



Research article

Addition of microcrystalline cellulose to an alkyd resin/titanium dioxide film: effect on dielectric properties

Adel Zyane^a, El-houssaine Ablouh^{a,b,*}, El Mouloudi Sabbar^c, François Brouillette^d, Ahmed Belfkira^a^a *Bioorganic and Macromolecular Chemistry Laboratory, Department of Chemistry, Faculty of Sciences and Technology, Cadi Ayyad University, 40000, Marrakesh, Morocco*^b *Center of Analysis and Characterization, Cadi Ayyad University, 40000, Marrakesh, Morocco*^c *Chouaib Doukkali University, Material Physicochemistry Laboratory, BP 20, 24000 El-Jadida, Morocco*^d *Innovations Institute in Ecomaterials, Ecoproducts, and EcoEnergies - Biomass Based (I2E3), Université du Québec à Trois-Rivières, Trois-Rivières, QC, G9A 5H7 Canada*

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ABSTRACT

In this study, various composite films were prepared by varying the amounts of long-oil Alkyd Resin (AR), Titanium Dioxide (TiO₂) and Microcrystalline Cellulose (MCC). The effects of each component, TiO₂ and MCC, on the properties and microstructure of the dry film were determined by examining images obtained by scanning electron microscopy (SEM) and studying the evolution of dielectric properties, the dielectric constant and the loss factor, against frequency at room temperature. Results showed that the introduction of the TiO₂/MCC powder with a weight ratio larger than 1 and a volumetric pigment concentration (VPC) less than 50% allowed a better dispersion of the particles and fitted well the Linchtencker logarithmic mixing law and the Maxwell-Garnett theory. Finally, a marked improvement of the dielectric constant with respect to those of pure alkyd resin and AR/TiO₂ composites was observed.

1. Introduction

The combination of organic and inorganic phases is a convenient way to obtain materials with properties not found in organic polymers or inorganic materials [1, 2, 3, 4, 5, 6]. Since the 1980s, researchers have been looking for composite materials demonstrating synergistic effects between their different organic and inorganic components [7, 8]. Until now, several polymer/ceramic composites have been obtained (e.g. epoxy resin–barium titanate [9], cellulose nanofibers/barium titanate [10], acrylonitrile–butadiene rubber/barium titanate [11]), but the most studied pairs are based on Polyvinylidene Fluoride (PVDF) and Barium Titanate and, in some cases, coupling agents [8, 12, 13, 14, 15]. However, these composites do not perform as expected. In fact, their properties are far from the dielectric characteristics of ceramic materials. Generally, they have a low dielectric constant and the level of ceramics cannot exceed a certain amount without causing a loss of mechanical properties [16]. In addition, the high shear required for ceramic dispersion can sometimes lead to polymer modification and air entrapment in the final material [17]. Finally, some components such as

ferroelectric particles are difficult, or even impossible, to disperse [18, 19].

Our recent studies on composites have shown that the introduction of microcrystalline cellulose (MCC) into a binary composite (Vinyl Resin/BaTiO₃) significantly improves the dielectric characteristics [20, 21]. For example, a Vinyl Resin/BaTiO₃/MCC ternary composite containing 4 wt. % MCC achieved a permittivity value of 13.24 [20]. This study presents a similar experimental scheme with a long-oil Alkyd Resin (AR) replacing the Vinyl Resin. The selected AR is widely used in coatings because of its low cost and ability to disperse pigments and fillers.

Several works have been devoted to the dielectric properties of alkyd resin (AR) such as those of S.A. El-Henawii who found that the value of the dielectric constant (ϵ') increases with temperature and decreases from 4 to 3 with frequency (10^5 – 10^7 Hz) [22]. The two dielectric quantities, ϵ' and loss factor ($\tan\delta$), varied with the oil used [23]. Also, the dielectric constant ϵ' of composite films based on modified alkyd resins and modified Zirconium Titanate (PZT) increased with the PZT fraction. It reached a value of 24 at room temperature, 15.65% PZT, and a frequency of 1 kHz [24]. Recently, the dispersibility of native and surface

* Corresponding author.

E-mail address: lhoussainiblah@gmail.com (E.-h. Ablouh).<https://doi.org/10.1016/j.heliyon.2020.e04977>

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treated Titanium Dioxide nanoparticles in an AR was studied. SEM images revealed the presence of TiO₂ nanoparticle agglomerates whose size depended on the agent used for surface modification [25]. However, the incorporation of TiO₂ particles gave a very slight improvement of dielectric properties [26].

The studies carried out before in our laboratory have shown that the introduction of microcrystalline cellulose, around 4%, significantly improves the dielectric properties of binary composites (vinyl resin/BaTiO₃) [20]. We attributed this improvement to the establishment of bridges and thus to the creation of macrodipoles between vinyl chains and BaTiO₃ particles by microcrystalline cellulose hydroxyl (OH) particles.

The alkyd resin alone or loaded with other ceramic particles did not lead to the expected synergy, no clear improvement in the dielectric constant. So, we have deemed it useful to introduce MCC particles with a hope of establishing links between the hydroxyls still available at the surface of the TiO₂ particles and the functions existing in the alkyd resin (ester, hydroxyl, carboxyls). There is also the fact that this resin is widely used in the paint industry. Its microstructure allows it to easily wet pigments and fillers. This ease of wetting makes it possible to avoid a certain number of problems mentioned before (high shear causing degradation of the binder, presence of agglomerates of particles and air). The objective of this study is to study the effect of adding MCC to the AR/TiO₂ binary composite. For this, we have prepared various TiO₂/MCC combinations by varying the amount of each component. We will finally interpret and discuss the effect of TiO₂ and MCC particles on AR dielectric properties.

2. Experimental

2.1. Materials

The materials used in this study are a long-oil Alkyd Resin (AR), Microcrystalline Cellulose (MCC), Cobalt Octanoate (siccative agent) and Titanium Dioxide (TiO₂). The characteristics of these materials are presented below.

2.1.1. Long-oil alkyd resin

The composition of the AR (70% solids in xylene) is 65% soybean oil; 10% Pentaerythriol and 26% Phthalic Anhydride. Table 1 gives other important AR properties. Figure 1 shows the general structure of this polyester, which comprises three groups: a fatty acid, a polyfunctional acid and a polyalcohol.

2.1.2. Microcrystalline cellulose MCC

MCC (Avicel PH-105) in white powder, 72% crystallinity rate was received from FMC Europe NV (Brussels, Belgium). The powder presents a wide polydispersity of particle shape and size. The average particle size is 20 μm.

2.1.3. Titanium dioxide (TiO₂)

TiONA 128 (previously known as Cristal 128) is an alumina/zirconia modified chloride-process rutile titanium dioxide pigment. Typically, this material has a TiO₂ content of 95% and a specific gravity of 4.1. Its surface is treated with alumina and zirconia. It has an oil absorption capacity of 18g/100g. The TiO₂ particles have a mean diameter (D_n) around 0.19 μm obtained by Disc Centrifuge and a calculated mean (OD) about 0.28 μm via Light Scatter as provided by the supplier.

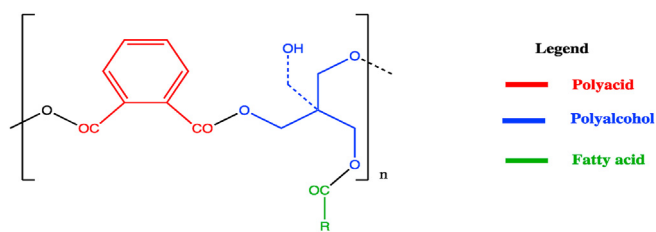


Figure 1. Simplified structure of the alkyd resin (AR).

2.2. Methods

2.2.1. Composite elaboration

Composite materials were prepared with different volumetric pigment concentrations (VPC) by varying the amount and ratio of powder materials (MCC and TiO₂). Beyond 50% VPC, the composite film loses its flexibility. This is why we have prepared films with VPCs ranging from 10 to 50% as shown in Table 2. Predetermined VPCs were obtained by weighting the appropriate masses of each powder (TiO₂ and MCC) before mixing them with the resin.

The dispersion system was a high-speed grinder (impeller apparatus) equipped with a mechanical agitator and a dispersion turbine. The AR, titanium dioxide and MCC were sequentially added under stirring in a stainless-steel beaker. The addition was carried out in small quantities. The fineness of grinding of the mixture was controlled using the North Gauge. As soon as the evolution of the milling reached a constant value, 1% of the siccative agent (cobalt octanoate) was added. A film of the dispersion was applied using a K-Hand coater on an aluminum foil forming one of the two electrodes (Figure 2). Before the dielectric measurement, a metallization of the other surface of the film was carried out by applying a silver lacquer.

2.2.2. Scanning electron microscopy

The surface morphology of composite films was analyzed using scanning electron microscopy (TESCAN, VEGA3), with an accelerating voltage of 20 kV.

2.2.3. Dielectric measurements

Dielectric measurements were recorded using a MTS Modulab (Solartron Analytical). The impedance was measured within a frequency range of 1Hz to 1 MHz at room temperature under a low voltage (0.8V). The surface diameter used in the dielectric measurements was 10 mm for thicknesses ranging from 80 to 100 μm.

3. Results and discussions

3.1. Composite films morphology

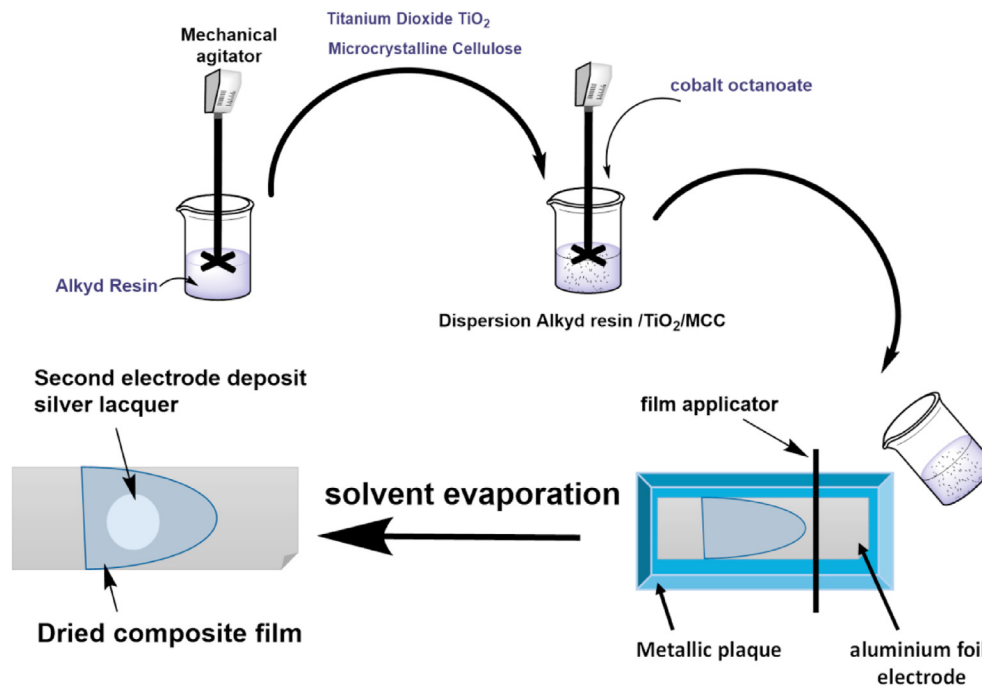
In our previous work on binary and ternary composite films, we have shown that the dispersion of particles has an important influence on the dielectric response [20, 21, 27]. In consequence, we begin the analysis of results by examining SEM images. Figure 3 shows SEM images illustrating the state of particle dispersion in the AR at different VPCs. A global analysis of these images shows that the Volumetric Pigment Concentration (VPC), the quantity of MCC, TiO₂ or the ratio of the two powders (MCC and TiO₂) have a great influence on the state of particle dispersion in the composite film. Indeed, the detailed examination of the

Table 1. Alkyd resin (AR) properties.

Property	Gardner Viscosity	Acid number	Color guard	Reduced viscosity at 25 °C	Dry state density
Value	Z3-Z5	12 max	5 max	46-100 Po	1.2 g/cm ³

Table 2. Composites compositions.

		Volumetric Pigment Concentrations (VPC)				
		10%	20%	30%	40%	50%
% TiO ₂ /%MCC	100/0	2.65	2.99	3.42	3.98	4.78
	50/50	1.33/0.49	2.99/1.09	5.12/1.88	7.97/2.92	12.07/4.37
	75/25	2.01/0.09	4.49/0.55	7.69/0.94	11.96/1.46	17.93/2.19
	25/75	0.25/0.73	1.5/1.64	2.56/2.81	3.99/4.38	5.97/6.56
	90/10	2.4/0.1	5.39/0.22	9.22/0.37	14.35/0.58	21.51/0.87
	10/90	0.27/0.88	0.6/1.97	1.03/3.37	1.59/5.25	2.39/7.87

**Figure 2.** AR/TiO₂/MCC composite film preparation method.

different images shows that no association of grains occurs in composites with $VPC \leq 50\%$ and $TiO_2/MCC > 1$ (Figure 3 a-f). However, Figure 3 (g-l) shows that a poor dispersion is obtained as soon as the volume fraction of MCC exceeds that of TiO_2 , for $VPCs < 50\%$. In these conditions, the presence of different size agglomerates increases with the MCC concentration and even at VPC values lower than 50%.

The particular structure of the AR, which contains hydrophobic and hydrophilic regions, explains its use in paint manufacturing. It can easily disperse surface treated particles such as pigments (TiO_2) and fillers ($CaCO_3$). Since MCC particles have surface hydroxyl groups, they will tend to remain strongly held together by hydrogen bonds. Therefore, their dispersion in a less polar medium remains difficult. The presence of surface treated TiO_2 particles, with $TiO_2 > MCC$, allows an easy dispersion of MCC particles. TiO_2 can be seen as a co-dispersion agent for MCC particles. This is clearly visible in Figure 3 (a-f). TiO_2 particles disperse easily and separate MCC particles. The presence of hydroxyl groups at the surface of TiO_2 particles makes it possible to establish interactions with MCC hydroxyls and thus lead to their separation. This can only take place if TiO_2 particles are more numerous in the medium and surround each MCC particle. If MCC particles are more numerous, MCC-MCC hydrogen bonds will predominate and they will remain agglomerated. The introduction of MCC into the medium also results in an increase of viscosity, which can only be explained by the establishment of a network of strong hydrogen bonds. The uniformity of the distribution of the particles in the

film is a proof of the stability of the particle dispersion in the liquid medium. The analysis of the dielectric response of composites will support these explanations.

3.2. Dielectric properties of composites

Figure 3 (a-e) shows experimental dielectric measurements. These measurements were made in the 100 Hz to 1 MHz frequency range at room temperature. Two types of references are needed to show changes brought by the introduction of each individual powder or their combination (MCC, TiO_2 , and TiO_2/MCC). The first one is the AR alone for binary and ternary composites. The other one, for ternary composites, are combinations of AR/ TiO_2 and AR/MCC. All following Figures will present the evolution of AR dielectric properties. Binary composites, AR/ TiO_2 and AR/MCC, will give the effect of the introduction of each single powder on the resin film. They will also serve as a reference for ternary composites AR/ TiO_2/MCC .

As it can be expected from materials subjected to high viscosity forces at frequencies above a certain threshold, permanent dipoles contribute lightly to the total polarization of the material. The dielectric constant ϵ' of all samples also decreases as the frequency increases. The variation of the dielectric constant ϵ' with frequency has already been used to determine the type of polarization occurring in a material. El-Saied et al. [28] found that dipole orientation polarization could be present in some

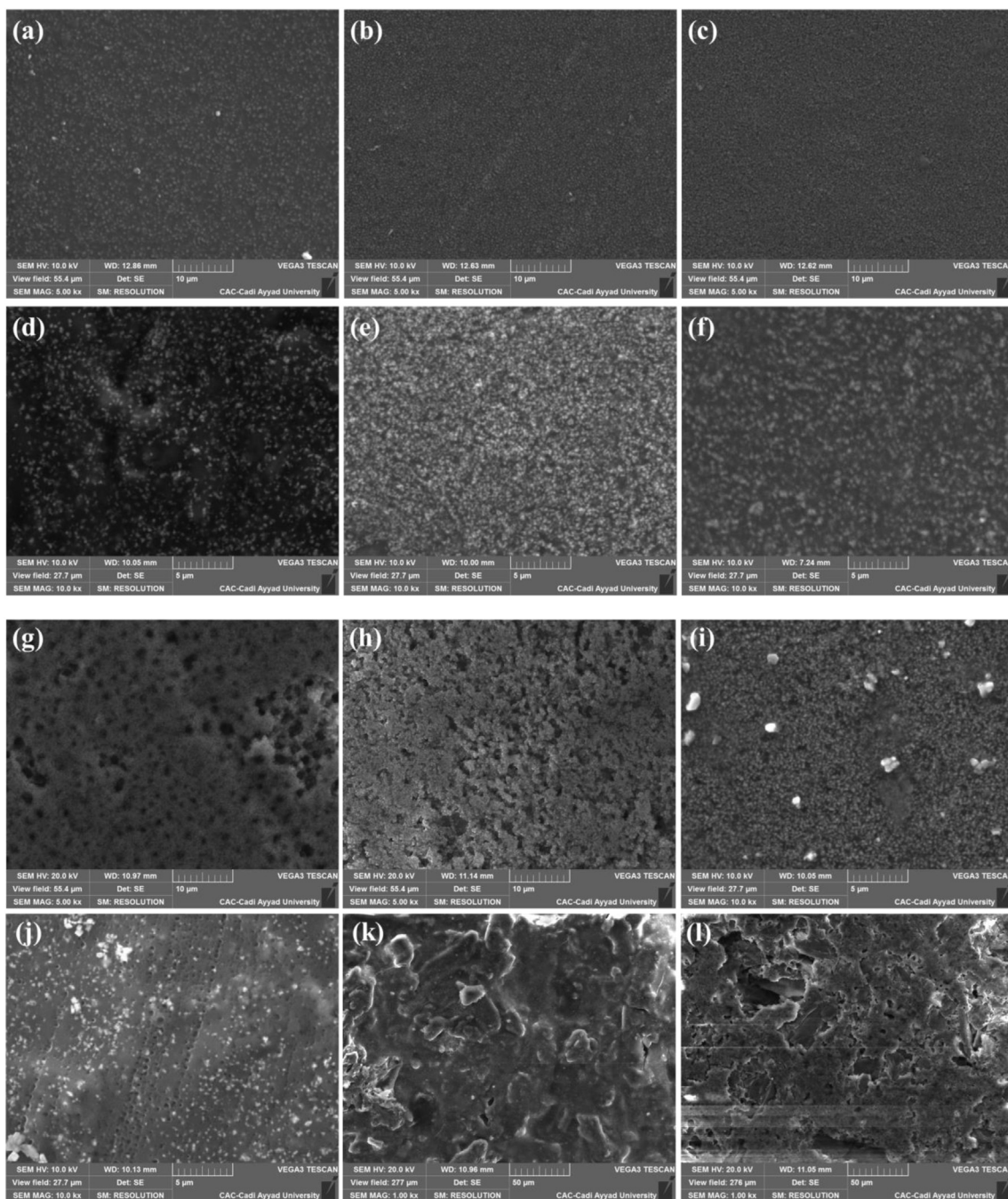


Figure 3. SEM images of composites at different VPCs and different TiO_2/MCC ratios: (a) VPC 10% TiO_2 , (b) VPC 30% TiO_2 , (c) VPC 50% TiO_2 , (d) VPC 30% (TiO_2/MCC 90/10), (e) VPC 30% (TiO_2/MCC 75/25), (f) VPC 30% (TiO_2/MCC 50/50), (g) VPC 40% (TiO_2/MCC 75/25), (h) VPC 50% (TiO_2/MCC 50/50), (i) VPC 30% (TiO_2/MCC 25/75), (j) VPC 30% (TiO_2/MCC 10/90), (k) VPC 40% (TiO_2/MCC 25/75) and (l) VPC 50% (TiO_2/MCC 25/75).

materials at frequencies up to 1010 Hz while electronic and ionic polarizations, still existed below 1013 Hz (Figure 4).

The evolution of the dielectric constant ϵ' of the resin alone, taken as a reference, is relatively constant as a function of frequency, with respect to composite composition. The reason is that the AR, which is polyester coated with unsaturated fatty acids, will give an insoluble and infusible three-dimensional network in the presence of siccative agents and oxygen (Figure 5). The formation of interchain bridges will reduce the mobility of polymer segments and consequently the orientation polarization. Even at low frequencies, there is almost no evolution of the dielectric constant for AR alone.

Our results, illustrated in Figure 4, show that the concentration, the nature of particles (TiO_2 , MCC) and their ratio in the material also have a great influence. The mixing of the resin with the different fillers, alone or in combination, will create more interface whose total area is a function of the dispersion of the different particles. Good dispersion will cause interfacial polarization and the establishment of macrodipoles. There is a clear improvement of the dielectric constant for both types of composite materials, binary and ternary, compared to the AR alone (Figure 4).

Both fillers used do not have the same number of surface functional groups (essentially OH on MCC surface) and particle size distribution. All these factors will influence the response of the composite. Indeed, the

affinity of the resin with respect fillers (TiO₂ and MCC), the interactions between fillers of either the same nature or different, will generate configurations in the space of which the dielectric response will depend.

The introduction of TiO₂ or MCC is marked by a very sharp increase in the dielectric constant ϵ' of the AR. For a frequency of 1 kHz, ϵ' values vary from 4 to 10 (a 2,5X increase) for VPCs of 50% in TiO₂ and 12% for MCC respectively. Despite the small amount of MCC introduced, relative to TiO₂, an identical increase is obtained. However, more MCC cannot be added to the AR due to the hydrogen bonding forces that hold these particles together and the hydrophobicity of the resin that prevents the wetting of MCC particles [29].

For the TiO₂ pigment, which particle surfaces are treated to facilitate their dispersion, there is a linear increase of ϵ' as a function of the amount added. This pigment was designed for the paint industry and therefore disperses well in the AR.

These two charges introduced individually improve the dielectric constant of the alkyd matrix. Their introduction creates more interface in the material generating an interfacial polarization, adding to the other existing types of polarization. The insertion of particles will even prevent rapid formation of bridges, leaving chain segments having more freedom

and thus participate in the total polarization of the material by orientation.

Figures 6 and 7 report the variation of ϵ' as a function of Volumetric Pigment Concentrations (VPC). These results can be used to explain the different dielectric response of the two binary composites. For surface-treated TiO₂, there is almost a proportionality between ϵ' and VPC, whereas for MCC, there is an initial linear relationship between ϵ' and VPC up to a volume percentage of 2%, then there is a random variation. MCC, which is difficult to wet with the long-oil Alkyd Resin (AR), tend to associate with hydrogen bonds from a certain concentration threshold in the medium.

The combination of two charges having a different affinity toward the AR in the same matrix gave better results than binary composites (Figure 4). Compared to the AR alone, there is an improvement of the dielectric constant and there is even a synergy between particles in the enhancement of the permittivity for a certain ratio. This is particularly the case for composites where the percentage of TiO₂ is greater than that of MCC. All composites with a TiO₂/MCC ratio >1 see their dielectric constant increase with the VPC (Figure 4 (a-c)). SEM images (Figure 3 (d-f)) corresponding to these composites show the good dispersion of solid

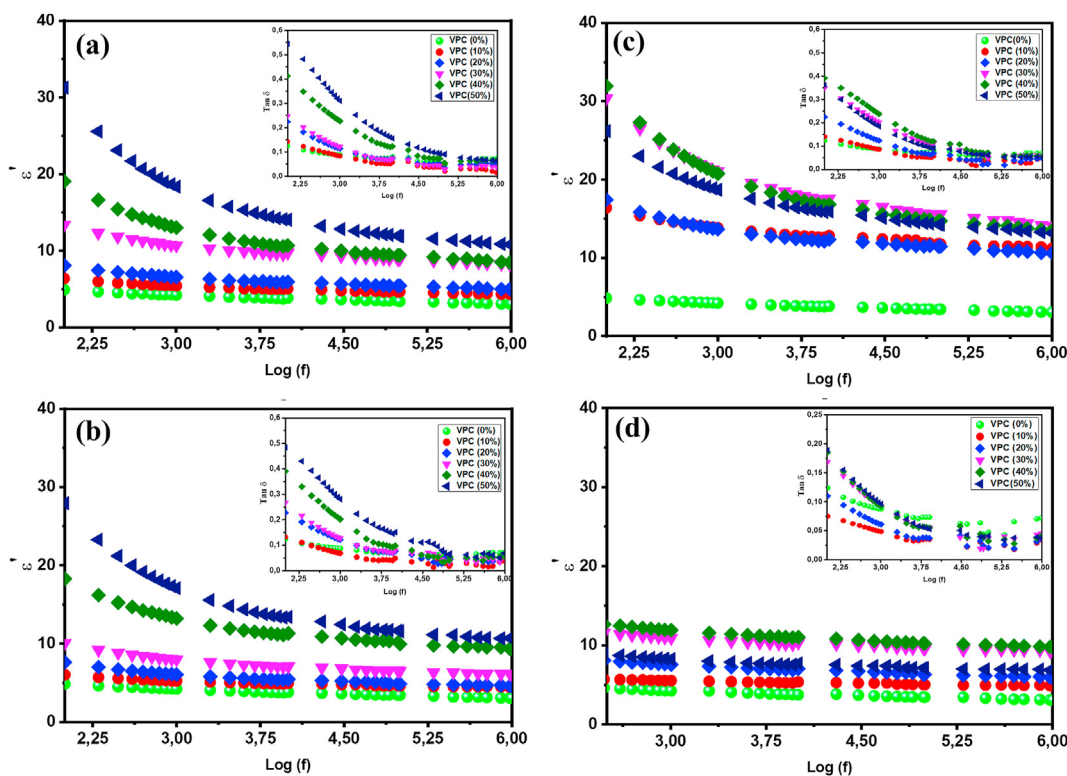


Figure 4. Relative permittivity (ϵ') and corresponding $\tan \delta$ as a function of $\text{Log}(f)$ at room temperature for different VPCs and TiO₂/MCC ratios: (a) 90%TiO₂/10% MCC, (b) 75%TiO₂/25% MCC, (c) 50%TiO₂/50% MCC and (d) 25%TiO₂/75% MCC.

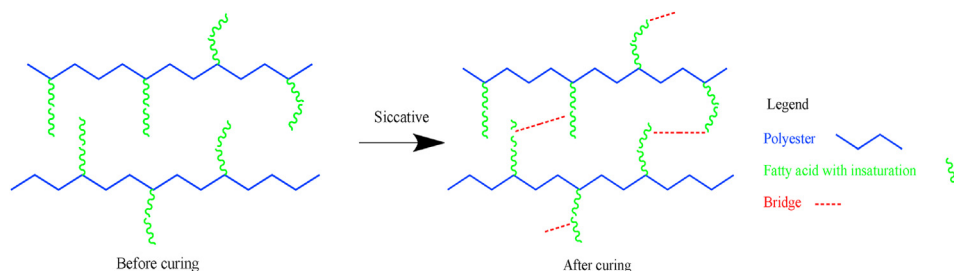


Figure 5. AR structure before and after curing.

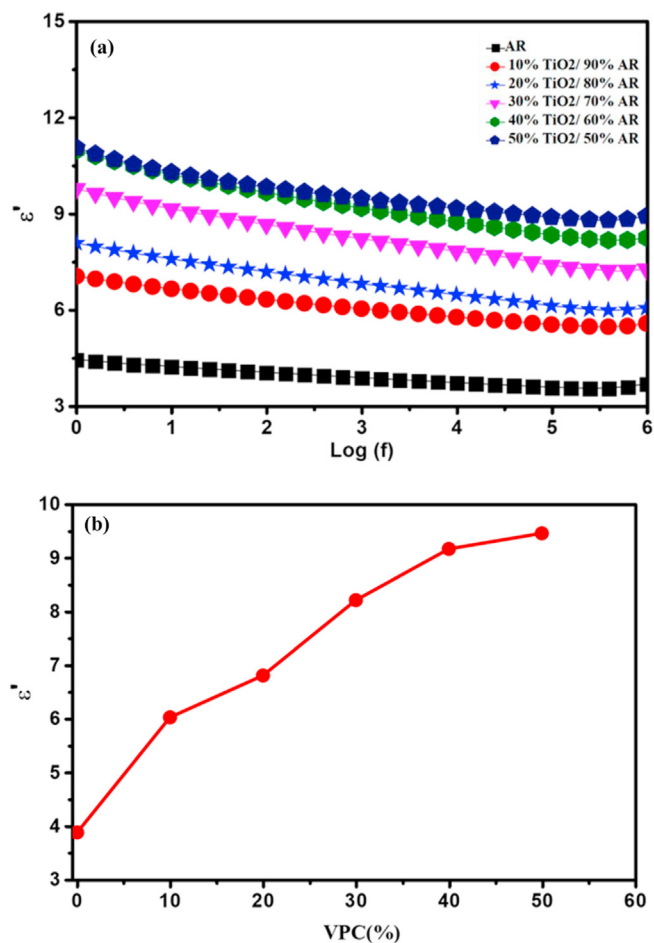


Figure 6. Dielectric response of AR/TiO₂ binary composites at different ratio of TiO₂ (a) and VPCs (b).

particles in the organic matrix. Good dispersion increases the probability of dipolar chain formation and consequently increases the capacity of the capacitor. It can also be seen from these images (Figure 3 (d-f)) and the corresponding dielectric responses (Figure 3 (a-d)) that there is a link between the quality of the dispersion and dielectric responses. Increasing the concentration of MCC particles causes a decrease in the dielectric performance of the composite. This is confirmed by SEM images of composites where the ratio $\text{MCC}/\text{TiO}_2 > 1$. This is clearly visible in SEM images (Figure 3 (h-l)) and on the dielectric response (Figure 4 d). Indeed, the presence of 75% of MCC compared to TiO₂ causes a significant decrease in dielectric constant and poor dispersion. As the number of MCC particles increases in the medium, the probability of collision and association by hydrogen bonding also increases. This results in a poor dispersion that leads to the creation of agglomerates, aggregates and voids preventing the formation of macrodipoles. Increasing the amount of MCC particles, which are very hydrophilic and have hydrogen-bonding promoter hydroxyls, in a hydrophobic medium, will promote the non-isotropy of the composite.

It is quite evident from Figure 8a that the dielectric constant increases with the VPC and the percentage of TiO₂ (Figure 8b) for composites with $\text{VPCs} < 30\%$ and $\text{TiO}_2/\text{MCC} \geq 1$. As discussed above, MCC being polar, it will disperse poorly in a hydrophobic medium. The presence of TiO₂ will allow its co-dispersion and will improve the creation of a certain percolation in the AR/TiO₂/MCC composite. Despite the fact that TiO₂ particles are surface-treated, they still have free hydroxyls and will insert between the AR and MCC, acting as an interfacial agent. Thus, when TiO₂

is in a sufficient amount, it will surround MCC particles and facilitate their dispersion in the resin.

SEM images (Figure 3 (d-f)) are also in good agreement for composites with $\text{VPCs} < 40\%$ and $\text{TiO}_2/\text{MCC} \geq 1$. They show isotropic particle distributions without agglomerates or voids in the material. Other formulations generated composites where the presence of agglomerates and empty spaces is clearly visible. This has repercussions on the dielectric properties of composite materials as shown in Figure 8b.

The two main factors contributing to the variation of the loss factor ($\tan \delta$) are dipole polarization and ion conductance [30]. Our materials have a classical responses, loss factor values are high in the low frequency region and decrease with the increase in frequency, because the polarization is minimal at very high frequencies [31]. Good dispersion creates more interface and thus increases both the constant and the dielectric loss. The examination of Figure 4 illustrates this phenomenon. The dielectric loss varies in the same way as ϵ' . The case of the 75% MCC/25% TiO₂ composite (Figure 4 (d)), is even more significant. The high concentration of MCC causes the formation of agglomerates and aggregates by hydrogen bonding, interrupting the formation of macrodipoles and creating more voids. As a result, the two dielectric characteristics of the composite are reduced. The same result was found in studies by J. Huang using BaTiO₃ as a filler [32].

Good dispersion of the ceramic particles results from the wetting of the particles by the AR and interactions between them. In our composite, ceramic particles are coated on all their surfaces by a film of resin. Examination of SEM images and the dielectric responses allowed us to conclude that there is some synergy for a $\text{TiO}_2/\text{MCC} > 1$ and $\text{VPC} < 50\%$. We compared these results with the most used theoretical models to

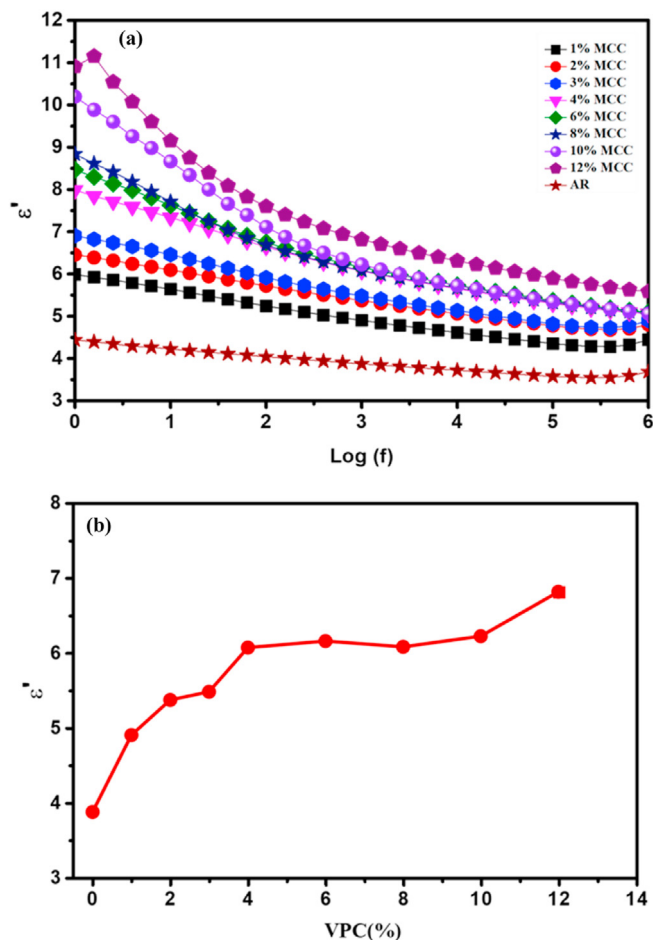


Figure 7. Dielectric response of AR/MCC binary composites at different ratio of MCC (a) and VPCs (b).

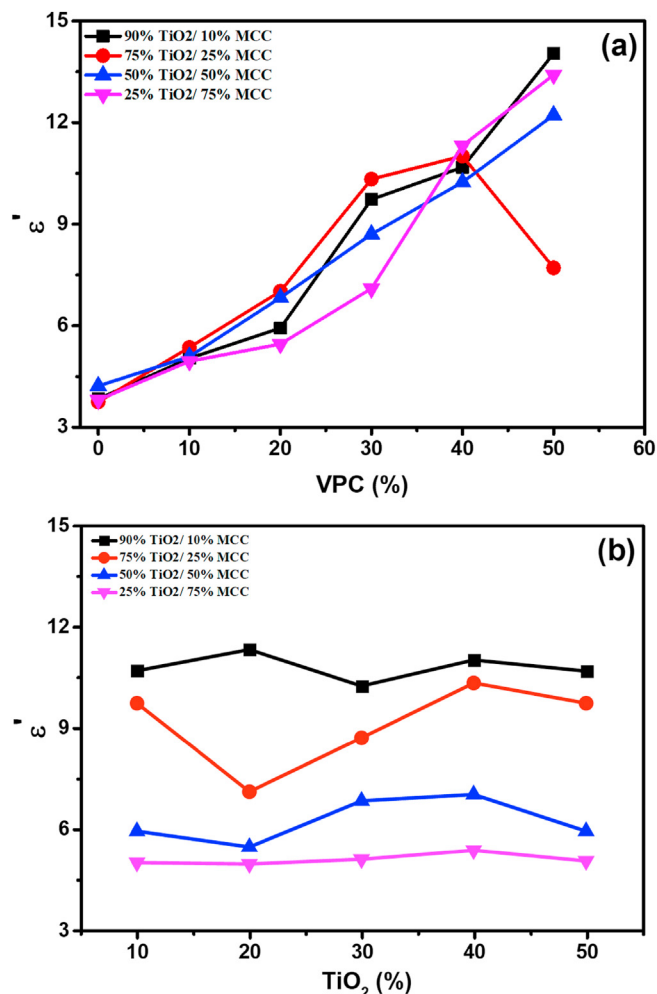


Figure 8. Evolution of the dielectric constant (a) as a function of the VPC for the different TiO₂/MCC ratios and (b) as a function of the % TiO₂ at various VPCs (f = 10kHz).

predict the dielectric behavior of a composite as a function of the particle dispersion state and its volume fraction [33, 34]. Figure 9, illustrates the evolution of ϵ' as a function of the volume fraction of TiO₂ for the two theoretical models, the Linchtencker logarithmic mixing law and the Maxwell-Garnett theory and those of our composites. These results confirm that for composites with TiO₂/MCC>1 and VPC<50%, the evolution of ϵ' as a function of the volume fraction TiO₂ follows the two models. However, as we have also shown before, our curves deviate from the models with the increase in MCC. Indeed, the non-wetting of MCC particles by the resin from a certain concentration level will create the combination of particles of the two charges and therefore not wetting of

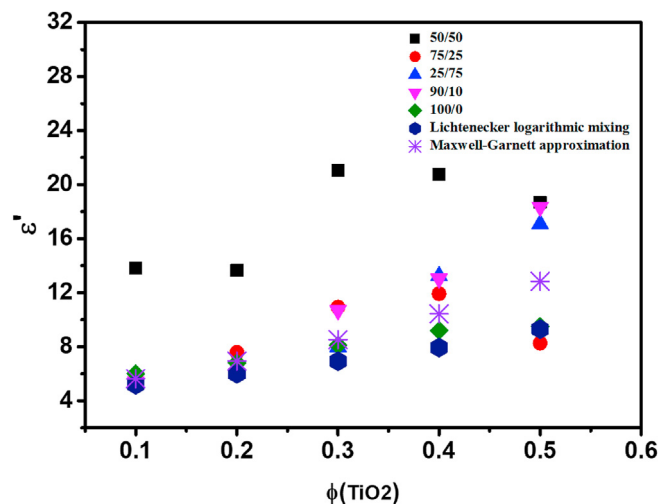


Figure 9. Comparison between numerical models (1 kHz, 25 °C, relative permittivity is 4.5 for AR and 80 for TiO₂) and experimental results of composites as a function of volume fraction.

all the particles contained in the material causing a deviation from the two models.

After studying these materials from the microstructural and dielectric point of view and confronting our results with the Linchtencker logarithmic mixing law and the Maxwell-Garnett theory, we propose the mechanism shown in Figure 9. It illustrates the two cases that illustrate our arguments for the dielectric response of our materials. On the right side of Figure 10, TiO₂ particles, more numerous than MCC, will coat them as surface-active agents for their dispersion. While in the left portion of Figure 10, MCC particles exceeding a tolerable threshold will associate leading to the creation of agglomerates and aggregates causing discontinuity in the material.

It is interesting to compare the results of this work with those reported in the literature, with other composite materials using the same fillers (cellulose derivative or titanium dioxide) but with different polymeric matrices (Table 3). As regards the matrices comprising the nitrile groups, very polar groups, these materials have interesting dielectric and even piezoelectric properties [35]. The values of ϵ' presented in Table 3 are between 3 and 4 except chitosan/modified cellulose, PVA-TiO₂ and MAM-TiO₂ where the values of ϵ' are respectively 8.5, 16 and 14. The high values of dielectric constant obtained for PVA-TiO₂ and MAM-TiO₂ are due to the formation of microcrystalline zones in the matrix [36]. It emerges from these bibliographical data, and those of our results, that the addition of a charge can play the role of a bridge between the various ceramic particles, having high dielectric constants like BaTiO₃ and TiO₂, in an organic matrix is necessary. Moreover, it is the same phenomenon with semiconductor polymers such as polyacetylene. Doping is necessary to have good conductivity. The addition of MCC to an alkyd/TiO₂ binary material markedly improves the dielectric constant.

Table 3. Dielectric constant values of composites materials based on cellulosic derivatives.

Composites materials	Dielectric Constant	References
Bacterial cellulose/polyurethane nanocomposite	3.0	[37]
Chitosan/Nitrile-modified cellulose nanocrystals Biocomposites	8.5	[38]
Aramid fibril/nanofibrillated cellulose composites	3.1	[39]
Nanocomposite polyvinyl-acetate/cellulose nanocrystals	4.7	[40]
S-SEBS/TiO ₂ Composites	3.2	[41]
Polyimide/titanium dioxide/montmorillonite nanocomposite	3.62	[42]
PVA-TiO ₂ nanocomposite and PMMA-TiO ₂ nanocomposite	16.0 14.0	[36]

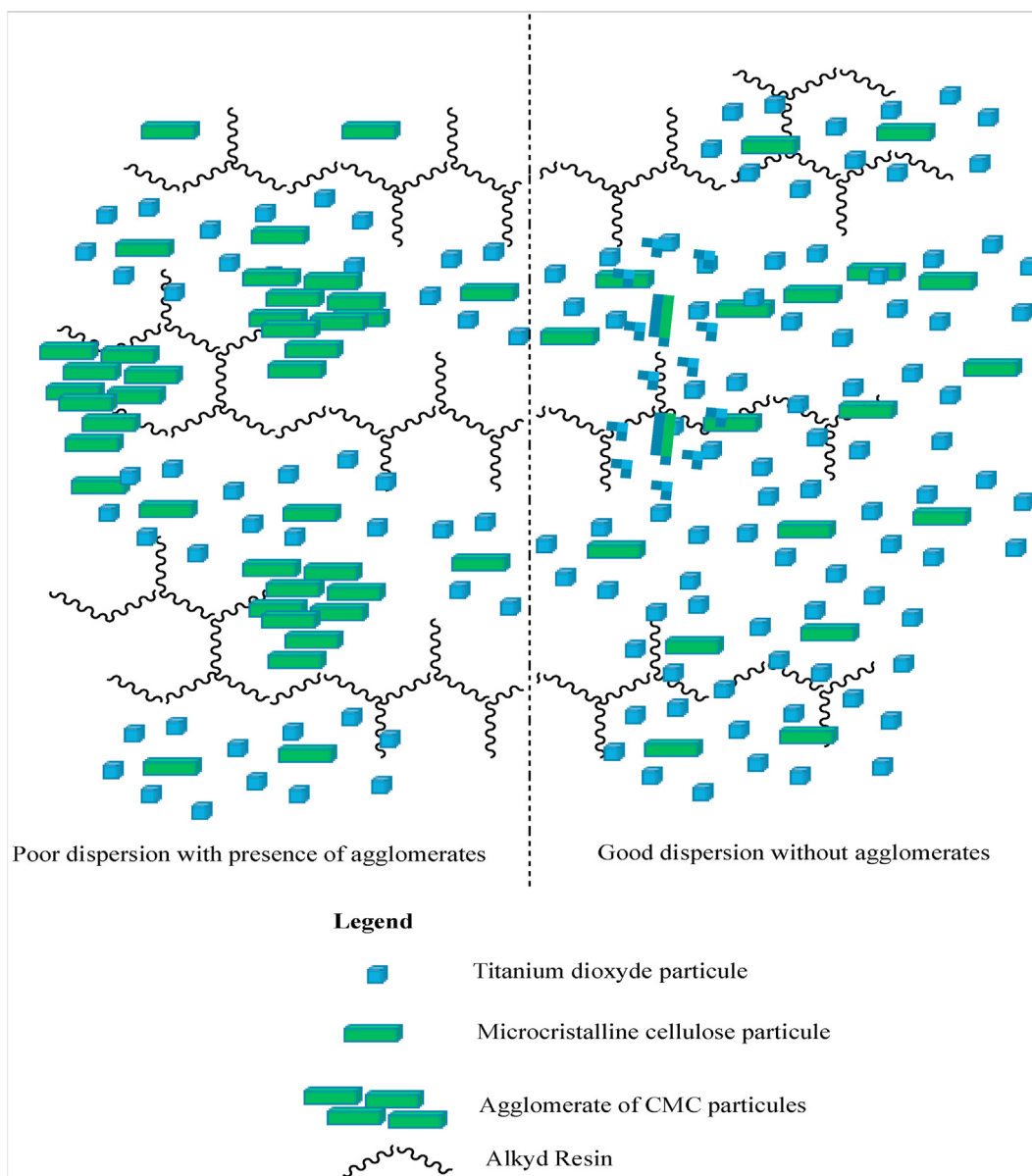


Figure 10. Composite structures for the two cases: $TiO_2 > MCC$ and $TiO_2 < MCC$ for a $VPC < 50\%$.

4. Conclusions

The search for a composite material having good dielectric characteristics continues despite recent performance improvements compared to resin only materials. Our composites, like others, have very modest ϵ' values (30 at low frequencies and around 10–14 at high frequencies). However, this study has shown that microcrystalline cellulose (MCC) particles, held together by strong hydrogen bonds, can be codispersed in an apolar medium with surface-treated TiO_2 particles. For Volume Pigment Concentration values ($VPC < 50\%$) and for $TiO_2/MCC < 1$, images obtained by scanning electron microscopy and the use of both Linchtencker logarithmic mixing law and Maxwell-Garnett theory models are in agreement with particles bathed in a resin (particles coated with the resin). Dielectric losses that accompanied the increase in permittivity are inevitable due to the appearance of space charges between MCC and TiO_2 particles and the polymer matrix.

This study is interesting first of all for paint manufacturers. Indeed, the most difficult step during the manufacture of paints is the dispersion of pigments. CMC, Microcrystalline Cellulose, has proven, in addition to

being ecological, an effective dispersing agent for pigments. The films obtained retain good gloss and excellent adhesion. The second important result is that the introduction of CMC, at low quantity (5%), makes it possible to improve the dielectric constant. We went from a value for ϵ' from 5 to 20 for a frequency of 1 kHz. These films can be used as a dielectric in capacitors and as insulators in electric generators.

Declarations

Author contribution statement

Adel Zyane: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

El-houssaine Ablouh: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

El Mouloudi Sabbar: Contributed reagents, materials, analysis tools or data; Wrote the paper.

François Brouillette: Analyzed and interpreted the data; Wrote the paper.

Ahmed Belfkira: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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