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Analysis of the mechanical properties of electrotechnical composites based on recycling materials

***Abstract**. Modern industry is looking for new functional materials with the same or better functional properties while reducing the burden on the natural environment. Innovative materials such as composites, which are a combination of components with different and complementary properties and the appropriate technology of their production meets these expectations. The preliminary studies carried out indicate the possibility of using the method of producing a metal / polymer composite for the construction panels and casings covering the electromagnetic field.*

***Streszczenie.** Współczesny przemysł poszukuje nowych materiałów funkcjonalnych o takich samych lub lepszych właściwościach użytkowych przy jednoczesnym zmniejszeniu obciążenia środowiska naturalnego. Innowacyjne materiały, takie jak kompozyty, które są połączeniem komponentów o różnych i uzupełniających się właściwościach oraz odpowiednia technologia ich wytwarzania, spełniają te oczekiwania. Przeprowadzone badania wstępne wskazują na możliwość zastosowania opracowanej przez wytwarzania kompozytu metal/polimer na płyty konstrukcyjne i obudowy pokrywające pole elektromagnetyczne. (**Analiza właściwości mechanicznych kompozytów do zastosowań w elektrotechnice bazujących na materiałach recyklingowych**).*

Keywords: metal – polyethylene composite, iron scale, volume resistance, mechanical properties.
Słowa kluczowe: kompozyt metal – polietylen, zendra, rezystancja skrośna, właściwości mechaniczne.

Introduction

Modification of polymeric materials carried out in a chemical, physicochemical or physical way, e.g. by thermal treatment or the addition of various types of fillers (in the form of dust, powder, flakes etc.) is increasingly used in various industrial branches [1-5]. This fact results, on the one hand, from the desire to change the mechanical, electrical and visual properties of the material and on the other hand, from the possibility of reducing the price of the final product [6,7].

One of the most commonly used in the production of polymeric or composite materials is high-density polyethylene (PE-HD), which, next to polypropylene and other grades of polyethylene, belongs to the group of polyolefin materials. It is estimated that the demand for polyolefins is over 40% of the demand for all polymers in Europe [8]. Consequently, it constitutes a significant proportion of the polymer waste [9]. High-density polyethylene is a semi-crystalline material, characterized by, among others, high resistance to chemical factors, low weight, good abrasion resistance, good dielectric properties and low water absorption. Due to the above properties, it has been used as a raw material for the production of e.g. any kinds of packagings, foil, toys, pipes, as well as machine components [10,11].

In turn, iron scale can be one of the fillers of composite materials used in electrical engineering. It is a coating that is formed on the surface of hot metal elements as a result of reactions taking place with the surrounding air. It consists of iron oxides Fe_2O_3, Fe_3O_4, FeO, permanently adhering to metal elements. It should be noted that the iron scale layer is thin – its thickness is equal 5 ÷ 65 μm [12].

In addition, it should be mentioned that in recent years there has been an increasing share of recycled materials and energy recovery in the production of new polymer or metal products. It is caused both by the growing awareness of citizens regarding the need to care for nature and the limited natural resources, as well as by the stiffening of environmental protection law [13,14].

The aim of the presented below research was to determine the thermal, mechanical and electrical properties of composites based on a polymer matrix with a filler coming from metal, post-consumer elements. The analysis was performed on samples prepared from mixtures containing different percentage compositions of high-density polyethylene and mill scales. As part of the work, impact strength, hardness and dynamic mechanical properties (DMTA) tests were carried out, a static tensile test was performed and the volume and surface resistance value was measured.

Research methodology

The tests were carried out on four different types of samples, with the following percentage compositions:

- unfilled polyethylene,
- 40% polyethylene + 60% iron scale,
- 30% polyethylene + 70% iron scale,
- 20% polyethylene + 80% iron scale.

Polyethylene used in the work was obtained from recycling, then the material was cleaned, ground and prepared for processing. In turn, for the needs of the analysis, the iron scale was mechanically removed from the post-consumer iron elements, whereby its chemical composition was determined using the scanning electron microscopy method with an electrodes dispersion detector (SEM-EDS). The obtained results of the analysis are presented in Table 1.

Table 1. The chemical composition of iron scale

Element	Percentage [%]
oxygen	25
sulphur	3
carbon	<1
aluminium	<1
iron	70

The polyethylene was ground to a form of powder whereas the iron scale – to flakes and from these types of raw materials appropriate mixtures were prepared. Then samples in the shape of square plates and paddles (Fig. 1) were produced using a hydraulic press with the following processing parameters:

- pressure of 15 MPa,
- mould temperature of 165 $^{\circ}$C.

The pressure and temperature values were selected experimentally until samples with a solid structure were obtained.

Fig. 1. Samples used in the carried out tests in shape of square plates (a) and paddles (b)

The obtained samples were subjected to the following tests: impact, hardness and tensile tests, as well as dynamic mechanical properties (DMTA) test. The volume resistance was also measured in order to investigate how the mechanical and electrical properties of obtained composite material depend on its percentage composition.

An impact test was carried out at room temperature (23 °C) by Charpy testing with applying a pendulum hammer of rigid construction at an energy of 5 J. This test was done using standardised notched specimens of type A. In turn, the hardness analysis was conducted by means of the ball indentation method, applying a hardness tester equipped with a steel spherical indenter of 5 mm diameter.

The tests of dynamic mechanical properties were carried out with the use of the Netzsch DMA 242 device with a three-point bending holder whereby beam-shaped samples with dimensions of 10 × 7 × 4 mm were used. The specimens were subjected to sinusoidally varying mechanical loads with a frequency of 1 and 10 Hz and and heating rate of 3 °C/min in the temperature range from -150 to 100 °C. As a result the graphs were obtained, which allowed for the determination of the values of the storage modulus *E*', loss modulus *E*'' and the tangent of mechanical loss angle tg*δ*.

Tensile strength tests were carried out with the use of the Galldabini Quasar 25 testing machine with the tensile speed of 5 mm/min. Stress at break, elongation at break and Young's modulus were determined in the static uniaxial tensile test.

In turn, the volume resistance R_v, which is the quotient of the DC voltage U and the steady through-current I_v, was measured on the test bench presented in Figure 2.

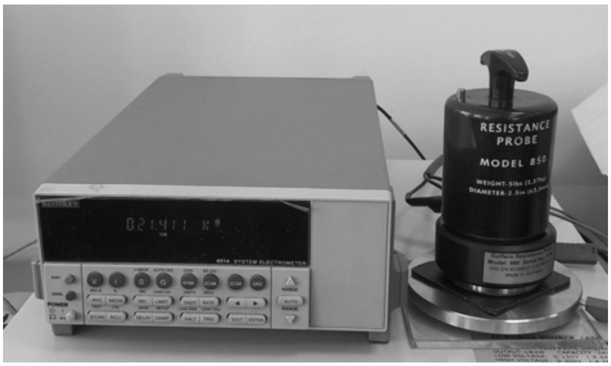

Fig. 2. Test bench used for measurement of the volume resistance

According to the measurement method presented in [15] the voltage *U* is applied between two electrodes, which are placed on two opposite surfaces of the sample, whereas the I_v current is the current flowing between these electrodes. It should be mentioned that during this measurement, the current flowing on the surface of the sample I_s is ignored.

Research results

The conducted research allowed to determine how the percentage composition of tested composite samples, consisting of PE-HD matrix and reinforcement i.e. iron scale flakes, has an influence on their mechanical and electrical properties. The results of impact, hardness, DMTA and tensile tests are presented in Figures 3-8.

As it can be seen from diagram 3, the lowest resistance to dynamically acting force was recorded for samples made of unfilled polyethylene. The addition of iron scale caused a significant increase in impact strength – in particular, for samples containing 70% of the filler, an increase in the tested property by 10.4 kJ/m^2 was noted. As the amount of filler was increased further, a sharp decrease in the impact strength was observed. As an example, samples with 80% iron scale show the impact strength value equal to 7.8 kJ/m^2, which is an increase of 1.4 kJ/m^2 compared to the unfilled material and a decrease by 9 kJ/m^2 in relation to samples with the composition of 30% PE-HD and 70% iron scale.

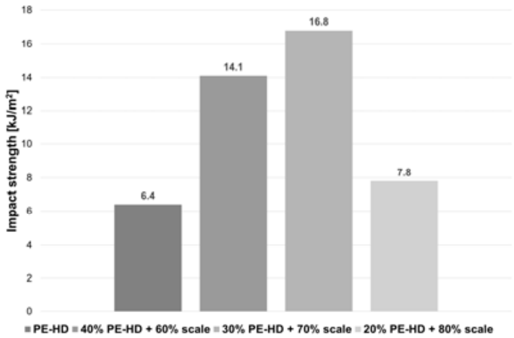

Fig. 3. The results of impact test for all studied composite samples

In turn, Figure 4 shows the results of hardness test by means of the ball indentation method. In the case of unfilled polyethylene, a hardness value of 39.2 MPa was recorded. However – compared to the impact tests – the opposite trend was observed. In the samples with 60% and 70% iron scale, a decrease in the hardness value was noted. On the other hand, for samples containing 20% PE-HD and 80% iron scale, an increase in hardness by 2.4 MPa was noted compared to the samples without the filler.

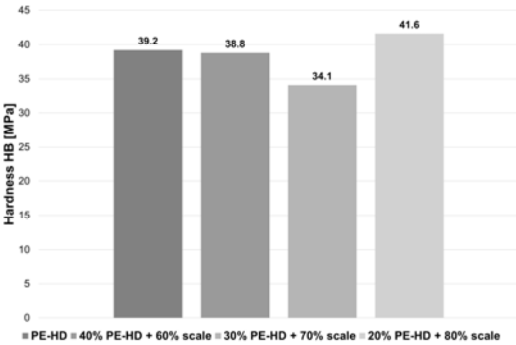

Fig. 4. The results of hardness test for all studied composite samples

The results of tests of dynamic mechanical properties of samples measured at the frequency of 1 Hz and 10 Hz are presented in Figures 5-6. From the tests carried out with the DMTA method, it can be concluded that the addition of the used fillers increases the value of the storage modulus and the tangent of the mechanical loss angle in the full temperature range. For temperatures below 120 °C, polyethylene is in glass state as evidenced by its high hardness and brittleness. With increasing the temperature value, the material undergoes the change of glass transition and enters the viscoelastic, then highly elastic state. This leads to the initiation of Brownian motion and the occurrence of reversible elastic strains. The achievement of the glass transition temperature in the DMTA diagram is seen as a reduction in the storage modulus. At the same time, with the glass transition, the loss angle tangent reaches its first maximum [16].

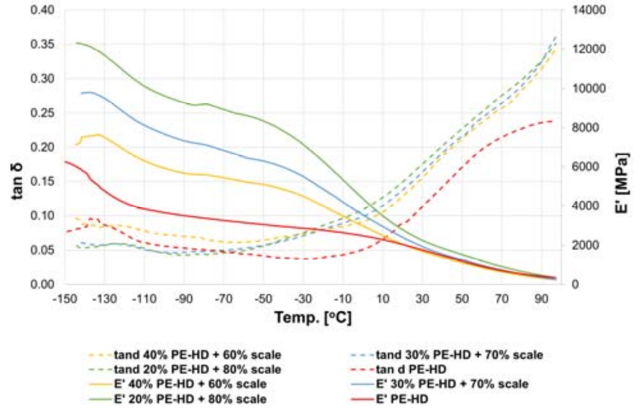

Fig. 5. The results of DMTA at frequency of 1 Hz

For the analysed samples, a similar course of the dependence of the loss angle tangent and the storage modulus on temperature was recorded both for the frequency of 1 Hz and 10 Hz. Moreover, in both cases of frequency for the studied samples, the glass transition temperature for unfilled polyethylene reached a similar value, equal to -127°C. Along with the increase of the filler content, an increasingly higher value of the glass transition temperature was recorded. For samples with 80% iron scale content, the glass transition temperature was 117°C.

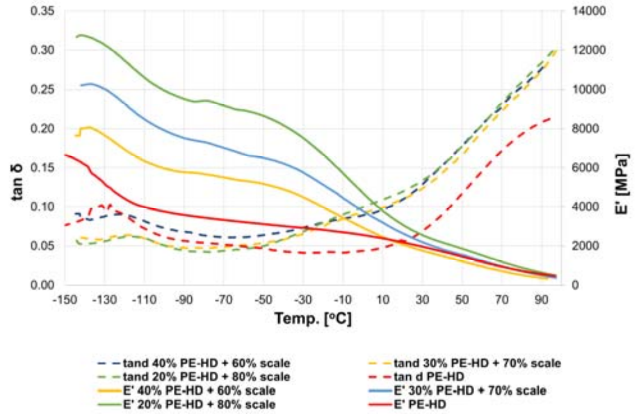

Fig. 6. The results of DMTA at frequency of 10 Hz

The greatest differences in the value of the storage modulus between polyethylene and filled samples in each case were recorded in the glass state. The highest value of the storage modulus, equal to 12317 MPa for 1 Hz and 12758 MPa for 10 Hz, was recorded in the glass phase for samples containing 80% iron scale. In turn, the lowest value of this parameter equal to 6283 MPa for frequency of 1 Hz and 6675 MPa for 10 Hz was observed below the glass transition temperature for unfilled samples.

Furthermore, in the phase of highly elastic strains, the course of the curves of the storage modulus and the loss angle tangent are similar. However, there are still differences in values. In the highly elastic range, most composites also show a greater influence of temperature on the value of the storage modulus. At 0°C for unfilled polyethylene, the value of the storage modulus was 2446.7 MPa for 1 Hz and 2551 MPa for 10 Hz. The highest increase of this parameter in this temperature value was recorded similarly to the glass phase for composites with the addition of 80% iron scale in the form of flakes. As it can be seen, for the frequency of 1 Hz, an increase by 1880.5 MPa was registered, and for 10 Hz by 2149.7 MPa compared to PE-HD. In each analysed case, at the temperature of about 90°C (i.e. in the end-use range of polyethylene), the differences in the course of the storage modulus curve are small. The modulus takes values closer and closer to zero, while the values of the tangent of the loss angle reach their maximums. It proves to a smaller and smaller reaction to the extortion of the material and its transition to a plastic state.

The results of research obtained in a static tensile test are presented in Figure 7-9. From these diagram it can be observed deterioration of the studied mechanical properties in the case of samples with the filler addition. Both in the case of Young's modulus, as well as elongation and stress at break, the greatest decrease in value was recorded for the sample with 80% iron scale content.

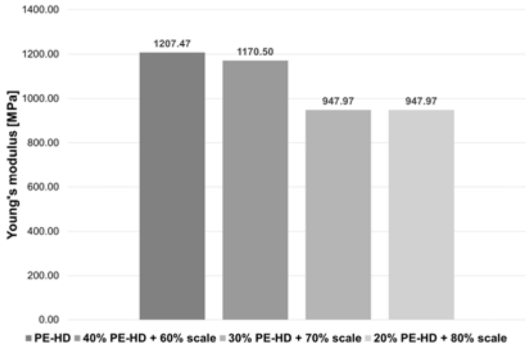

Fig. 7. The results of static tensile test – Young's modulus for studied composite samples

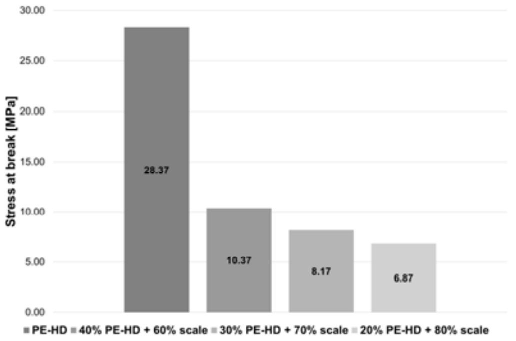

Fig. 8. The results of static tensile test – stress at break for studied composite samples

For Young's modulus, a decrease of 259.5 MPa was noted and a significant drop in the value of elongation and stress at break was observed. The samples with the highest filler content at the moment of fracture had an elongation of 17.53% at a stress of 6.87 MPa. On the other hand, for unfilled polyethylene, the elongation value of 81.87% and the stress of 28.37 MPa were recorded. Deterioration of mechanical properties in the static tensile test along with the increase in the amount of iron scale is related to the geometry of the used filler. The content of iron scale in the form of flakes causes irregular distribution of the filler, which results in the possibility of the occurrence of places with a small amount of material (polymer matrix). This, in turn, leads to a reduction in mechanical properties under this type of load.

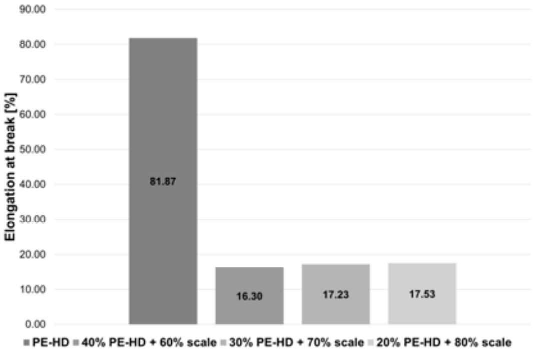

Fig. 9. The results of static tensile test – elongation at break for studied composite samples

As part of the research, the volume resistance of the obtained composite samples was also determined, the results of which are presented in Table 2. As it can be seen for samples from unfilled polyethylene, a volume resistance value above 200 GΩ was recorded. Significant decrease in resistance was noted for samples with filler content. As an example, for the PE-HD + 60% iron scale composite, a value of 46.25 kΩ/mm was measured. In turn, in the case of samples with 70% iron scale, a decrease in the volume resistance was noted by 43.82 kΩ/mm compared to the above mentioned composite. Further increase of the filler content did not significantly affect the measured parameter, as indicated by the results for samples with 80% iron scale - their volume resistance value was 2.31 kΩ/mm.

Table 2. The results of volume resistance measurements of composite samples

Sample	Volume resistance R_v [Ω/mm]
PE-HD	$>200 \circ 10^6$
PE-HD + 60% iron scale	$46.25 \circ 10^3$
PE-HD + 70% iron scale	$2.43 \circ 10^3$
PE-HD + 80% iron scale	$2.31 \circ 10^3$

Conclusions

The carried out research allowed to determine the mechanical and electrical properties of the considered composites. The analysis shows that the content of the filler in the form of iron scale flakes significantly affects the properties of polyethylene composites. The greatest differences in properties were recorded between the unfilled material and the samples containing 20% PE-HD and 80% iron scale. In composite samples of this composition, an increase in the impact toughness and hardness values as well as an improvement in dynamic mechanical properties were noted. In the samples with 70% and 90% iron scale, a significant reduction in the volume resistance was recorded. The applied form of the filler causes deterioration of the mechanical properties recorded in the static tensile test.

The obtained research results provide the basis for the use of recycled polyethylene composites and iron scales for the production of electro-insulating materials with satisfactory thermomechanical properties.

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REFERENCES

[1] Legocka I., Wierzbicka E., Al-Zahari T.M., Osuwaru O., Wpływ modyfikowanego haloizytu na strukturę, właściwości cieplne i mechaniczne poliamidu 6, *Polimery*, 58 (2013), nr 1, 24-32.
[2] Gnatowski A., The research of mechanical properties and structure of the polyethylene composites with cardboard fibre, *Composites Theory and Practise*, 8 (2008), No 1, 99-103.
[3] Ostrowska Czubenko J., Pieróg M., Gierszewska M., Modyfikacja chitozanu - krótki przegląd, *Wiadomości chemiczne*, 70 (2016), nr 9-10, 657-679.
[4] Kolasińska, E., Szmaja, A., Napolski, G., Świeboda, T., Tazbir, I., Modyfikacja właściwości elektrycznych polimerów i tworzyw sztucznych, *Maszyny Elektryczne: zeszyty problemowe*, 119 (2018), nr 3, 29-34.
[5] German J., *Podstawy mechaniki kompozytów włóknistych*, Politechnika Krakowska, Kraków 2001.
[6] Jakubas, A., Łada-Tondyra, E., Makówka, M., Chyra, M., Sochacka, O., Suchecki, Ł., Koncepcja wykorzystania surowców pochodzących z recyklingu do wykonania kompozytowych materiałów ekranujących pole elektromagnetyczne. *Przegląd Elektrotechniczny*, 97 (2020), nr 12, 182-184.
[7] Jakubas, A., Chyra, M., Gnatowski A., The Influence of Reinforcement Fibers on Mechanical and Electrical Properties of the Electrical Engineering Composites. *Acta Physica Polonica A*, 135 (2018), No. 2, 193-195.
[8] PlasticsEurope Market Research Group (PEMRG)
[9] Kakar, M. R., Mikhailenko, P., Piao, Z., Bueno, M., & Poulikakos, L. (2021). Analysis of waste polyethylene (PE) and its by-products in asphalt binder. *Construction and Building Materials*, 280, 122492.
[10] Czaja K., *Poliolefiny*, WNT, Warszawa 2005.
[11] Malpass, D. B., *Introduction to industrial polyethylene: properties, catalysts, and processes* (Vol. 45). John Wiley & Sons, 2010.
[12] Tomaszewska, A., Kuc, D., Bednarczyk, I., Żak, A., Woźniak, D., Krawczyk, A., Paterek, W., Wpływ parametrów nagrzewania na grubość i skład fazowy warstwy zgorzeliny wsadów walcowanych. *Hutnik - Wiadomości Hutnicze*, 85 (2018), 8, 278--282.
[13] Małuszyńska I., Bielecki B., Wiktorowicz A., Małuszyński M.J., Recykling materiałowy I surowcowy odpadów samochodowych, *Inżynieria Ekologiczna*, (2012), nr 28, 111-118.
[14] Romański L., Kowalczyk M., Ocena procesu odzysku wybranych odpadów pochodzących z eksploatacji pojazdów samochodowych na przykładzie przedsiębiorstwa Moto-Polis, *Inżynieria Rolnicza*, 2 (2010), nr 120, 149-156.
[15] Lisowski M., *Pomiary rezystywności i przenikalności elektrycznej dielektryków stałych*, Oficyna wydawnicza Politechniki Wrocławskiej, 2004
[16] Pramoda K.P., Liu T., Effect of moisture on the dynamic mechanical relaxation of polyamide-6/clay nanocomposites, *Journal of Polymer Science, Part B: Polymer Physics*, 42 (2004), No. 10, 1823–1830.