



Prediction of the Dielectric Properties of Some Eco-composite Materials for Energy-related Applications

ANDREEA IRINA BARZIC¹, IULIANA STOICA¹, RALUCA MARINICA ALBU^{1*},
BOGDAN OPRISAN²

¹ Petru Poni Institute of Macromolecular Chemistry, Department of Physical Chemistry of Polymers, 41A Grigore Ghica Voda Alley, 700487, Iasi, Romania

² Grigore T. Popa University of Medicine and Pharmacy, Faculty of Medicine, Discipline Biophysics and Medical Physics, 16 University Str., 700115, Iasi, Romania

Abstract: *The study establishes a theoretical evaluation through several models concerning the dielectric properties of some new eco-composites made of a cellulosic derivative matrix – hydroxypropyl methylcellulose (HPMC) – in which distinct sorts of fillers (ceramic, metallic and bio-derived) were introduced. The investigation describes the impact of the filler addition on the dielectric constant, the dielectric breakdown, and finally how these two factors are contributing to the electric energy density of purposed eco-composites. After incorporation of the reinforcement agents, the dielectric constant significantly increases comparatively with the matrix, as a function of the type of filler used. Moreover, by assessing of the dielectric breakdown, it is observed that with the increase of filler quantity, this parameter slightly decreases for all samples. The data concerning the electric energy density reveal that, by filler insertion in the HPMC matrix, an improvement occurs, especially for the barium titanate system owing to its large dielectric constant. These data are promising for design of new eco-composites having improved dielectric features as demanded for green energy storage devices. Since the materials have biodegradable and biocompatible character, they also have importance in bio-related applications.*

Keywords: *biopolymer, composite, dielectric constant, applications*

1. Introduction

The advantages introduced by polymers make them the materials of choice for numerous applications. Relatively new trends in polymer research are devoted to exploration of materials that are less damaging for the environment [1-3]. Particularly, polymers that are abundant in the nature, like cellulose, are intensively studied owing to their biodegradable, biocompatible and good mechanical resistance properties [4]. Moreover, the drawbacks arising from the low solubility were resolved by chemical approaches [5,6], which showed that the resulted derivatives have a remarkable multifunctionality. For instance, cellulose ethers are generally produced by the reaction of cellulose with aqueous sodium hydroxide and subsequently combined with an alkyl halide, leading to water-soluble compounds even at low degrees of substitution [7-9]. Hence, these cellulose derivatives are occupying a highly important position on the market as a result of their remarkable multifunctionality [10,11], which attracted a huge applicability as anisotropic layers for optical devices [12], additives for pharmaceutical [13] or food products [14], thickening agents for paints or cosmetics [15], components for making adhesives [16], textile fibers [17], and so on.

Hydroxypropyl methylcellulose (HPMC) is a cellulose-ether that has been obtained by replacing hydroxyl groups with methoxy and hydroxypropyl ones [18]. The properties of this biodegradable compound are including solvent-tunable flow behavior [19], good wettability [19], excellent optical features [20], good viscoelasticity [21], insulating character [22] and relatively low refractive index [23]. These remarkable properties can be further improved by incorporation of specific fillers or additives. For instance one may upgrade the dielectric performance of HPMC by inserting compounds known for their relatively high dielectric constant (ϵ), such as ceramic particles, carbon-based fillers, metallic particles or other substances. However, literature is not abundant on data concerning the dielectric

*email: albu.raluca@icmpp.ro

constant enhancement upon HPMC filling [24]. Moreover, in the current context of producing polymer-based devices that are not damaging the environment, it is preferable to use not only biodegradable matrices, but also bio-based fillers are recommendable [25]. Among these, one may mention cereals flower, seeds, leaves, natural resins, minerals and so on. Such materials are known to possess a dielectric constant ranging above 5, depending on the chemical structure, moisture and other experimental conditions [26-28].

This article has the goal to predict the dielectric performance of some eco-compatible composites that have HPMC as matrix. Several types of fillers were selected based on their dielectric performance, namely ceramic compounds (barium titanate (BT), silicon carbide (SiC), strontium titanate (SrTiO₃)), metallic particles (manganese-doped zinc oxide (ZnMnO), titanium dioxide (TiO₂), hafnium oxide (HfO₂), tantalum pentoxide (Ta₂O₅) and bio-derived fillers (fly ash, wood dust, white mica, bentonite). The polymer matrix properties are predicted by means of connectivity index theory elaborated by Bicerano [29]. Upon reinforcement with the selected materials, the dielectric constant is assessed by applying several mathematical models. The results are discussed in regard to the resulted composite suitability for energy storage uses from the point of view of the dielectric performance.

2. Materials and methods

2.1. Materials

Hydroxypropyl methylcellulose (HPMC) is a semi-synthetic polymer, which can be purchased from Sigma Aldrich.

The ceramic compounds, such as barium titanate, silicon carbide and strontium titanate are found as nanopowders at Sigma Aldrich and have dielectric constants ranging between 22-4500 [30-32].

The metallic particles, like manganese-doped zinc oxide, titanium dioxide, hafnium oxide and tantalum pentoxide are acquirable from Sigma Aldrich and have dielectric constants ranging between 23-100 [33-36].

The selected bio-derived fillers (fly ash, wood dust, white mica, bentonite) can be obtained from natural resources and have dielectric constants ranging between 6.4-10.1 [26-28].

2.2. Theoretical methods

The connectivity index theory elaborated by Bicerano [29] enables to calculate several physical properties of polymers based on their structural peculiarities. In this case, this model is applied to predict the dielectric constant (ϵ), as revealed by equation 1:

$$\epsilon = 1.412014 + \frac{0.001887E_{coh} + N_{dc}}{V_{vw}} \quad (1)$$

where E_{coh} is the cohesion energy, N_{dc} is a structural parameter related to the amount of N, O, S, SO₂, F, Si, Cl, Br, while V_{vw} is the van der Waals volume.

The cohesion energy is defined in the work of Bicerano [29] as follows:

$$E_{coh} = 9882.56 \zeta_1 + 358.7(6N_{at} + 5N_{gr}) \quad (2)$$

where ζ_1 is the first order connectivity index, N_{at} is a parameter describing the influence of S, SO₂, Cl, Br, and cyanide atoms, N_{gr} defines the effect produced by hydroxyl, amide, non-amide, carbonyl, alkyl ether and nitrogen groups.

The van der Waals volume could be determined by formula 3, according to Bicerano [29]:

$$V_{vw} = 2.28694 \zeta_0 + 17.14057\zeta_{1,v} \quad (3)$$

where ζ_0 is the zero order connectivity index and $\zeta_{1,v}$ is the valence first order connectivity index.

For the assessment of the dielectric constant of the reinforced materials, several models are proposed [37-41]. Here, the following approaches are chosen to discern the dielectric performance of the analyzed composite samples, namely that additive (or parallel) model (eq. 4), Lichtenecker model (eq. 5), Maxwell–Garnett model (eq. 6) and Knott model (eq. 7) [38-41]:

$$\varepsilon_c = \varepsilon_p w_p + \varepsilon_f w_f \quad (4)$$

$$\ln(\varepsilon_c) = w_p \ln(\varepsilon_p) + w_f \ln(\varepsilon_f) \quad (5)$$

$$\varepsilon_c = \varepsilon_p \frac{2\varepsilon_p + \varepsilon_f + 2w_f(\varepsilon_f - \varepsilon_p)}{2\varepsilon_p + \varepsilon_f - w_f(\varepsilon_f - \varepsilon_p)} \quad (6)$$

$$\varepsilon_c = \varepsilon_p \left[1 - \frac{(\varepsilon_p - \varepsilon_f)w_f}{\varepsilon_f + (\varepsilon_p - \varepsilon_f)w_f^{0.33}} \right] \quad (7)$$

where ε_c is the dielectric constant of the composite, ε_p is the dielectric constant of the polymer, ε_f is the dielectric constant of the filler, while w_p and w_f are the weight fractions of the polymer and filler, respectively.

3. Results and discussions

The dielectric properties of both matrix and proposed composites are discussed in the next sections of the article. A brief critical analysis of the models results is also performed. In the final part of the paper, the electric energy density is evaluated for the selected systems.

3.1. Dielectric constant of the HPMC matrix

The dielectric properties of a polymer are affected by its structural peculiarities involved in the mechanisms of polarization. They can be assessed easily based on the chemical structure of the polymer. The latter is rendering the degree of polarity of the material and hence the probable behavior under a wide of electrical situations. There are few studies in the literature that address the dielectric behavior of pure HPMC [42,43]. Rachocki et al. [42] have investigated the dielectric relaxation and showed that local motions of chain sequences through the glucosidic linkages generate β -type relaxation. Shareef et al. [43] showed that the dielectric constant of HPMC is decreasing upon enhancing the frequency of the electric field. This could be attributed to the fact that at large frequencies atoms and dipoles are not capable to follow the oscillations of the electric field and only electrons contribute to the dielectric constant. The reported measurements were conducted above 35°C and at 10⁶ Hz a dielectric constant of about 6 is attained for pure HPMC. In the case of our HPMC sample, the Bicerano method was employed resulting: cohesion energy of 22350 J/mole, Ndc parameter of 137 and van der Waals volume of 229.85 cc/mole. By introducing these data in equation 1, the theoretical dielectric constant of HPMC matrix is 2.192. The difference between our result and that of Shareef et al. [43] resides in the fact that their measurements were performed at a higher temperature and their sample might contain a high level of moisture, which is known to enhance the dielectric constant.

3.2. Dielectric constant of the HPMC/ceramic particles composites

The ceramic fillers are largely employed for dielectric composite preparation since they provide high polarizability along with electrical insulation [44]. Barium titanate and strontium titanate are compounds with perovskite structure that renders interesting characteristics, including large dielectric constant, along with elevated thermal stability. They are useful for capacitors for the range 500-10000 pF. Silicon carbide is hard chemical compound with crystalline structure with semiconducting features. The dielectric behavior of such ceramic composites having HPMC matrix is further analyzed. According to the results from Figure 1, the highest dielectric constant is acquired for the composites filled with barium titanate, followed by those reinforced with strontium titanate and silicon carbide.

Ferroelectric substances display residual dipoles induced by means of static atomic displacements, which are mainly connected to the perovskite structures. This explains the relatively high dielectric

constant of this group of dielectric materials, which easily exceeds 200, in regard to silicon carbide. Moreover, the differences between these composites are also dictated by the used model for calculation of the dielectric constant. Also, it is revealed that by adding more ceramic fillers (up to 30 wt%) in the HPMC matrix the dielectric properties are considerably enhanced. It seems that this level of loading induces a considerable improvement of the dielectric constant of the composite, in regard to the neat cellulosic matrix. For this composition, the increase in composite dielectric constant is at least of about 1.68-1.91 times, according to Knott model and, at largest of about 3.71-616.58 times, according to additive model.

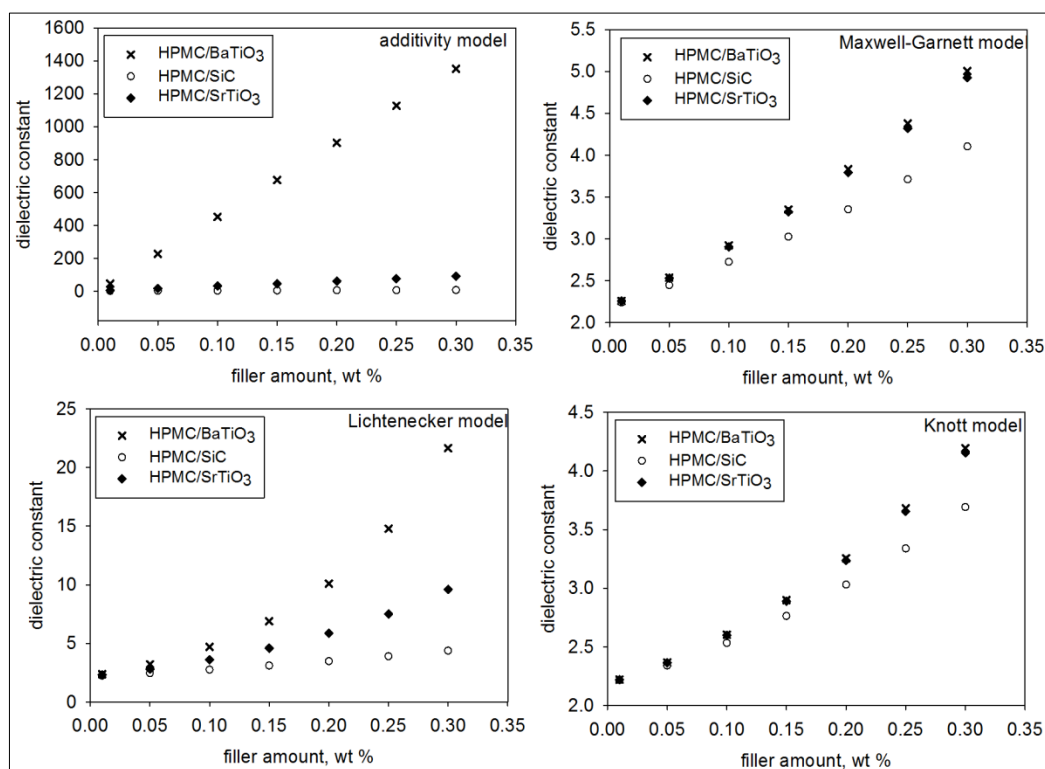


Figure 1. The predicted dielectric constant values for the composites made by reinforcing HPMC with various amounts of the selected ceramic fillers

3.3. Dielectric constant of the HPMC/metal particles composites

The metallic particles that are known for their raised dielectric constant are manganese-doped zinc oxide, titanium dioxide, hafnium oxide and tantalum pentoxide. Generally, these metal oxides are narrow band gap compounds having relatively high leakage current [45]. However, their dielectric performance is comparable to that of some ceramic fillers, like silicon carbide, but is still below that of those with perovskite structure (BaTiO₃ or SrTiO₃).

According to their polarization nature, the metal oxides present nonpolar atomic arrangements that determine the residual polarization to stay for wider periods of time after the imposed voltage is stopped.

As seen in Figure 2, titanium dioxide and tantalum pentoxide are producing the best improvements in terms of HPMC dielectric performance, while insertion of the zinc oxide and hafnium oxide compounds in the same matrix is inducing smaller variations in the estimated permittivity values. The incorporation of the selected metallic fillers in the HPMC matrix leads to composites with permittivity comparable or a little smaller than that of the ceramic-based composites. Therefore, at 30 wt% loading, the enhancement of the composite dielectric constant (in regard to neat HPMC) is at least of about 1.69-1.86 times, according to Knott model and, at largest of about 3.85-14.39 times, according to additive model.

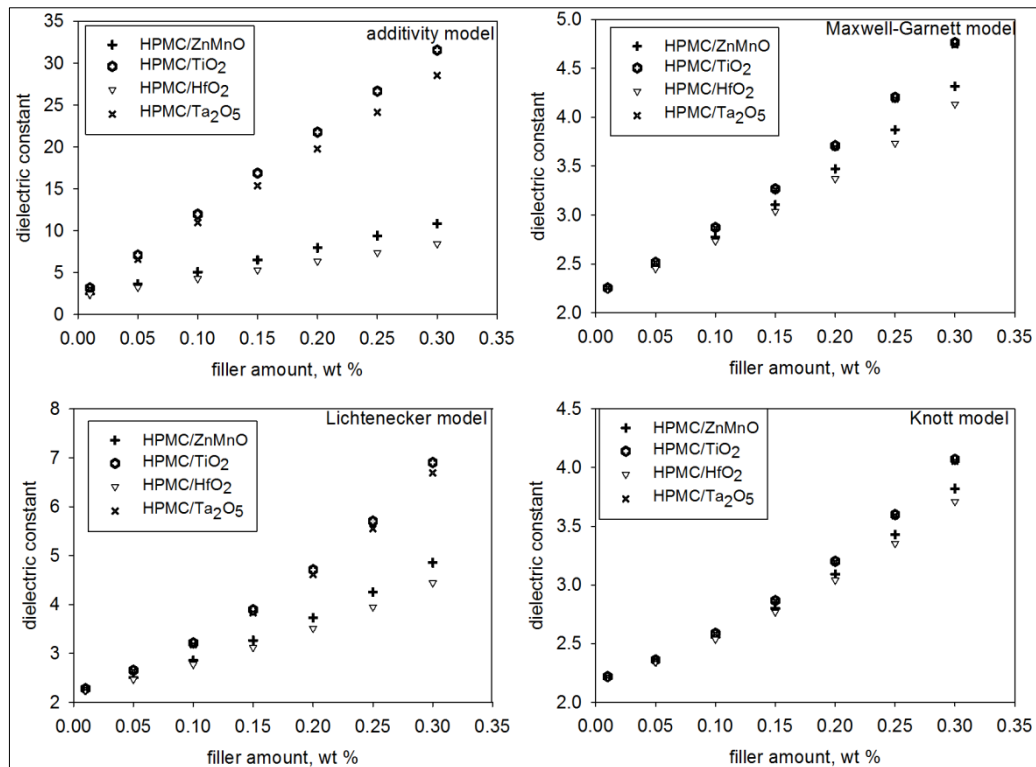


Figure 2. The predicted dielectric constant values for the composites made by reinforcing HPMC with various amounts of the selected metallic fillers

3.4. Dielectric constant of the HPMC/bio-derived filler composites

The utilization of bio-derived fillers is less common for the design of high-permittivity materials. The dielectric analysis of substances originating from biomass or other similar resources is not so much explored in the literature, at least not with the purpose of energy-related applications. Depending on the chemical peculiarities, moisture absorption or other factors, bio-derived fillers exhibit relatively high dielectric constant, but not at the level of previous described fillers. In any case, as noted in Figure 3, these reinforcement agents lead to promising results regarding the dielectric properties of the HPMC eco-composites. The data from Figure 3 indicate that wet wood and bentonite seem to generate higher values of the dielectric constant of the HPMC composite, whereas insertion of the fly ash and mica are leading to lower permittivity values. Analysis of the results at 30 wt% loading, reveal that the magnification of the composite dielectric constant (in regard to neat HPMC) is at least of about 1.23-1.49 times, according to Knott model and, at largest of about 1.32-2.07 times, according to additive model.

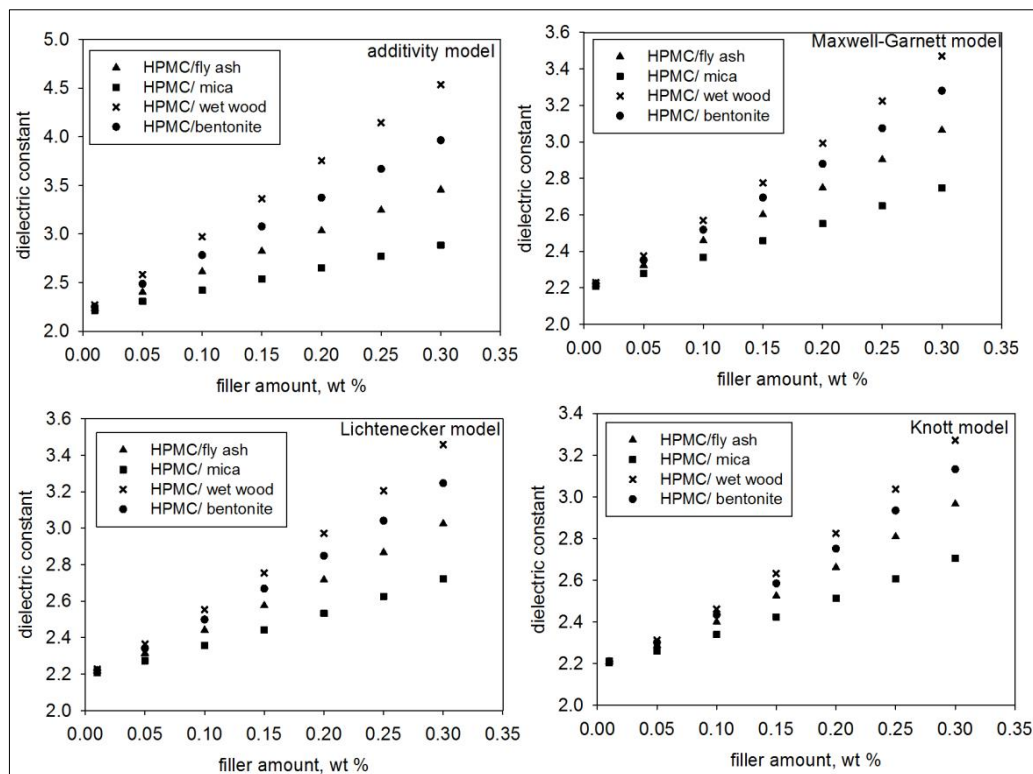


Figure 3. The predicted dielectric constant values for the composites made by reinforcing HPMC with various amounts of the selected bio-derived fillers

3.5. Critical analysis of the models results

The additive or parallel model is useful for estimation of the certain property of polymer composites with at least two phases. It represents the simplest approach that considers the pursued property as being attained by adding the properties of each component of the composite in agreement with the composition expressed by means of the weight fractions or volume fractions. It can be noted that the data attained with this approach (eq. 4) can be regarded as the upper limit for the dielectric constant of each analyzed system.

The Lichtenecker model (eq. 5) considers that the composite dielectric constant is determined by a logarithmic mixing rule. It is utilizable for the cases when the spatial distribution of the loaded particles, their form and orientation is random. Regardless the type of used filler, this model provides high values for the permittivity (next under the upper limit established with parallel model).

The Maxwell-Garnett model (eq. 6) is a mixing rule suitable for homogeneous and isotropic two-phase composites. It assumes that all polymer particles in clusters are spherical. The latter aspect is adequate to simplify and avoid utilization of additional adjustable parameters related to shape of the fillers. The magnitude of the dielectric constant derived from this model for the HPMC composites are smaller than those predicted by additive or Lichtenecker rules.

The Knott model (eq. 7) is an approach that considers the filler as being a small cube covered by matrix medium of equal dimensions. This model is employed for estimation of the dielectric constant of polymeric composites loaded with spherical particles. Even though spherical fillers are slightly distinct from cubic lattices, it is proved that such small difference in the form of fillers is not majorly affecting magnitude of the permittivity of composites. The results achieved for HPMC samples indicate that Knott model leads to the smallest values for the dielectric constant in comparison with the other used approaches.

For all studied composites, the reinforcement determines a linear or non-linear enhancement of the dielectric constant as a function of used model. As the reinforcement level is higher, the differences among the results conveyed by each model are more obvious.

3.6. Electric energy density

The electric energy density of a composite is affected by the magnitude of its permittivity and dielectric breakdown strength. For such dielectric materials, the largest discharge energy density can be described by equation 8:

$$U_d = \frac{1}{2} \varepsilon_0 \varepsilon_c E_{bd}^2 \quad (8)$$

where U_d is the largest discharge energy density, E_{bd} is the dielectric breakdown strength, while ε_c and ε_r are permittivity of dielectrics and vacuum, respectively.

To evaluate the maximum capacity of the studied samples to store electric energy, the data rendered by the parallel model are further employed. Also, the discussions are resumed to the materials that are predicted to give the best results in terms of permittivity, namely barium titanate, titanium dioxide and bentonite. The dielectric breakdown of the studied composites is estimated similarly with the dielectric constant by using the additive or parallel approach. The values of E_{bd} of these fillers are taken from other reports [46-48]. For the polymer matrix, this parameter is evaluated by knowing that the dielectric breakdown is affected proportionally by the yields stress. The latter is found using theory of Bicerano [29] as being of about 3.55 MPa, which leads to a dielectric breakdown for HPMC of about 617 kV/mm. It is observed that as the reinforcement degree is larger, the E_{bd} magnitude of the composite is decreasing, regardless the used filler type. According to Table 1, slightly higher values are attained for bentonite, followed by titanium dioxide and barium titanate.

Table 1. The results of dielectric breakdown for the composites made by reinforcing HPMC with various amounts of barium titanate, titanium dioxide and bentonite

Filler amount, wt%	Ebd, kV/mm		
	HPMC/BaTiO ₃	HPMC/TiO ₂	HPMC/bentonite
0.010	611.567	611.692	611.730
0.050	587.343	587.968	588.155
0.100	557.061	558.311	558.685
0.150	526.780	528.655	529.216
0.200	496.499	498.999	499.747
0.250	466.218	469.343	470.278
0.300	435.937	439.687	440.809

These aspects together with the dielectric constant properties are further reflected in the electric energy density of the samples, as seen in Figure 4. The electric energy density is augmented at higher loadings. Also, in the case of barium titanate-containing composites, it seems that U_d is higher owing to its tremendous high dielectric constant. The predicted values for electric energy density are slightly higher than those reported for other composites [49,50]. These results are promising for further development of green materials utilizable in energy storage applications. Since the studied materials have biodegradable and some of them display biocompatible character, the samples also have great relevance in bio-related applications.

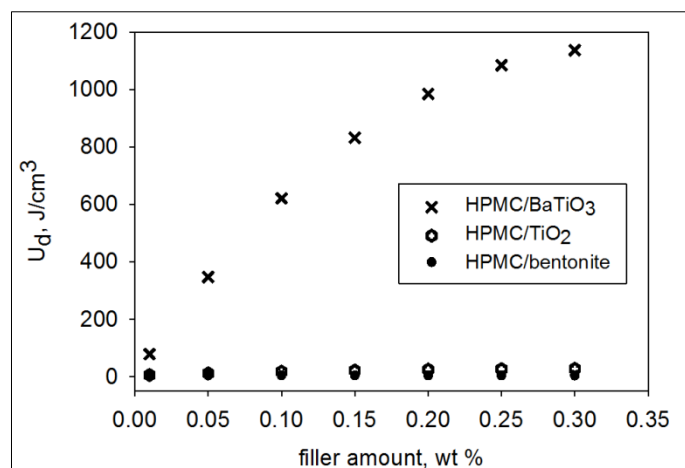


Figure 4. The electric energy density versus filler loading for the composites made by reinforcing HPMC with various amounts of barium titanate, titanium dioxide and bentonite

4. Conclusions

This work reports theoretical studies on novel composites based on a biodegradable cellulosic matrix with several sorts of fillers (ceramic, metallic and bio-derived ones). The dielectric constant of the HPMC is very low ($\epsilon=2.192$) in regard to those of the reinforcement agents (e.g. > 4500 for barium titanate), but upon reinforcement its increases considerably. The best results are given upon filling HPMC with ceramics like barium titanate ($\epsilon=1351$), while incorporation of bio-derived fillers like bentonite provide lower values ($\epsilon=3.96$). Further, the dielectric breakdown of the composite is evaluated, and it is found to decrease after polymer filling, regardless to filler nature. The electric energy density is improved by the filler addition in HPMC, especially for the barium titanate system owing to its large dielectric constant. These data are recommending these composites for energy storage devices with eco-compatible character, but they also can be implemented in biomedical devices.

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