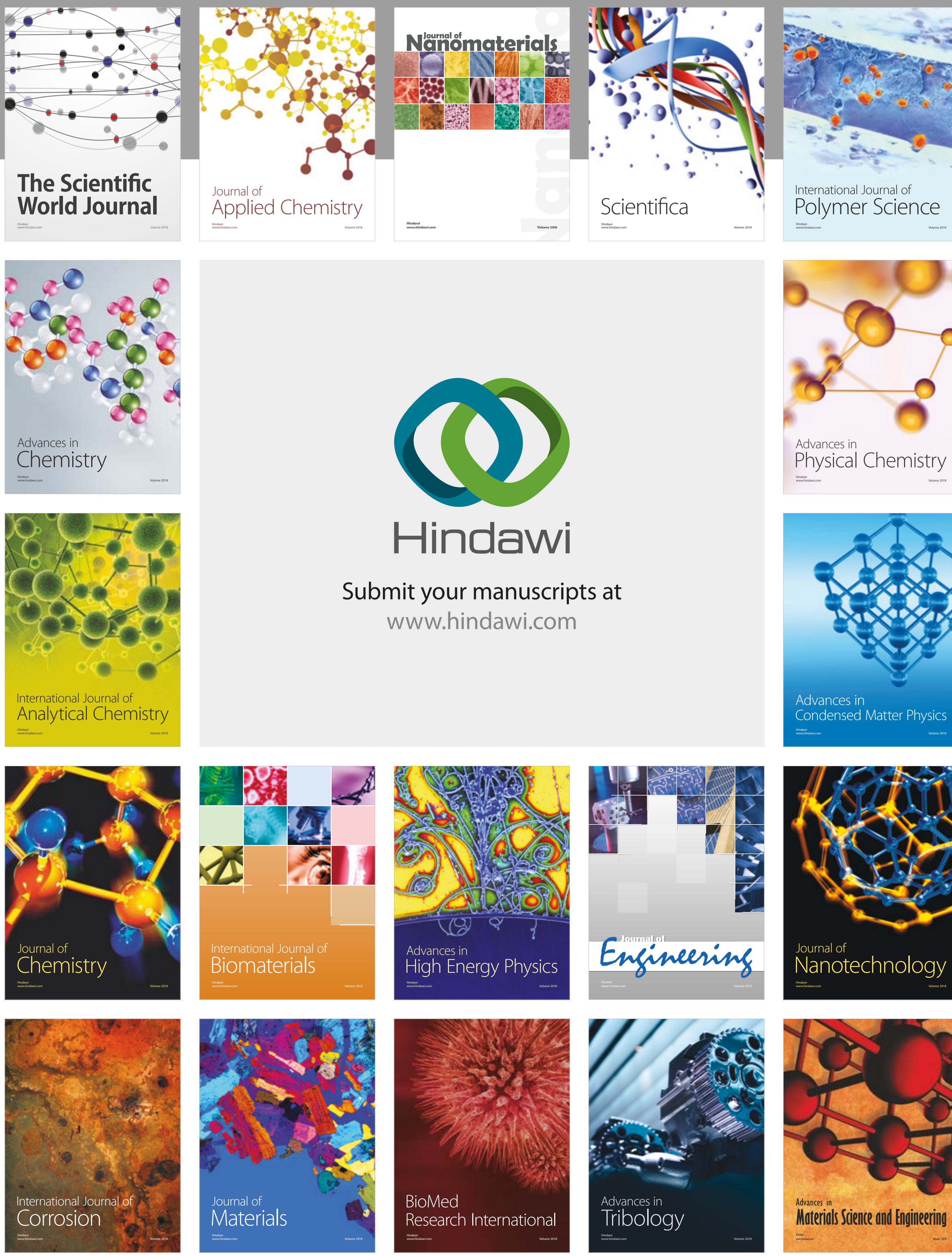
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