Metal-polymer composites for electromagnetic interference shielding applications

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Abstract: Electromagnetic properties of materials are an important topic due to their commercial, military, communication and environmental protection applications [1–14]. Electromagnetic interference (EMI) effects occur due to the emitted EM radiation by electric and/or electronic devices. EMI may affect the devices causing malfunction or fail, which might have very serious consequences, *e.g.* for medical equipment, aeronautics and cars. Polymer composites with metallic fillers are the subject of increasing interest as potential materials which may effectively shield the electromagnetic field. Polymer composites filled with metal particles are advantageous because they are characterized by low specific weight, high corrosion resistance, plasticity and simple, low cost processing methods. Wide variety of modification of polymer composites (in terms of selection of matrix polymer, type of filler and its structure and content), gives possibility to control their electromagnetic properties depending on particular application. That is why polymer composites with metal fillers are still one of the most important materials to be considered for EMI shielding application. Current review covers theoretical bases and discusses selected experimental results concerning important polymer composites EMI shields developments. Polymer composites EMI shields is a multidisciplinary subject and current review should be useful for the specialist from different areas of research and technology.

Keywords: polymer composites, electromagnetic interference shielding, metal filler.

Kompozyty polimerowe z napełniaczem metalicznym do zastosowania w ekranowaniu pola elektromagnetycznego

Streszczenie: Materiały o właściwościach ekranujących promieniowanie elektromagnetyczne stały się ważnym obiektem badań ze względu na możliwość ich zastosowania militarnego, komercyjnego, komunikacyjnego, a także w ochronie środowiska. Interferencja elektromagnetyczna (EMI), pojawiająca się w wyniku wzajemnego oddziaływania promieniowania elektromagnetycznego emitowanego przez urządzenia elektroniczne, może powodować bardzo poważne zakłócenia, np. w działaniu urządzeń medycznych, samochodów itp. Wśród materiałów skutecznie ekranujących pole magnetyczne coraz większą uwagę skupiają kompozyty polimerowe z udziałem napełniaczy metalicznych. W porównaniu ze stosowanymi obecnie do tego celu osłonami metalowymi, polimerowe materiały kompozytowe charakteryzują się elastycznością i łatwością przetwarzania, małym ciężarem właściwym oraz dużą odpornością na korozję. Szerokie możliwości modyfikacji kompozytów w zakresie wyboru osnowy polimerowej, rodzaju napełniacza, jego zawartości i struktury umożliwiają sterowanie właściwościami elektromagnetycznymi w zależności od docelowej aplikacji.

Przedstawiono podstawy teoretyczne oraz wybrane wyniki prac doświadczalnych dotyczące kompozytów polimerowych z udziałem napełniaczy metalicznych. Ze względu na multidyscyplinarny charakter wyniki te mogą być przydatne w różnych obszarach badań.

Słowa kluczowe: kompozyty polimerowe, ekranowanie pola elektromagnetycznego, napełniacze metaliczne.

Due to rapid growth of electronic industry and electronics devices electromagnetic interference (EMI) shielding continues to be a serious problem [1–15]. EMI is noise emitted by electrical circuits that interferes with the function of other electronic devices. The most common type of EMI is in the radio frequency range and comes from many sources including computer circuits, radio transmitters, electric motors, overhead power lines and others. Preventing it is increasing demand due to the abundance and sensitivity of electronics, particularly radio frequency devices which tend to interfere with digital devices. EMI may affect the devices causing malfunction or fail, which might have very serious consequences, *e.g.* for medical equipment, aeronautics and cars. There is a growing demand for high-speed electronic devices operating at higher frequencies. Especially, mobile phones and smartphones are

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typically operating at 800–900 MHz, and around 2 GHz for data transmission and for this reason most of EMI properties are studied at this frequency range. The effects of EMI can be reduced or diminished by the application of a proper shielding material to protect humans and devices from the adverse effects of this radiation. Consequently, there is a large number of papers published (isiknowledge.com website counted almost 2000 papers covering EMI shielding subject from 2010 to 2016) as well as different aspects of the problem were studied. In the current review the selected aspects of the polymer composites to EMI shielding will be presented according to the lines of the studies, which were carried out at the Industrial Chemistry Research Institute (ICRI) in Warsaw.

In electromagnetic interference (EMI) shielding applications mostly metal based screening materials were conventionally used due to metals high electrical conductivity. In comparison to metals, polymer composites EMI shields are advantageous because they are characterized by low specific weight, high corrosion resistance, plasticity and simple, low cost processing methods. However, despite a very wide range of different types of polymer composites studied much should be done to increase the knowledge about such materials characterization, especially in the area of electrical properties. The most common method for preparing electrically conductive (CPCs) and EMI shielding effective polymer composites is by mixing conductive solid fillers such as metallic powders, metal flakes, metal-coated fibers, metal nanowires and different carbon based materials as carbon black, graphite, graphen or carbon nanotubes into the polymer matrix. EMI polymer composites and nanocomposites can be manufactured in different structures such as bulk, foam and layered structures and their effect on EMI attenuation has been widely studied, too [5].

BASIC NOTIONS OF EMI THEORY

The basic term [3] describing quantitatively shielding properties of the material is shielding efficiency (SE_T). For a transverse electromagnetic wave propagating into a sample with negligible magnetic interaction, the total shielding efficiency (SE_T, dB) of the sample is expressed by the following equation:

$$SE_T = 10 \log (P_{in}/P_{out}) = SE_A + SE_R + SE_I \qquad (1)$$

where: P_{in}, P_{out} – the power incident on and transmitted through a shielding material, SE_A, SE_R – the absorption and reflection shielding efficiencies, respectively, SE_I – a correction term related to the reflecting waves inside the shielding barrier (multireflections). This term is negligible for a single layer material when $SE_A > 15$ dB [16]. The most important mechanism for EMI shielding is reflection, which requires the shielding material to have mobile charge carriers. However, electrical conductivity is not a condition for EMI shielding and it is generally accepted that materials effective in EMI applications should be characterized by volume resistivity around 1 Ω · cm and lower [7, 8, 15]. In general, the reflection loss is dependent on the type of field, frequency and the wave impedance. Consequently, it is required to comprehensively understand/study the nature of noise to design proper shielding material. The absorption loss is a function of the product $\sigma_r \cdot \mu_r$ and the reflection loss is a function of the ratio σ_r/μ_r, where σ_r is the electrical conductivity relative to copper and μ_r is the relative magnetic permeability. Due to their high conductivity, metals such as, *e.g.*, silver, copper, gold and aluminum are excellent for reflection. For instance, superpermalloy and mumetal are excellent for absorption, due to their high magnetic permeability [15]. The reflection loss decreases with increasing frequency, whereas the absorption loss increases with increasing frequency. Other than reflection and absorption, a mechanism of shielding is multiple reflections, which refer to the reflections at various surfaces or interfaces in the shield. This mechanism requires the presence of a large surface area or interface area in the shield. An example of a shield with a large surface area is a porous or foam material. Another example of a shield with a large interface area is a polymer material containing filler characterized by a large surface area. Internal reflections are especially common for composites filled with small dimensions fillers, *e.g.* nanoparticles. They might be polymer composites filled with electrically conductive fillers such as particles/nanoparticles of metals, carbon or carbon fibers. The loss due to multiple reflections can be neglected when the distance between the reflecting faces or interfaces is large compared to the skin depth [6]. Their shielding effectiveness is higher when filler concentration is greater and for higher aspect ratio parameter of filler (ratio of fiber length to thickness/diameter) [6, 17].

The addition of a conductor or semiconductor to an insulator affects the electrical properties of the composite according to the degree of filling and proximity of the conductive particles to other conductive particles. When the conductive particles are isolated, the conductivity of the composite is changed only slightly, even though the dielectric properties may change significantly. However, when the conductive fillers are close to each other electrons can jump the gap between particles, creating a current flow [18]. From the point of view of the current review it is particularly interesting to consider possibility to predict composite polymer EMI shielding properties using theoretical approach. Among many theories describing electrical properties of composite materials the most cited and commonly used is a percolation theory.

One of the aspects of percolation theory refers to the conductivity of a polymer composite system near to the metal-insulator transition. Using the percolation concept, the electrical conductivity above the percolation threshold can be correlated using appropriate equation predicted by the theory. According to the percolation theory there is a threshold concentration of the filler when its particles are close enough to establish a continuous electric current pathway and above this concentration electrical conductivity of the polymer composite increases substantially. The general dependence of the conductivity as a function of the conductive filler concentration is given in Fig. 1 [18, 19].

It should be added that there are two different theoretical models describing the dielectric properties of polymer composites. The first is matrix media model, where the matrix phase surrounds the granular (particle) phase at all volume fractions and the distance between the conducting particles is greater than the tunneling distance for electrons. This is usually best described using the Maxwell-Wagner effective media equation (also known as the Maxwell-Garnet equation). The second nanostructure is where the conducting particles, in a two-phase material, make electrical contact with each other, when the volume fraction of the conducting particles (V) reaches a certain critical (V_c). At this point, a critical cluster is formed and there is a sharp (usually many orders of magnitude) change in the DC conductivity. The complex electrical conductivity of these systems is best described by the two-exponent phenomenological percolation equation (TEPPE) also known as the general effective medium (GEM) equation [19].

Finally, the question arises how thick the polymer composite of the cable should be? Theoretically, when we assume predominantly reflective shielding the thickness should not be too high. The following equation (2) for skin depth is given below. Note that skin depth (δ_s) is a function of only three variables, frequency (f), volume/bulk resistivity (ϱ), and relative permeability (μ_r).

$$\delta_s = [(2\varrho)/(2\pi f \cdot \mu_0 \cdot \mu_r)]^{1/2} \tag{2}$$

where: ϱ – bulk resistivity ($\Omega \cdot$ m), f – frequency (Hz), μ_0 – permeability constant ($4\pi \cdot 10^{-7}$ H/m), μ_r – relative permeability constant (usually ~ 1).

For calculating the approximate skin depth we should measure the volume/bulk conductivity (resistivity) of polymer composite.

For instance, according to the above equation the skin depth of Cu is 0.66 μm at the frequency of 10 GHz and 66 μm at the frequency of 1 MHz. Consequently, a thickness of 0.1 mm should be enough to efficiently shield from frequencies above 1 MHz. At lower frequencies the materials with a good magnetic conductivity (as well as electric conductivity when dealing with eddy currents) and thicker material may be needed.

OVERVIEW OF POLYMER COMPOSITES MATERIALS FOR EMI SHIELDING

The interesting summary of the current status of EMI materials is presented in patent application [17]:

"In spite of considerable effort, there is still a need for electromagnetic interference shielding that effectively operates at higher frequencies, is compact, thin, lightweight, and is suitable for wide frequency bands. Further, there is a need for simpler and versatile methods to prepare these materials for use in electromagnetic interference shielding." From the literature search it is evident that polymer composites consisting of usually dielectric polymer host/matrix and the fillers are the most promising materials for EMI shielding applications. There is a large number of publications dealing with EMI shielding polymer composite materials. In this paper only the basic information concerning the correlation between the dielectric (conductivity) properties and EMI shielding effectiveness will be presented.

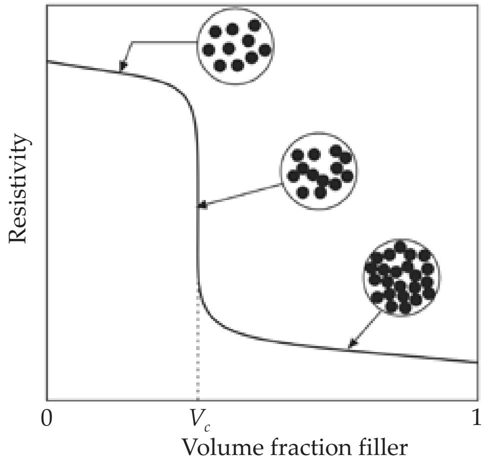

Fig. 1. Percolation phenomenon as applied to conductive composites, showing the development of conductive pathways with an increase in volume fraction of filler [18, 19]

The review of theoretical models concerning the correlations between the polymer composite composition/structure and dielectric properties indicates that the theoretical models may not be useful as quantitative description of the most polymer composites materials with metallic fillers since the composite material structure and properties are not so well defined in those cases. Instead a qualitative/quasi-quantitative description of the development directions can be deduced from the theory. The general direction of EMI composite materials studies is to design the material in such a way that percolation threshold is observed at low concentrations of the filler. It is well known that fibers and flakes metallic fillers are better than spherical fillers due to lower concentrations necessary to achieve percolation. The most promising composite materials are nanocomposites and one of the most important factors to consider is aspect ratio L/d which is the ratio of the length/size and thickness/diameter of nanowires, (carbon) nanotubes or nanoflakes. It is believed that nanocomposites containing high-aspect-ratio conductive metal nanoparticles should be considered as alternatives to carbon nanotube composites for use in electrostatic dissipation, electromagnetic interference, and other electronic applications, such as heat sinks, that

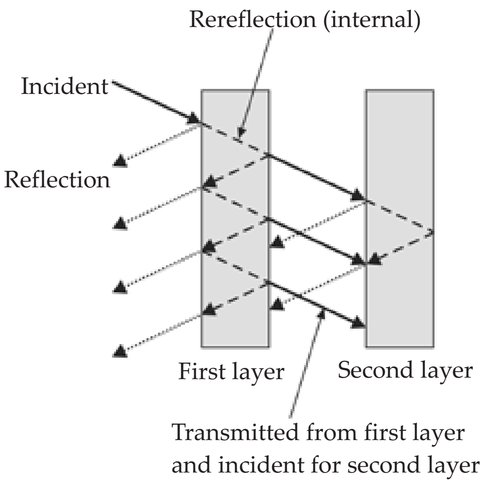

Fig. 2. Effect of stacking method on EMI shielding effectiveness of layered composite [22]

utilize the good thermal conductivity of metals [20]. The paramount importance of the aspect ratio influence on electrical properties of composites can be illustrated on the example of carbon nanotubes [21] where percolation threshold depends very strongly on its value.

The EMI shielding material should be in general characterized by high reflection and low absorption. In this case the best strategy is to apply a few layers of the reflecting copper filled composite material as it is shown in Fig. 2 [22]. This strategy may be illustrated by the results of the paper [22] where electromagnetic interference (EMI) shielding effectiveness (*SE*) of multiwalled carbon nanotubes–poly(methyl methacrylate) (MWCNT–PMMA) composites prepared by two different techniques was established. By stacking seven layers of 0.3 mm thick MWCNT–PMMA composite films EMI shielding effectiveness of up to 40 dB in the frequency range 8.2–12.4 GHz (X-band) was achieved compared with 30 dB achieved by stacking only two layers of 1.1 mm thick MWCNT–PMMA bulk composite. It is interesting to note that according to [22] the reflection shielding efficiency, SE_R increases when number of the layers increases.

Another strategy to improve the properties and economic viability of the composite polymers is functionalization of filler nanoparticles/particles. For instance in [23] the morphological, electrical and rheological characterization of polystyrene nanocomposites containing copper nanowires (CuNWs) functionalized with 1-octanethiol is presented and characterization by SEM (Scanning Electron Microscopy) and TEM (Transmission Electron Microscopy) shows that surface functionalization of the nanowires resulted in significant dispersion of CuNWs in the PS (polystyrene) matrix. The electrical characterization of the nanocomposites indicates that functionalized CuNWs start to form electrically conductive networks at lower concentrations (0.25 vol % Cu) than using un-functionalized CuNWs (0.5 vol % Cu). The organic coating on the nanowires prevents significant changes in the electrical resistivity in the vicinity of the percolation threshold. Usually, functionalization is used to improve dispersion of filler within the polymer as well as cohesion between filler and polymer matrix.

Summing up literature review we may conclude that although it is still not possible to design the composite polymer using a quantitative theoretical model one may find a very useful indications concerning the directions of a new EMI material development. The key is the knowledge of correlation between the polymer composite structure, composition and dielectric properties (measured at laboratory conditions) and shielding efficiency measured on real products, *e.g.* cables.

COMPARISON OF EXEMPLARY POLYMER COMPOSITES MATERIALS WITH METAL FILLERS FOR EMI SHIELDING

In the review article [6] the exemplary data concerning different metal fillers are presented in Table 1. It is evident that the best EMI results are obtained when nickel filler is used. Comparing the results presented in Table 1 for different types of nickel fillers the strong influence of nickel filler shape and aspect ratio are very clear, *i.e.* the highest *SE* is obtained for Ni fibers of the highest aspect ratio. This result could be predicted from the above-presented basic theoretical considerations.

According to [12] the best normalized shielding efficiency is obtained for glass covered amorphous ferromagnetic microwires of $Co_{68.7}Fe_4Ni_1B_{13}Si_{11}Mo_{2.3}$ and $Co_{67.05}Fe_{3.85}Ni_{1.44}B_{11.53}Si_{14.47}Mo_{1.66}$ as the fillers. The comparison of different materials and fillers as presented in [12] is shown in Table 2. However, the manufacturing cost of such sophisticated filler might be a substantial barrier

Table 1. Electromagnetic interference shielding effectiveness at 1–2 GHz of polyethersulfone matrix composites with various fillers – sample thickness 2.8 mm [6]

Filler	Vol, %	EMI shielding effectiveness, dB
Al flakes (15 x 15 x 0.5 mm)	20	26
Steel fiber (1.6 mm dia. x 30–56 mm)	20	42
Carbon fiber (10 mm dia. x 400 mm)	20	19
Ni particles (1–5 mm dia.)	9.4	23
Ni fibers (20 mm dia. x 1 mm)	19	5
Ni fibers (2 mm dia. x 2 mm)	7	58
Carbon filaments (0.1 mm dia. x >100 mm)	7	32
Ni filaments (0.4 mm dia. x >100 mm)	7	87

Table 2. The *SE* of several shielding candidates composites with varying volume fraction of filler loading, sample thickness, and the normalized shielding efficiency at 1–2 GHz in comparison to [12]

Filler	Vol, %	Normalized shielding efficiency dB · (vol %)$^{-1}$ · mm^{-1}	Ref.
Ferromagnetic wires /913 E-glass prepregs	0.026	1	[12]
Al flakes/PES (=PESU – polyethersulfone)	20	$4.3 \cdot 10^{-4}$	[6]
Carbon fiber/PES	20	$3.1 \cdot 10^{-4}$	[6]
Ni fiber/PES	7	$2.7 \cdot 10^{-3}$	[6]
Ni powder/SIM-2030M	8.7	$1.9 \cdot 10^{-3}$	[12]
Stainless steel fiber/ PC (polycarbonate)	1.1	$1.7 \cdot 10^{-2}$	[12]
MWCNT/PAK (polyacrylate)	8.1	$1.9 \cdot 10^{-3}$	[12]
CNT BP (buckypaper)	100	$1.4 \cdot 10^{-2}$	[12]
CNT BP/PE	38	$1.9 \cdot 10^{-2}$	[12]
Fe-MWCNT/PMMA	27	$5.2 \cdot 10^{-3}$	[12]

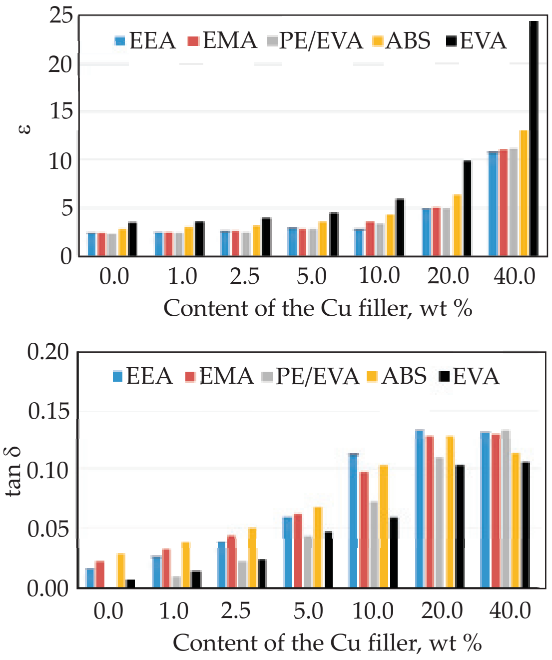

Fig. 3. Complex permittivity (ε) and loss tangent (tan δ) of polymer matrix composites based on EEA, EMA, PE/EVA, ABS and EVA with copper flakes (average thickness 150 nm) measured using split post dielectric resonator method at the frequency 4.7 GHz

in their wider practical application (apart from research subject interests).

Wide range of polymer composites EMI shielding materials was studied by the research groups at ICRI [24–31]. Different fillers metals and polymer matrices were used to manufacture such composites. The dielectric properties of the obtained polymer composites were in most cases measured in the GHz frequency range using split post dielectric resonator method developed by J. Krupka *et al.* [32]. From our studies and literature data it is very

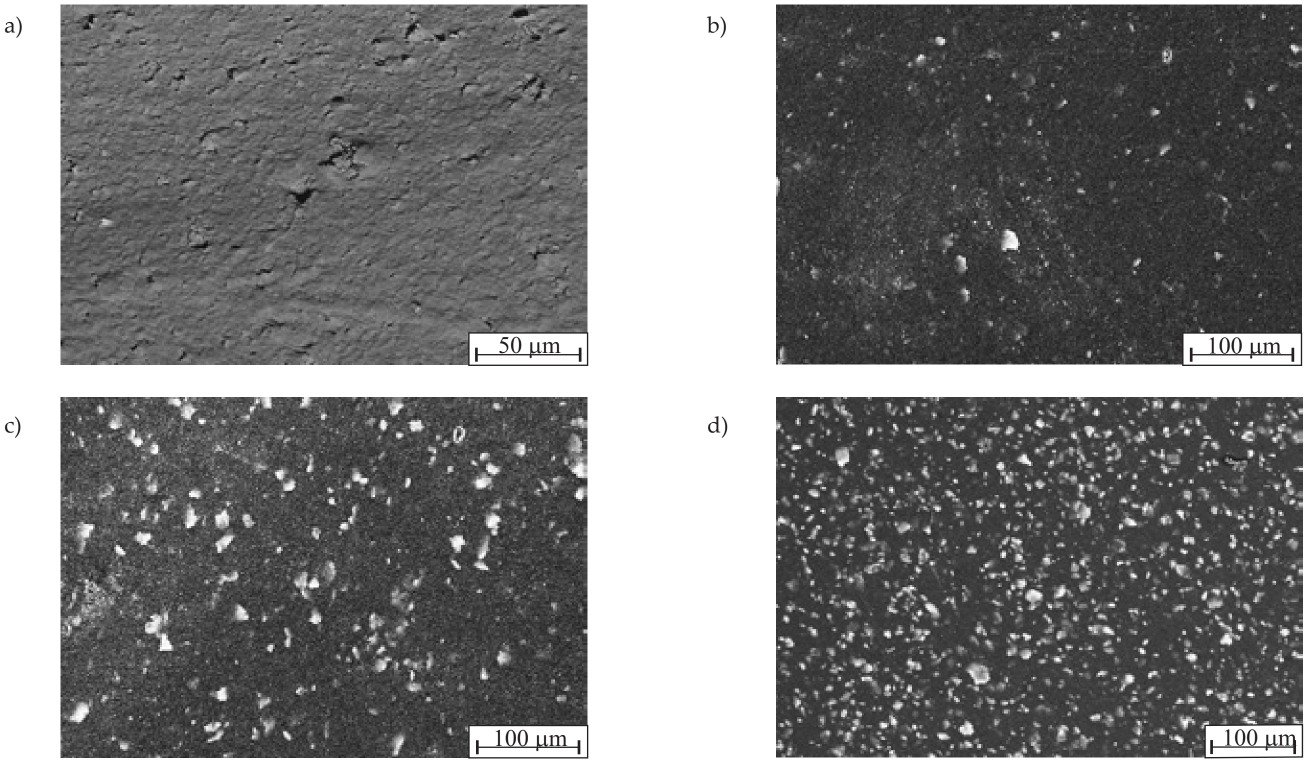

Fig. 4. SEM images of polymer composites based on EVA with increasing content of copper flakes filler: a) 0 wt %, b) 5 wt %, c) 20 wt %, d) 40 wt %

T a b l e 3. Complex permittivity (ε), loss tangent (tan δ) and quality factor (*Q*) of sandwich-type polymer composites based on EEA, EMA, and EVA with different metallic fillers, measured using split post dielectric resonator method at the frequency 1.9 GHz [24]

Metallic filler	Polymer matrix											
	EEA				EMA				EVA			
	wt %	ε	tan δ	*Q*	wt %	ε	tan δ	*Q*	wt %	ε	tan δ	*Q*
Cu flakes	10.0	4.54	0.094	1144	9.7	5.09	0.102	952	7.3	19.74	0.121	670
Ni flakes	2.0	7.60	0.126	416	2.0	4.20	0.134	702	0.7	90.48	0.194	28
Ag powder	5.7	6.87	0.079	905	5.5	5.21	0.173	558	5.7	14.86	0.287	123

clear that comparisons of *SE* presented in [12] are not very reliable because different polymer matrices are used. As it was shown in [24–31] the polymer matrix may influence strongly the electrical/EMI properties of polymer composite when the same filler is used. The influence of the polymer matrix is even more pronounced when sandwich/multi-layered type of polymer composites are used [24]. Exemplary results illustrating the range of different materials studied are presented in Fig. 3. The exemplary structure of polymer composites with copper filler is presented in Fig. 4.

In our previous studies [24–31] electromagnetic properties of polymer composites based on several polymer matrices: ethylene-(ethyl acrylate) plastic (EEA = EEAK), ethylene-(methacrylic acid) plastic (EMA), acrylonitrile-butadiene-styrene plastic (ABS), laboratory prepared polyethylene (PE) and ethylene-(vinyl acetate) plastic (EVA = EVAC) waste blend [PE/EVA (86/14)], and EVA filled with copper and nickel flakes as well as other metals such as silver, were presented. The electromagnetic properties were measured by split post dielectric resonator method [32]. The obtained results showed that with increasing content of copper filler the values of dielectric constant increase which according to the theory should improve their electromagnetic shielding properties. Multilayered polymer composites of sandwich type filled with copper, silver and nickel were also studied (see results below). These composites consist of two layers of polymer with metallic filler between them. In such materials, shielding effectiveness is expected to be higher due to the presence of reflections at the boundary of each of the individual layers (internal reflection). The best measure of the shielding effectiveness in this case is the quality factor *Q* and secondly the value of dielectric constant and loss tangent. The structure of studied sandwich type polymer composite is presented in Table 3 and Fig. 5.

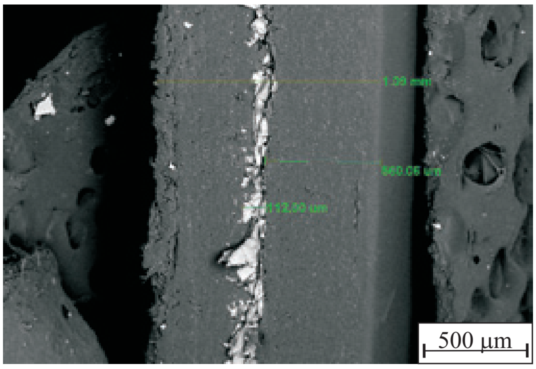

Fig. 5. SEM image of sandwich-type polymer composite based on EVA filled with 0.7 wt % copper flakes

It was concluded that such materials are the most promising in potential practical applications since they show increasing shielding effectiveness along with reduced costs of manufacturing. It should be noted that the selected materials developed in ICRI have been already patented [25] as electromagnetic metamaterials [28, 30].

CONCLUSIONS

On the basis of the above literature review and our (ICRI) previous experimental studies one may propose the following directions of the EMI shielding metal filled polymer composites development (which predominantly is characterized by reflective shielding efficiency):

– Shielding material should possess (if possible) multilayered structure or in general the structure (*e.g.* foam) which enables multireflections.

– The content of copper filler should be decreased by the usage of high aspect ratio metal flakes and/or wires.

– Metal flakes can be functionalized (*e.g.* by treatment of methanol) to improve dispersion, corrosion resistance and adhesion of the metal flakes to the polymer host.

– Polymer composites filled with electrochemically obtained metal flakes are very effective as EMI shields. The ICRI developed and patented method of potential controlled electrolysis (PCEl) was used in the process of the filler manufacturing. The polymer composites with the PCEl obtained metallic fillers are characterized by significantly lower production costs [24–31].

– Metal fillers are still very interesting in the design and manufacturing of the polymer composites EMI shields since they enable relatively easily to obtain the materials of expected electromagnetic, mechanical and other commercially important properties. A very good example are materials developed and studied at ICRI where high dielectric permittivity and low loss tangent composite polymers were obtained and studied.

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