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High-density Polyethylene - Expanded Perlite Composites: Structural Oriented Analysis of Mechanical and Thermomechanical Properties

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Abstract: *As part of this work, research was carried out on the effect of the addition of expanded perlite (PR) on the mechanical and thermomechanical properties of high-density polyethylene (PE) composites. Composites containing from 1 to 10 wt% of the inorganic filler were produced. Polyethylene-based composites manufactured by twin-screw extrusion and formed in the compression molding process were subjected to mechanical, thermomechanical, and structural analyses. The structure of polymer composites and filler was analyzed using scanning electron microscopy (SEM). It has been correlated with the static tensile tests and results of dynamic thermomechanical analysis (DMA). As part of the work, several thermomechanical parameters were calculated, and the obtained results were discussed with the evaluation of interfacial adhesion based on microscopic analysis. The research indicate that despite introducing a 10 wt% of particle-shaped filler, the composites show increased stiffness without noticeable deterioration in tensile strength, simultaneously reducing toughness and brittleness. The analysis of the thermomechanical properties showed the lack of significant effects of the filler influence on the polymer matrix.*

Keywords*: polyethylene, perlite, composites, mechanical properties, dynamic mechanical analysis*

1. Introduction

For many years, polymer composites have been one of the most dynamically developing fields in material engineering. The search for new solutions aimed at adjusting the polymers' properties so far to new applications and limiting the amount of consumed petrochemical materials results in the presence of new solutions and the use of new materials as fillers. The enhancement of particular matrix properties is strongly dependent on the shape and aspect ratio of applied fillers. In most cases, fibrous fillers increase the mechanical strength of the final products concerning the polymer matrix [1,2]. On the other hand, introducing powder and particle-shaped fillers into the polymer may be aimed at modifying thermal, thermomechanical, tribological, and electrical properties or be caused by the necessity to limit the price of the composite material [2-5]. In the case of powder and particle-shaped fillers, also their chemical structure is often essential for the performance of composites since they do not offer possibilities for such an exceptional stress transfer compared to fibers.

Perlite is a glassy mineral of volcanic origin that is classified into three grades with different water content, i.e., obsidian (below 2 wt%), perlite (2-5 wt%), and pitchstone (above 5 wt%). It contains SiO_2 (65-75%), Al_2O_3 (10-18%), $K_2O + N_2O$ (6-9%), MgO + CaO (2-6%) and Fe_2O_3 (1-5%). The sources of perlite can be found in the Mediterranean countries such as Turkey, Greece as well as Armenia, Hungary, China and the USA. This material was formed by solidification in an aqueous environment. As a result of annealing at the temperature of 900-1000°C, it is possible to obtain it in an expanded form due to evaporation of the bound residual water [6,7]. Expanded perlite, due to its extensive specific surface area and low density, is most often used as an insulation material, as well as an additive for the production of

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elements used in construction, such as gypsum boards or lightweight concrete, in agriculture and horticulture as a soil amendment as well as a filler for polymer composites with reduced density [8-10]. The combination of mechanical and insulation performance of perlite with the low density may enable significant costs reduction in construction sector due to the reduced weight of particular building components.

In so far published studies, one can find works related to perlite as a filler for polymer composites, including those based on polyolefins. This direction of studies is understandable, as polypropylene and different grades of polyethylene comprise about 49.2 of total plastic demand in European Union [11]. Polyolefines are lightweight, easy to process, and can be obtained at a relatively low cost, so they are likely to remain the plastics industry leaders. However, they can also be characterized by relatively low mechanical and thermomechanical properties, which can be improved by adding an inorganic particle-like filler such as perlite. The high-density polyethylene (HDPE) nanocomposites reinforced with nanometric-sized perlite particles were investigated in the course of their mechanical and thermal properties by Lapcik et al. [12]. As shown by the results of static tensile measurements and non-destructive vibrator testing, the addition of inorganic nanosized filler caused a 37% improvement in the elasticity modulus compared to the unmodified polymer. However, despite described strong bonding between polymer and filler particles, nano-perlite acts as a stress concentrator providing the embrittlement of the composites. Mattausch and co-workers [13] investigated the effect of the configuration of mixing segments of a co-rotating twin-screw extruder on the properties of poly-propylene (PP) composites filled with expanded perlite. The research highlights the possibility of damaging the honeycomb-like structure of expanded perlite due to melt processing. At the same time, despite the general deterioration of the strength properties of composites due to the addition of perlite, the mechanical tests carried out showed obtaining more favorable strength effects for the series produced with the use of higher shear forces. This effect was due to the more homogenous dispersion of lower particle-sized filler and the PP matrix. Szadkowski et al. [14] studied the effect of perlite compared to vermiculite on the properties of ethylene-propylene (EPM) composites crosslinked with dicumyl peroxide with crosslinking co-agents. Apart from basic mechanical and barrier tests, the analysis focused on assessing thermal properties and flammability. The research showed that the addition of perlite in 20 wt% leads to a significant reduction of the heat release rate (HRR) and total heat release (THR), 67 and 75%, respectively, compared to pure EPM. Interesting results of perlite addition in the production of polypropylene composites were also described in [15]. PP composites with the addition of 15, 30, and 50 wt% perlite were subjected to gamma irradiation, with a radiation range of 10-100 kGy. It has been shown that the composite samples containing the inorganic filler after irradiation with higher radiation doses (above 50 kGy) were less susceptible to the deterioration of mechanical performance compared to unmodified PP. In conclusion, many attempts to expand perlite introductions as a filler into polyolefin matrix have been undertaken so far; however, mechanical and thermal properties changes induced by the inorganic fossil filler are often unpredictable. This is mainly due to changes in the filler structure caused by both its natural origins and changes resulting from pre-processing and processing of the composites.

In the works described so far, despite the introduction of expanded perlite with various particle sizes and manufacturing methods, there were no comprehensive studies relating to parameterizing changes in thermomechanical properties using dedicated parameters determined based on data from dynamic thermomechanical tests (DMA). This work aimed at a synthetic approach to describe the complex changes in the mechanical properties and thermomechanical of polyethylene-based composites caused by introducing an inorganic powder filler, mainly expanded perlite (PR). As part of the work carried out, thermomechanical parameters such as the efficiency of the filler, adhesion factor, and constrained chain volume as well as structural analysis and mechanical properties evaluation, were determined and discussed.

2. Materials and methods

2.1. Materials and preparation of polymer composites

High-density polyethylene (HDPE), type M300054, delivered by SABIC (Bergen op Zoom, The Netherlands), characterized by the mass flow index (MFI) of 30 g/10 min (190°C, 2.16 kg) and density of 0.954 g/cm^3, was applied as a matrix to prepare investigated composites. Expanded perlite (PR) was preliminary grinded with using Retsch GM200 knife mixer. Obtained filler was characterized by a bulk density of 1.4419 g/cm^3, and its average particle size was 59 μm (value estimated based on 400 measurements of particle radius by scanning electron microscopy (SEM). The example images of the filler made using SEM with various magnifications, and presenting the structure of expanded perlite, are presented in Figure 1.

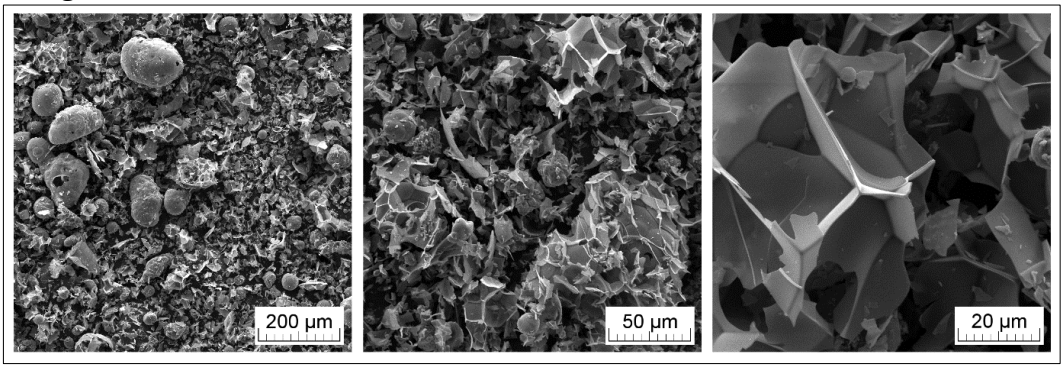

Figure 1. SEM image of perlite used in a study

The composites were prepared by mixing in a molten state. The HDPE pellets were pulverized into a fine powder using a Tria 25-16/TC-SL high-speed knife grinder (Tria S.p.A., Cologno Monzese, Italy). Then, the polymeric powder was preliminary mixed with 1, 2, 5, and 10 wt% of perlite powder using the Retsch GM200 knife mixer (Retsch GmbH, Haan, Germany operating at 3000 rpm for 3 min). The physical mixtures were processed using a ZAMAK EH16.2D co-rotating twin-screw extruder (Zamak Mercator Sp. z o. o., Skawina, Poland) operating at 100 rpm with the maximum temperature of the process of 190°C. The obtained materials were cooled in forced airflow and pelletized. The resulting composites were then compression molded using Fontjine LabManual 300 hydraulic press at 170°C and 10 MPa for 2 min, then kept under pressure at room temperature for another 5 min, and freely cooled until achieving solidification fully. All manufactured samples were conditioned for seven days in temperature of 23°C and average humidity of 50% before mechanical testing. Unfilled HDPE was subjected to melt processing procedure under conditions identical to composites to exclude the influence of thermal history on the structure and properties of the reference samples. The assignation of the samples was related to the filler content in the composite, e.g., material series were described as PE/XPR, where X stands for the filler content. In Figure 2 the scheme of composite series preparation is presented.

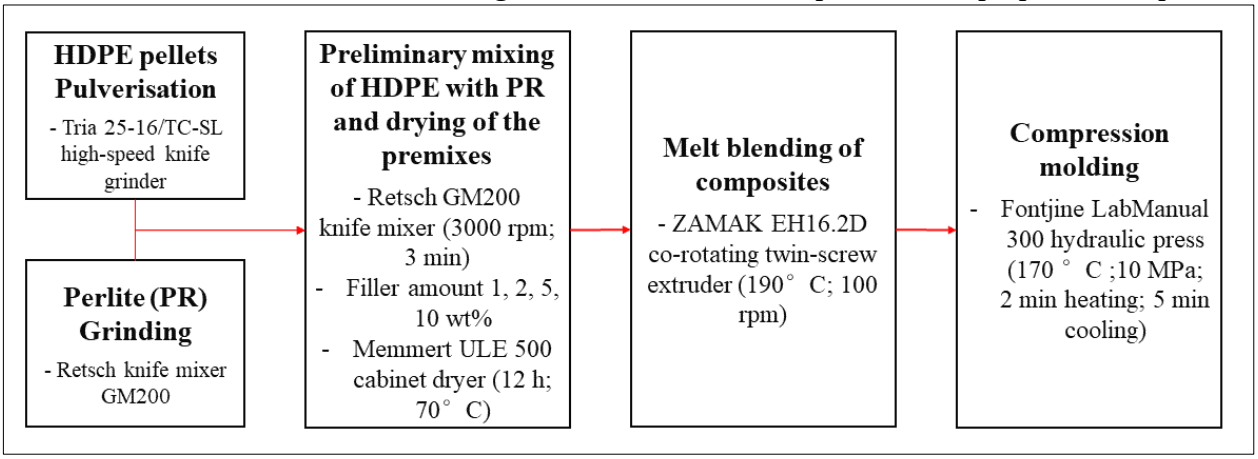

Figure 2. Composite samples preparation scheme

2.2. Methods

The scanning electron microscope (SEM), model Tescan MIRA3 (Brno, Czech Republic), was used to evaluate the structure of composites. The structures of the carbon-sputtered cryofractured surfaces of the polyethylene and composites samples were assessed with an accelerating voltage of 12 kV.

The density of filler and composites was determined using Ultrapyc 5000 Foam gas pycnometer from Anton Paar (Graz, Austria). Following measurement settings were applied: gas - helium; target pressure - 18.0 psi; flow direction - sample first; temperature control - on; target temperature - 20.0°C; flow mode - monolith; cell size - small, 10 cm^3; preparation mode - flow; time of flow - 0.5 min.

The tensile strength, elongation at break, and elastic modulus were estimated following PN-EN ISO 527 standard, using the Zwick/Roell Z020 apparatus (Zwick Roell Group, Ulm, Germany) with a cell load capacity of 20 kN. Samples type 1BA were used. Tensile tests were performed at a constant speed of 1 mm/min (for elastic modulus) and 20 mm/min (tensile strength and elongation at break). Five samples were analyzed for each specimen.

Shore hardness type D was estimated using Zwick 3131 durometer (Zwick Roell Group, Ulm, Germany) following PN-ISO 868. The presented hardness value was the mean of at least 11 measurements for each series.

The dynamic mechanical analysis was conducted on a DMA Q800 TA Instruments apparatus (Waters Corporation, New Castle, Delaware, USA). Samples with dimensions of 40 x 10 x 2 mm were loaded with variable sinusoidal deformation forces in the single cantilever bending mode at the frequency of 1 Hz under the temperature rising rate of 4°C/min, ranging the temperature from -100 to 100°C.

3. Results and discussions

3.1. Structure analysis

Figure 3 shows the brittle fractures of samples formed by compression molding from high-density polyethylene and composites containing different concentrations of perlite. Brittle fractures were obtained by breaking the samples frozen in the liquid nitrogen. Freezing down the samples significantly below their glass transition temperature enabled obtaining brittle fractures. Additionally, in the case of a composite containing 10 wt% of PR, an additional image was taken at higher magnification to assess the polymer-filler interphase and analyze the adhesion. Considering the particle size of the filler shown in Figure 1, it can be seen that shearing effects did not significantly mechanically degrade it during melt mixing; however, there have been no spherical-shaped closed filler domains. Similar to the observations of Mattaush et al. [13], perlite particles saturated by polyethylene with an exposed honeycomb structure are visible at the composite samples' fracture. While some of the filler particles shown in Figure 1 exhibit intact spherical structure, melt processing using intensive shearing conditions resulted in a small reduction of the expanded perlite particle size. Individual hollow domains of characteristic filler structures can be observed (PE/5PR and PE/10PR samples). The dispersion of the filler in all composites can be assessed as acceptable, which confirms that mixing in a molten state of the composites was performed correctly. The distribution of the perlite particles is homogeneous; it should additionally be emphasized that there were no pull-out holes that could arise in the case of improper adhesion of the filler to the polymer. In the SEM image made with higher magnification for PE/10PR sample, no void in the interphase space was noted, which points to the low porosity of composites, beneficial for their mechanical performance. Moreover, it can be noticed that the filler particle is broken in the plane of the brittle fracture. This proves the good adhesion of the polymer to the filler. Despite the different nature of PE and PR (hydrophobic polymer and hydrophilic filler), good adhesion will usually result from an extensive specific surface and mechanical interlocking of the particles in polymeric matrix super-saturating them. Therefore, it can be decided that perlite does not need any chemical modification or coupling agents to show good affinity to the polymeric matrix, which is highly advantageous. Such an approach is in line with the Green Chemistry Principles [16].

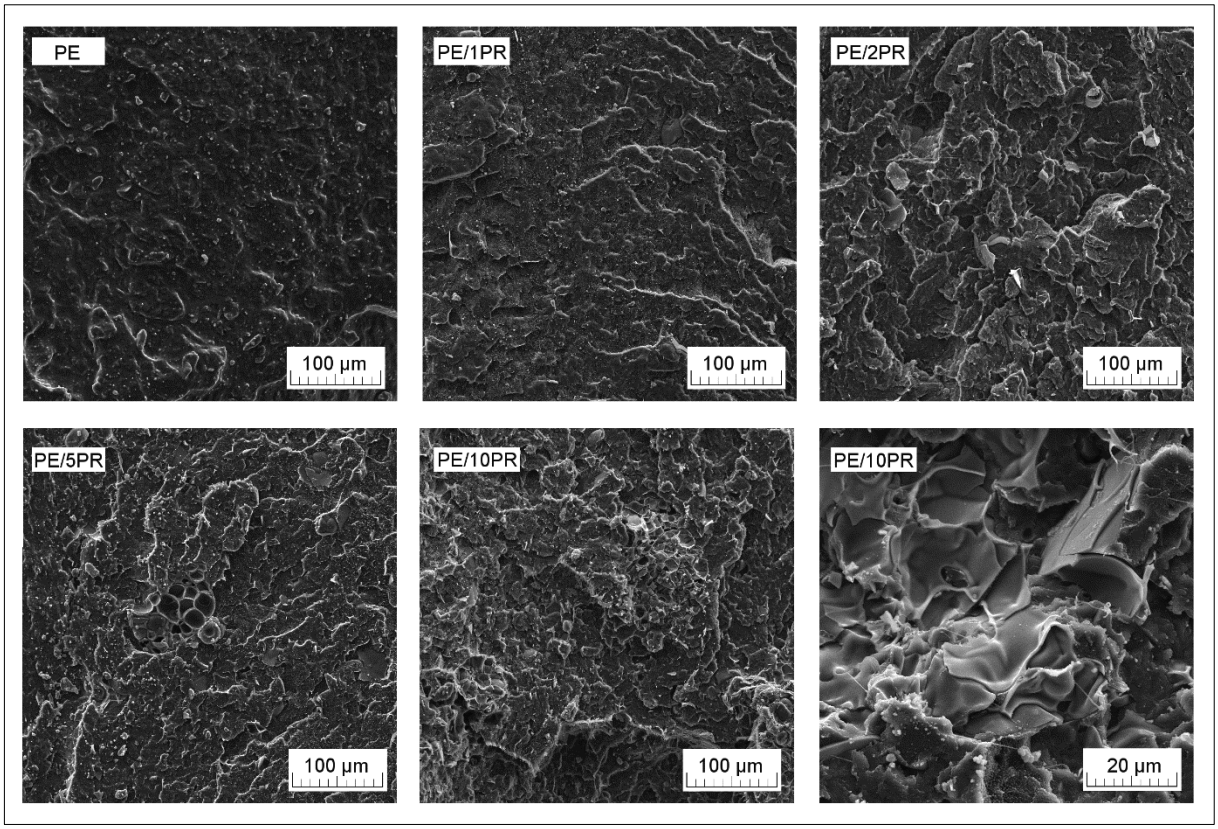

Figure 3. SEM images of PE and PE/PR composites

The results of performed analyses related to the physical properties of polyethylene/perlite composites, including density-based calculation of the porosity, are presented in Table 1. We reported the theoretical and experimental density values, which enabled the determination of materials' porosity. As demonstrated in the previous work, theoretical composites' density and porosity values were calculated using the rule of mixtures using neat matrix and filler density values [17]. Values of density were applied to determine the volume fractions of matrix and filler, which enabled calculation of theoretical composites' density. Porosity is a crucial factor for the mechanical performance of composite materials since it quantitatively determines the number of voids and discontinuities in the material. In the case of composite materials, voids are often present at the interface between matrix and filler, as well as in the fillers' agglomerates, particularly observed for powder and particle-shaped fillers. Most of the work related to the adverse effects of porosity has focused on analyzing the properties of thermoset polymer-based laminates [18]. Of course, the presence of pores may positively affect the damping of mechanical vibrations. Still, it may lead mainly to weakening the composite structure by creating non-load-bearing discontinuities in its structure. Saenz-Castillo et al., in their work on poly(ether ether ketone) (PEEK) reinforced with carbon fibers (CF) [19], showed that the critical pore content was about 1.2-2.7%. Considering the obtained test results, where only in the case of PE/10PR the porosity value above 2% was achieved, it can be concluded that the porosity in the selected case should not be a significant problem and should not have a substantial impact on the change of mechanical properties. It should also be considered that the obtained results of porosity may result not so much from entrapped air domains in polymeric structure, which were not observed by SEM analysis, but from the porous structure of the inorganic filler itself, the volume content of which was 8.8% in the case of the highest concentration. Bearing in mind the data compiled in [20], it is assumed that composites with a pore content below 1% are described as excellent quality, materials in the range of 1-5% of porosity as good quality, and only products with a porosity content of more than 5% are classified as for defective materials with poor quality. Summarizing, it can be stated that polyethylene/perlite composites produced

using extrusion and compression molding are of good quality, which was confirmed by physicochemical analysis and structural evaluation.

Table 1. Physical properties of PE and PE-PR composites

Parameter	PE	PE/1PR	PE/2PR	PE/5PR	PE/10PR
Theoretical density, g/cm^3	0.9517	0.9549	0.9582	0.9683	0.9855
Experimental density, g/cm^3	0.9517	0.9520	0.9524	0.9545	0.9639
Porosity, %	-	0.307	0.608	1.422	2.193
Filler volume fraction, %	-	0.66	1.32	3.34	6.80

3.2. Mechanical and thermomechanical properties

The mechanical properties in Table 2 indicate that the addition of the expanded perlite did not cause the deterioration of the tensile strength, despite the introduction of 10 wt% of the filler. The differences between tensile strength of PE/10PR sample and neat PE are within the range of standard deviation. This effect is very beneficial considering the slight increase in the density of the composite itself. In most cases reported in the literature, using a powder filler to produce composites based on nonpolar thermoplastic polymers is associated with a significant deterioration in mechanical performance. This is mainly due to improper adhesion at the polymer-filler interface. Such an effect efficiently limits wider application of such materials, which often do not have industrial application or are generated as by-products in various processes and their incorporation into polymer matrices could provide an excellent method for their reuse and recycling. In the studied case, this tendency does not occur, indicating good compatibility of the composite components, which was also indicated by the SEM observations. Moreover, the effect of a gradual increase in the stiffness of the composite with the increasing share of the filler in the composite was also noted. In the case of the samples containing the highest concentration of PR, an increase in Young's modulus by 200 MPa was noted compared to unmodified polyethylene. The increase in composite stiffness is observed in most thermoplastic polymers' modifications using particle-shaped fillers [21, 22]. The effect is caused by dispersion of the rigid structures, which limit the mobility of the polymeric chains during the strain, and is especially pronounced in the case of the good polymer-filler adhesion [23, 24]. The addition of inorganic filler caused a significant decrease in elongation at break in composites compared to unmodified HDPE. While in the case of the lowest filler content, there was a decrease in this value by one order of magnitude, in the case of 10 wt%, there was almost a decrease by two orders of magnitude. As a result, the composite materials produced with the perlite should be characterized as stiff and brittle materials.

Table 2 presents also the values of tensile toughness - τ, determined as the amount of energy needed to break a sample in static tensile conditions, which can be calculated as the area under the stress-strain curve, using formula (1) [25]:

$$\tau = \int_0^{\varepsilon_b} \sigma \, d\varepsilon \tag{1}$$

where: ε_b - is elongation at the break of the material, %.

Due to the complex problem of defining the brittleness of polymeric materials, Brostow et al. [26] proposed a method for determining this material parameter based on multi-criteria analysis and results obtained with various measurement techniques in both quasi-static and dynamic conditions (DMA and static tensile test). As a result of their considerations, the equation (2) was defined:

$$B = 1 / (E' \cdot \varepsilon_b) \tag{2}$$

where: B - brittleness, 10^{10} %·Pa; ε_b – elongation at break, %; E' – storage modulus at 25°C, Pa.

Considering the parameters mentioned above described by equations (1) and (2), brittleness can be considered the toughness antagonist. Material characterized by low brittleness should be able to withstand high forces over the possibly most comprehensive range of deformations. Nevertheless, these parameters are not inversely proportional. Although the elongation at break is considered in both cases, toughness considers the tensile strength, while brittleness is the storage modulus determined by DMA analysis. As presented in Table 2, results the decrease of the elongation at break mean values depression may be indirectly correlated with unfavorable changes in the toughness and brittleness of the composites. Summing up, while the addition of the filler did not cause significant changes in the strength of composites and even resulted in an improvement in their stiffness, the introduction of perlite had a negative effect on the ductility of the PE/PR systems. Such an effect is typical for polymer composites containing micrometric powder and particle-shaped fillers.

Table 2. Mechanical and thermomechanical parameters of PE and PE/PR composites

Parameter	PE	PE/1PR	PE/2PR	PE/5PR	PE/10PR
Tensile strength, MPa	23.0±1.1	23.3±0.8	22.8±0.4	22.4±0.5	22.1±0.1
Elongation at break, %	407±54	66.4±11.1	10.3±1.1	8.2±0.2	5.9±1.0
Young's modulus, MPa	947±43	969±30	980±32	1081±72	1181±64
Toughness, J/cm^3	5726±658	946±98	165±38	131±15	105±14
Brittleness, 10^{10} %·Pa	0.0141	0.0961	0.5822	0.7258	0.8756
E' at -90 °C, MPa	3157.9	2894.1	3073.6	3062.3	3467.7
E' at 25 °C, MPa	1735.1	1567.6	1666.7	1676.9	1924.9
tan δ at 25 °C	0.0597	0.0569	0.0582	0.0585	0.0571
tan δ at T_g	0.0545	0.0536	0.0530	0.0507	0.0477
T_g, °C	-51.47	-51.34	-51.14	-50.60	-50.38
C factor	1.000	1.014	1.013	1.003	0.989
Constrained chain volume, %	0.00	1.34	2.30	6.01	10.79
Adhesion factor at 25 °C	-	-0.0410	-0.0070	0.0133	0.0270

In addition to the results of mechanical tests, Table 2 also lists the thermomechanical properties of PE and PE/PR composites obtained on the basis of dynamic thermomechanical analysis (DMA). The storage modulus (E') and damping factor (tanδ) vs. temperature curves are presented in Figure 4. In addition to the basic parameters such as storage modulus (E') in the ambient temperature (25°C) and the temperature of -90°C, i.e., in the glassy and rubbery state, the thermomechanical parameters that indirectly describe the interactions in the considered materials are presented. Typically, E' values correlate with Young's modulus values measured in the static tensile test. In the case under consideration [27], however, only for the composites with the highest filler concentration, an increase in E' was recorded compared to the sample made of unmodified PE, both at room and low temperature. The remaining samples showed slightly lower values of E 'composites to PE. Such an effect was noted both in glassy and rubbery state.

Except for the data related to the stiffness of composites, DMA provides information on the material's damping properties. They are related to the damping factor (tanδ) of material, which characterizes the ability of a material to dissipate the mechanical energy. The temperature position of tanδ peak is also often used for determination of the glass transition temperature (T_g) of the polymers. The local maximum of tanδ is connected with changes in mobility of macromolecular chains. In dependence on the chemical structure of the polymer, relaxations may be related to changes in mobility of both the crystalline phase and amorphous phase of the thermoplastic polymer. Changes in T_g caused by the increasing share of the filler are gradual, leading to a decrease in this parameter. Still, the maximum change, i.e., an increase by 1.09°C, should be considered negligible. In the case under consideration, the described changes in T_g occur in the range of β-relaxation of the polyethylene. It is supposed that β-relaxation is related to the

motion of branches, which in the case of high-density polyethylene do not constitute a significant share. Although the assumptions of determining T_g for polyethylene are part of a broader discussion, as described by Khanna and co-workers [28], in the case of HDPE, the choice of β-relaxation seems to be justified due to the lack of significant changes in the intensity of the recorded local maximum values on the thermomechanical curves as a result of content changes branches in polyethylene structure. Considering values of tanδ at peak, all composite samples reveal lower values, which may be interpreted as lower mechanical damping ability.

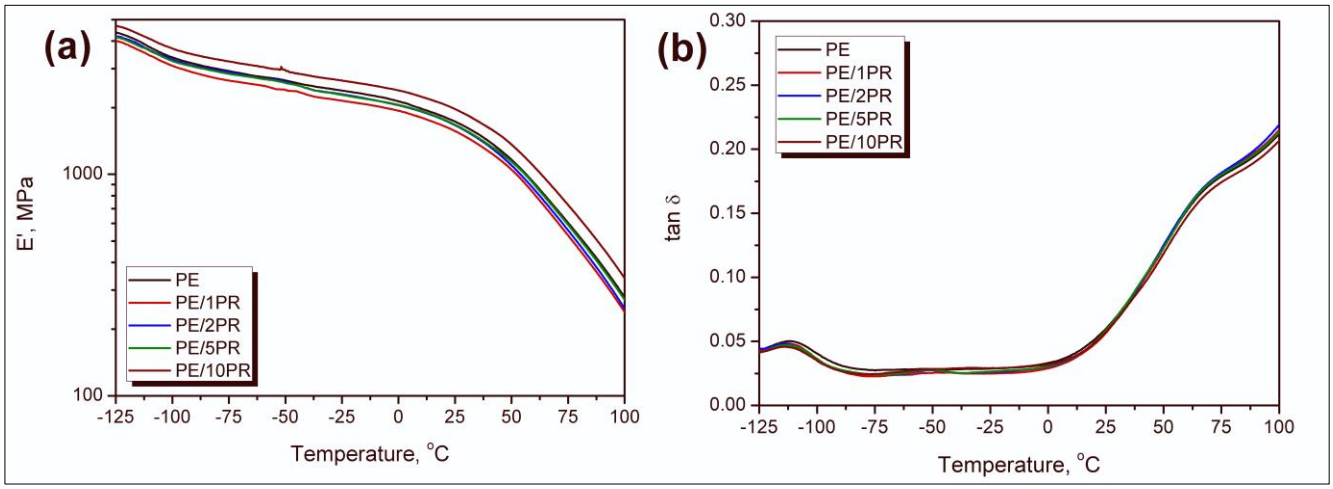

Figure 4. Storage modulus (E') and damping factor (tanδ) vs. temperature curves obtained by DMA

To evaluate the efficiency of fillers towards modification of the modulus of composites in different conditions, the C factor was calculated according to the following equation (3) introduced by Chua [29]:

$$C = ((E'_{gc} / E'_{rc}) / (E'_{gm} / E'_{rm})) \quad (3)$$

where: E'_g - storage modulus in the glassy state (-90°C), MPa; E'_r - storage modulus in the rubbery state (25°C), MPa. Presented equation provides relatively simple and straightforward insights into the efficiency of introduced fillers and could be used to compare various materials or their modifications. The lower values of C factor point to the higher efficiency of filler on the modulus of the composite. Only in the case of the PE/10PR sample the positive effect of the filler can be observed in the form of the calculated C factor value concerning the unmodified PE. This result seems reasonable, taking into account the fact, that only in the case of the composite containing the highest filler content the obtained E' values were higher in comparison with the ones of the HDPE sample.

Because of the fact that brittleness and toughness are "antagonistic" properties, the relationship between brittleness and toughness of various polymers has been described in literature [30], and allow to formulate the following equation (4):

$$\tau = (b + c \cdot B) / (1 + a \cdot B) \quad (4)$$

where: τ - toughness, J/cm^3; a, b, c - constants.

In our previous work [31], we presented this relationship using power function in the form of following equation (5):

$$\tau = d \cdot B^e \quad (5)$$

where: d, e - constants. Such presentation enables easier interpretation of obtained results. The value of d constant determines the base value of composites toughness, while e constant plays a similar level for

brittleness. For data presented by Brostow et al. [30], values of d and e are 178.380, and -0.984, respectively. In the presented work, values of 94.657 and -1.0220 were obtained, so experimental data points lie below the curve proposed by Brostow et al. [30]. They developed a toughness-brittleness relationship mostly for homopolymers. Therefore, they did not take into account the impact of interfacial adhesion, which is often inferior to the cohesion of polymer matrix. Obtained values of the above mentioned constants point to the insufficient compatibility of perlite with polyethylene matrix because no reinforcing effect was noted.

Apart from the glass transition temperature, tanδ peak can be analyzed in terms of the molecular motions occurring inside the composite. Its magnitude is related to the composites' ability to dissipate the energy through molecular motions. The reduction of polymer chains' mobility is expressed by the decrease of tanδ peak value [32]. It can be seen that the introduction of perlite particles caused the drop of tanδ peak magnitude, proportional to the volume fraction of filler. Such an effect is associated with the incorporation of solid particles into the polymer and has been confirmed numerously by other researchers [33-35], and is resulting from suppressed damping ability of the composites [36]. Solid filler particles act as nodes of the network and provide the effect qualitatively similar to the crosslinking of structure in terms of the damping ability. Reduced mobility of the polymer macromolecules inside the composite may suggest an increase in the number of polymer chains constrained by filler particles [37]. Their volume can be determined based upon the information presented by various researchers [32,38], using the following equation (6):

$$C_v = 1 - (((1 - C_0) \cdot W) / W_0) \quad (6)$$

where: C_v - volume fraction of the immobilized polymer chains, %; C_o - volume fraction of the immobilized chains in pure polyethylene (taken to be 0), %; W and W_o - energy loss fractions for the analyzed sample and pure PE, respectively. Energy loss fraction W can be calculated from the tanδ in accordance with the following equation (7):

$$W = (\pi \cdot \tan \delta) / ((\pi \cdot \tan \delta) + 1) \quad (7)$$

For a more detailed analysis of the interfacial adhesion in presented composites and its dependence on the type and content of applied fillers, in Table 2 there are also presented values of the adhesion factor. The concept was proposed by Kubát et al. [39]. It is based on the assumptions that the mechanical properties of composite materials are determined by the performance of matrix, filler, and interface. In our previous work [17], the adhesion factor was evaluated was proposed according to the following formula (8):

$$A = (1 / (1 - \varphi_f)) \cdot (\tan \delta_c / \tan \delta_m) - 1 \quad (8)$$

where: A - adhesion factor. Lower values of the adhesion factor indicate strong interfacial adhesion between matrix and filler. Negative values are resulting from the simplifications made during development of the above-mentioned equation by Kubát et al. [39]. Interestingly, the A parameter is rising with the increasing content of the filler. According to Abhimanyu et al. [40], the composites with the lowest content of the filler (1 wt%) reveal maximum interaction between the filler and polymeric matrix. The higher the filler content, the lower the restricted mobility of the polymer chains near the particle than at the bulk matrix.

4. Conclusions

As part of the work carried out, it was shown that it is possible to produce polyethylene composites modified with an expanded perlite in the form of micrometric powder filler, with favorable mechanical properties. The addition of 10 wt% of the inorganic filler increased Young's modulus by 24% relative to the reference sample but, at the same time, did not deteriorate the tensile strength, which is an uncommon

but highly advantageous result in the case of a hydrophilic filler characterized by a low aspect ratio. It was decided that this behavior results from sufficient adhesion between phases in the composite, which results from the high specific surface of the expanded filler and mechanical interlocking of the polymer and the filler. At the same time, the introduction of honeycomb-structured filler significantly reduced the ductility of the composites and increased their brittleness. The mechanical tests were supplemented with structural analysis and multi-criteria analysis of thermomechanical parameters to assess adhesion. The tests confirmed the initial observation that the adhesion of the filler was at an acceptable level, taking into account the lack of surface modification and the use of a nonpolar polymer matrix. Presented results provide auspicious insights into the potential application of expanded perlite as filler of thermoplastic polymer composites. Such an approach could yield novel utilization methods for this inorganic material in the future. Except for the increased use of perlite, it could reduce the amount of used polymer matrix, which is crucial considering the current environmental crisis.

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