Original Article

Dielectric Analysis of Lanthanum Oxide/Polyvinyl Pyrrolidone Composites for Advanced Electrode Applications

Jagdish P. Yadav [1] , Jitendra K. Mishra [1] , Suchitra Yadav [1] , Anju Yadav [2] , Vikas Kumawat [3] , Balram Tripathi [3] , Dinesh K. Yadav [1, ✉]

1. *Department of Physics, University of Rajasthan, Jaipur, IND*
2. *Department of Physics, Seth RL Sahariya Government PG College, Jaipur, IND*
3. *Department of Physics, S.S. Jain Subodh PG College, Jaipur, IND*

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Abstract

Polymer composites have significant importance in energy storage applications. In the present work, polymer composites of polyvinyl pyrrolidone (PVP) and lanthanum oxide (La_2O_3) have been synthesized with filler (La_2O_3) concentrations varying from 1 mwt% to 5 mwt% using high-energy planetary ball milling. The prepared polymer composite powder is compressed into pellets using a hydraulic press for dielectric measurements. Bonding, structural, morphological, and dielectric characteristics of these composites have been investigated using Fourier transform infrared spectroscopy, X-ray diffraction, scanning electron microscopy, and impedance spectroscopy. The dielectric constant and dielectric loss of LO/PVP composites for varying filler concentrations have been investigated using an impedance analyzer across a broad range of frequencies at room temperature. Results show that the dielectric characteristics of PVP enhance with increasing La_2O_3 filler concentration due to interfacial polarization. The dielectric constant of pristine PVP is observed to be 4.73, and with 5 mwt% La_2O_3 filler concentration, it increased to 28.38 at 1 kHz, making these composites appropriate for energy storage and advanced electronic applications.

Categories: Advanced Materials, Nanotechnology, Polymer Science and Engineering

Keywords: lanthanum oxide, polymer composites, pvp, energy storage, dielectric properties

Introduction

Recently, polymer composites were widely used by researchers in energy storage applications due to their flexibility, straightforward processing, lightweight nature, and excellent electrical insulating properties [1]. Polymers such as poly(vinylpyrrolidone) (PVP) are widely used in polymer composite fabrication due to their flexibility, light weight, ease of processing, excellent dielectric strength, excellent solubility, film-forming ability, and compatibility with various filler materials [2]. However, pristine polymers' low dielectric performance limits their effectiveness in high-performance applications. Dielectric properties of composite materials have attracted significant interest recently due to their wide range of applications, where their ability to store and release electrical energy is important, such as in advanced electronic devices, energy storage devices, and capacitors [3]. To improve dielectric characteristics of polymers, ceramic and metal oxide fillers are often incorporated. Previously reported studies have primarily examined metal-oxide fillers such as Al_2O_3, TiO_2, SiO_2, and ZnO to enhance the dielectric properties of polymer matrices. Lanthanum oxide (La_2O_3) is a rare-earth oxide and exhibits a high dielectric constant ($k \approx 30$), low dielectric loss, excellent thermal and chemical stability, and excellent insulating properties [4,5]. When La_2O_3 filler is incorporated into the polymer matrix, dielectric performance of the composite can be enhanced by interfacial polarization at the matrix-filler interface. These properties make La_2O_3 an attractive and suitable candidate for developing polymer-based dielectric composites designed for energy storage applications. Therefore, La_2O_3 had been selected as the filler material in this present work to analyze its effects on structural and dielectric properties of PVP polymer matrix.

The study of the dielectric behavior of polymer composites is important for developing materials that can operate efficiently under various electrical and thermal conditions [6]. Improvement of dielectric properties of filler-polymer composites depends on various factors, such as size, filler aspect ratio, synthesis method, and interfacial adhesion between fillers and matrix. Recognizing how these parameters affect the dielectric constant, dissipation factor, and dielectric strength of polymer composites is crucial for enhancing their performance in applications such as capacitors, insulators, and other electronic devices [7].

In previous studies, the effect of La_2O_3 incorporation in PVP polymer matrix has not been comprehensively reported on structural characteristics and dielectric response over a wide range of frequency. Therefore, this research paper focuses on the synthesis and investigation of dielectric properties of La_2O_3/PVP composite materials (with 1-5 mwt% of LO) synthesized via the ball-milling technique, which is a solvent-free technique for uniform mixing and effective filler dispersion. By varying the concentration of LO in the PVP matrix, the aim of this study is to determine dielectric stability, dielectric loss, and frequency-dependent behavior. By understanding how an increase in LO filler concentration affects the dielectric performance of composites, we want to develop materials suitable for use in high-energy storage devices. The results may contribute to the development of materials for capacitors, energy storage, and advanced electronic applications where high dielectric performance is required [8]. Dielectric properties of synthesized LO/PVP composites are investigated with a wide frequency range [9]. We systematically studied the effect of La_2O_3 filler concentration (1-5 mwt%) using pelletized samples.

Materials And Methods

Materials

PVP K30, purchased in powder form from LOBA Chemie Pvt. Ltd., Mumbai, Maharashtra, was used as the polymer matrix, and La_2O_3 A.R. with a minimum of 99.9% purity, purchased from HiMedia Laboratories Pvt. Ltd., Maharashtra, was used as the filler. Both materials were used as received, without further purification.

Synthesis of La_2O_3/PVP composite

PVP and La_2O_3 powders were weighed according to the calculated composition that is done with the required molecular weight percentage. Both powders were placed in a ball-milling jar of a high-energy conventional planetary ball mill along with stainless steel balls by maintaining a ball-to-powder ratio of 10:1. Milling was performed at 100 RPM for 2 hours and 40 minutes with a 5-minute pause after every 3-minute milling cycle to minimize thermal effects. During ball milling, PVP and La_2O_3 were uniformly mixed, and agglomeration of La_2O_3 particles was reduced with uniform dispersion of La_2O_3 filler within the polymer matrix. Uniform mixing of filler particles into the PVP polymer matrix and reduced agglomeration were confirmed by scanning electron microscopy (SEM) analysis. After milling, a fine and homogeneous composite powder was obtained. Mixed powder was compacted into cylindrical pellets with a thickness of 1 mm and a diameter of 12 mm using a hydraulic press under a compaction pressure of ~700 MPa. For dielectric measurements, electrodes of silver paste were applied to both the sides of pellets and dried at room temperature for 12 hours to remove residual solvent and ensure proper adhesion, and stable electrical contacts.

Characterization

The powder X-ray diffraction (XRD) patterns of pristine PVP, La_2O_3, and LO/PVP composite materials with 1 mwt% and 5 mwt% filler concentration were recorded using a Rigaku (Miniflex-600) X-ray diffractometer in reflection mode with Cu-K_α radiation of wavelength and a graphite monochromator and Ni filter for structural analysis. Pristine PVP, La_2O_3, and LO/PVP composite materials were analyzed under standard XRD scanning conditions at 40 kV and 15 mA.

SHIMADZU (IRSpirit) Fourier transform infrared (FTIR) spectrometer was used for functional group analysis of pristine PVP, La_2O_3, and the composite material LO/PVP in the wavenumber range of 4000 cm^{-1} to 500 cm^{-1} with a resolution of 4 cm^{-1} and 45 scans per sample at room temperature in ATR mode to identify chemical bonding and functional groups present in the LO/PVP sample.

Surface morphological and chemical composition studies of synthesized LO/PVP composites were performed using a Carl Zeiss (EVO 18) SEM equipped with energy-dispersive X-ray spectroscopy (EDX) with different resolutions.

Impedance spectroscopy measurements were conducted using the impedance analyzer WAYNE KERR to determine the frequency-dependent dielectric response and capacitance in the frequency range of 20 Hz-1 MHz at room temperature.

Results And Discussion

X-ray diffraction

XRD of pristine PVP, La_2O_3, and LO/PVP composites containing LO concentrations of 1% and 5% is displayed in Figure *1*.

The XRD pattern of pristine PVP exhibits broad humps in the range of 10° to 15° and 18° to 24°, confirming the amorphous nature. The XRD pattern does not have any sharp peaks, and the result indicates that long-range crystalline order is absent in it.

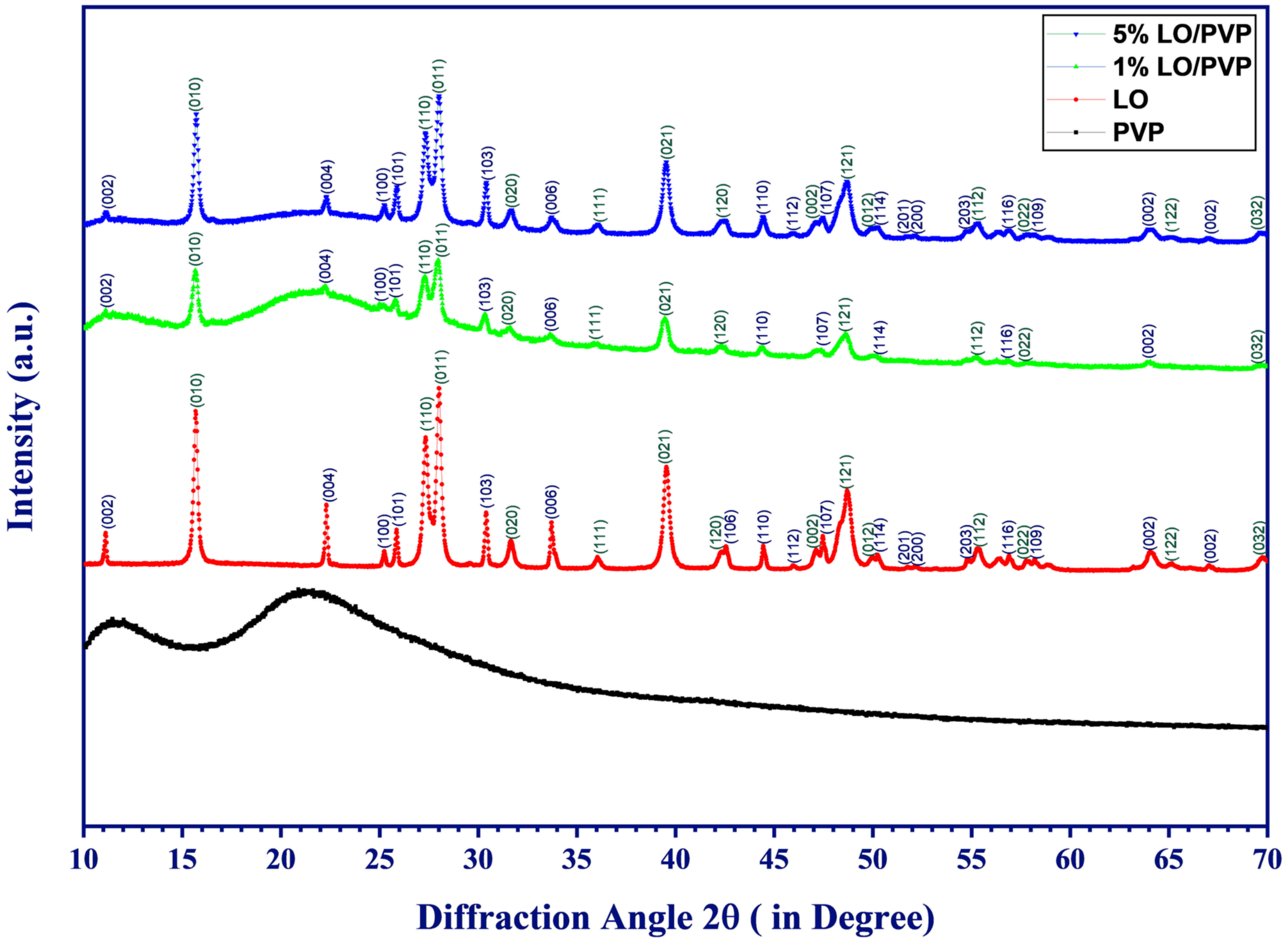

FIGURE 1: XRD Patterns of Pristine PVP, Lanthanum Oxide, $LO_{1\%}$/$PVP_{99\%}$ and $LO_{5\%}$/$PVP_{95\%}$

XRD, X-ray diffraction; PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

The XRD pattern of La_2O_3 has several sharp and intense diffraction peaks, which confirm its highly crystalline nature [10]. Several peaks correspond to $La(OH)_3$ due to moisture, and some peaks correspond to lanthanum dioxycarbonate due to atmospheric CO_2 absorption. Diffraction peaks observed at 15.68° (010); 22.3° (004); 27.34° (110); 28° (011); 30.4° (103); 33.72° (006); 39.54° (021); 48.7° (121); etc., are well matched with standard ICDD reference data (PDF No.: 96-100-0464 and 96-403-1382) [11,12]. Corresponding characteristic peaks are related to well-defined lattice planes and indicate an ordered arrangement of La_2O_3 atoms. Absence of other non-relevant peaks indicates phase purity and confirms the absence of impurities in the La_2O_3 sample. Crystallite size was calculated using the Debye-Scherrer equation, using full width at half maximum, and found to be approximately ~132 nm on average. Lattice parameters calculated by using Bragg's law and the interplanar spacing formula were found to be a = b = 6.51 Å and c = 14.2 Å with a hexagonal crystal structure, and the volume of the unit cell is 521.17 $Å^3$, which is in good agreement with reported standard values.

The XRD pattern of LO/PVP composites exhibits combined structural effects of La_2O_3 and PVP. The pattern exhibits broad humps corresponding to PVP in lower concentration samples of the filler, and crystalline peaks of La_2O_3 appear in the pattern with a gradual increase in peak intensity, sharpness, and minor shifting with increasing filler concentration [13,14]. Gradual increments in peak intensity confirm uniform dispersion of La_2O_3 particles in the PVP matrix. However, a broad hump of the polymer is observed in the XRD pattern of the composites indicates partial amorphous character. No impurity peaks were detected in pristine materials as well as in LO/PVP composite samples, which indicates that La_2O_3 particles were successfully dispersed within the PVP matrix, that no chemical phase transformation occurred during the synthesis process of the composites, and that the composite samples show a semi-crystalline phase, and an increase in filler concentration enhances crystallinity.

Fourier Transform Infrared

FTIR spectra of pristine PVP, La_2O_3, and LO/PVP composites (1-5 mwt%) showing characteristic functional groups are shown in Figure *2*.

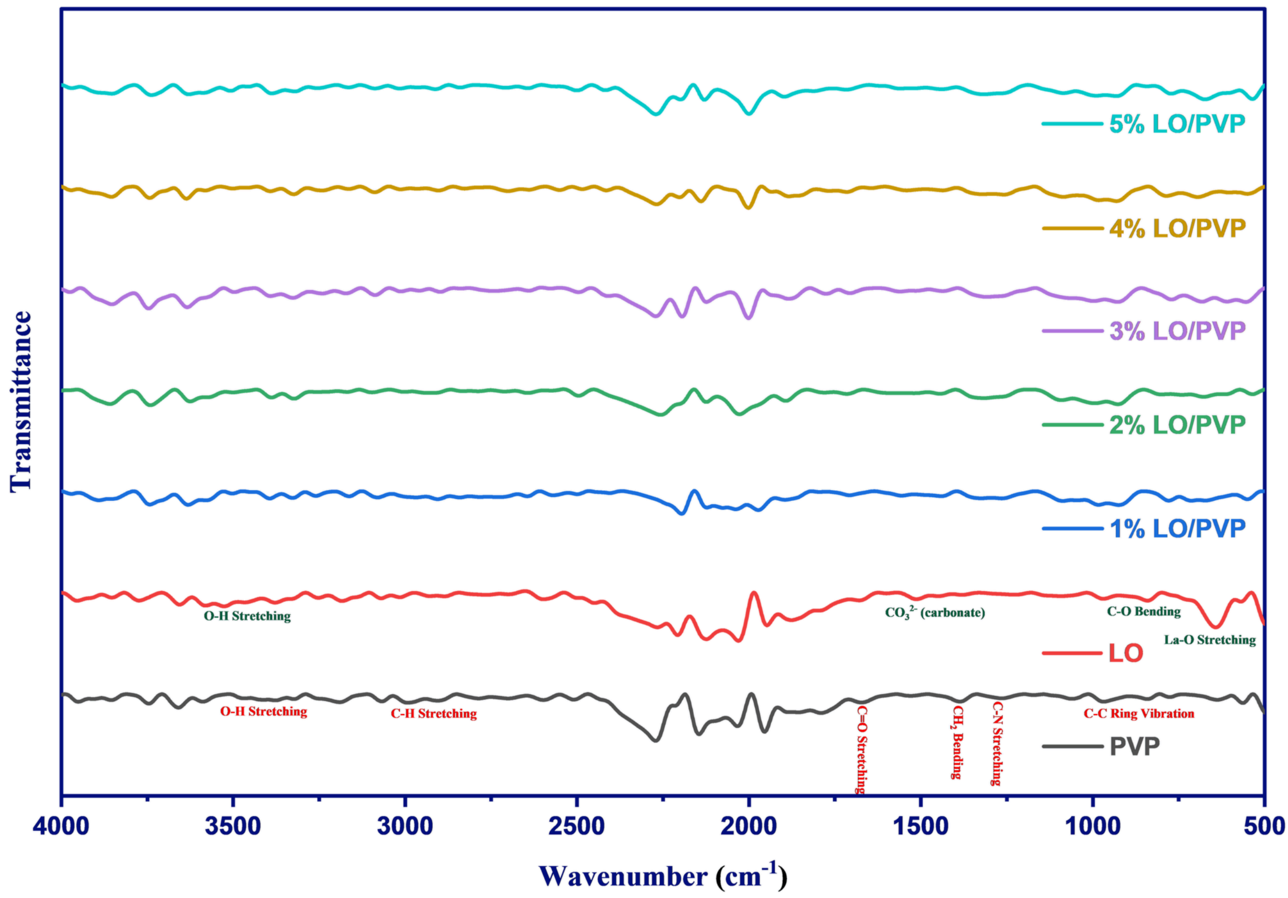

FIGURE 2: FTIR Spectrum of Pristine PVP, Pure Lanthanum Oxide and LO/PVP Composites (1–5 mwt%) with Characteristic Functional Groups

PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

In the FTIR spectrum of pristine PVP, bands observed at ~970 cm^{-1}, ~1270 cm^{-1}, ~1390 cm^{-1}, ~1670 cm^{-1}, ~2990 cm^{-1}, ~3400 cm^{-1} correspond to functional groups C-C ring vibrations, C-N, CH_2, C=O, C-H, O-H, respectively. These peaks correspond to characteristic peaks of PVP reported in previously reported studies. In the FTIR spectrum of La_2O_3, bands observed around ~640 cm^{-1}, ~836 cm^{-1}, ~1515 cm^{-1}, ~3400 cm^{-1} correspond to La-O stretching, C-O bending, (carbonate), and O-H stretching, respectively. Carbonate groups are observed due to atmospheric CO_2 absorption [15]. In the FTIR spectrum of LO/PVP composites, it exhibited corresponding peaks of PVP and La_2O_3, confirming successful incorporation of LO into the PVP matrix. A slight peak shift is observed with increasing LO filler concentration, indicating interfacial interaction. Peak intensity increases with increasing LO filler concentration and minor shifts corresponds to carbonyl and hydroxyl-related absorption bands is observed [16], which indicates interfacial interactions and effective dispersion of LO filler into the PVP matrix. The FTIR observations are consistent with the XRD results, which confirm the coexistence of crystalline La_2O_3 and amorphous PVP without the formation of secondary phases. Moreover, the interfacial interactions inferred from FTIR analysis are expected to contribute to enhanced interfacial polarization, which may be responsible for the observed improvement in dielectric performance of the composites at higher filler concentrations.

Scanning Electron Microscope

Surface morphology of pristine PVP, La_2O_3, and LO/PVP composites containing LO concentrations of 1% and 5% was investigated using SEM, as shown in Figure 3(a-d). SEM micrographs of pristine PVP as shown in Figure 3(a) show irregular, flake-like, and fragmented structures, and the surface is relatively smooth [17]. These micrographs of pristine PVP indicate the amorphous nature of PVP because no well-defined crystalline boundaries appear are observed. SEM micrographs of La_2O_3 shown in Figure 3(b) show fine granular particle structures with highly agglomerated particles. Micrographs show irregular morphology, and a non-uniform size distribution is present. SEM micrographs of $LO_{1\%}/PVP_{99\%}$ (Figure 3(c,d)) composites show the presence of spherical and semi-spherical particles, and LO particles are uniformly distributed in the PVP matrix. These micrographs suggest that excellent dispersion was achieved between LO and PVP [18,19]. Agglomeration is minimal at this low concentration of LO. The observed agglomeration at higher filler concentrations is comparable with previously reported studies. SEM micrographs of $LO_{5\%}/PVP_{95\%}$ composites, as shown in Figure 3(e,f), show higher particle density, and particles become irregular and clustered. Agglomeration is observed at this high concentration of LO [20]. The surface becomes rougher compared to lower filler concentration, indicating enhanced interfacial area.

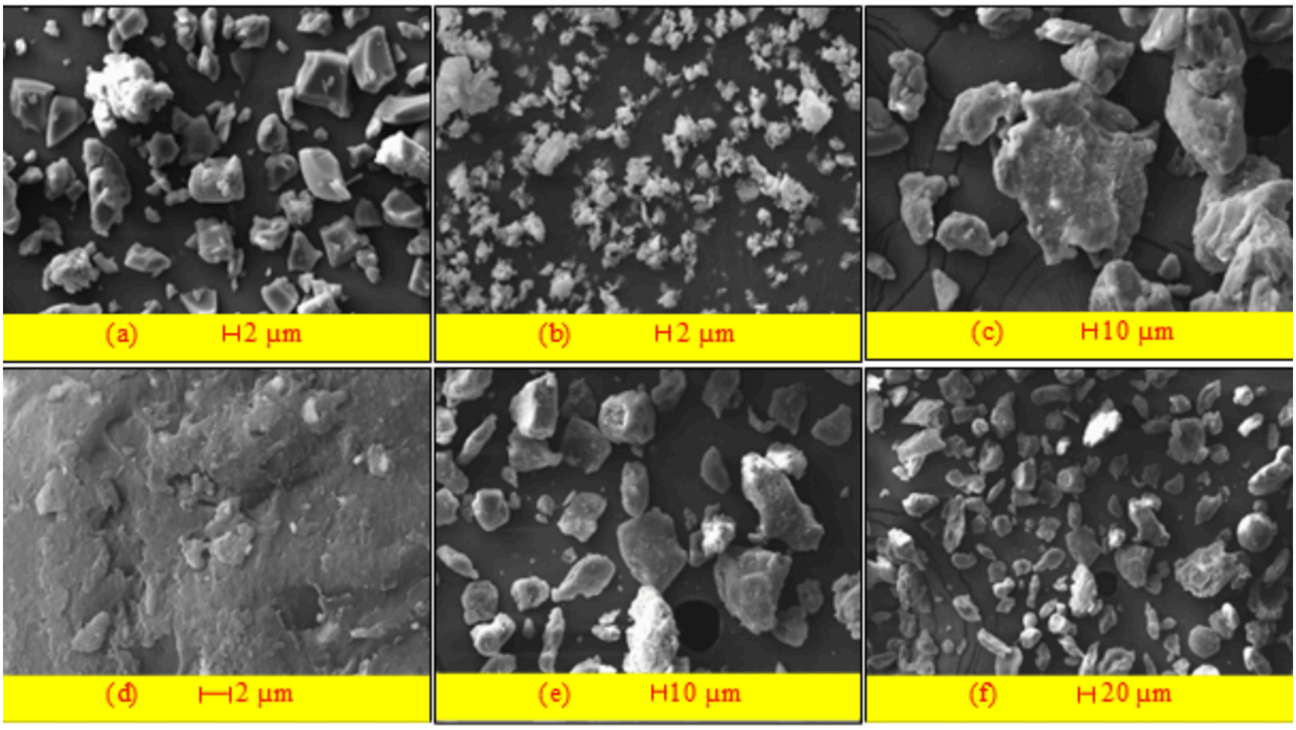

FIGURE 3: SEM Micrographs of (a) Pristine PVP, (b) Pure Lanthanum Oxide, (c,d) $LO_{1\%}/PVP_{99\%}$ and (e,f) $LO_{5\%}/PVP_{95\%}$

LO, lanthanum oxide

Energy dispersive X-ray spectroscopy

In EDX spectra of pristine PVP, Figure 4(a) only has carbon (C) and oxygen (O) peaks present, confirming the purity of the polymer matrix without any other impurities. In the EDX spectrum of pure (Figure 4(b)), the presence of lanthanum (La) and oxygen (O) peaks confirms La_2O_3 without significant impurities; some other minor peaks are secondary peaks associated with lanthanum and oxygen. In the EDX spectrum of LO/PVP composites (Figure 4(c,d)), carbon, oxygen, and lanthanum peaks were observed, so the presence of these elemental peaks indicates uniform distribution and successful incorporation of La_2O_3 filler into the PVP matrix [21, 22]. In this spectrum, it is noted that lanthanum peak intensity increases as filler concentration increase in PVP. An additional peak is observed around 2 KeV that is due to the gold coating used during SEM characterization.

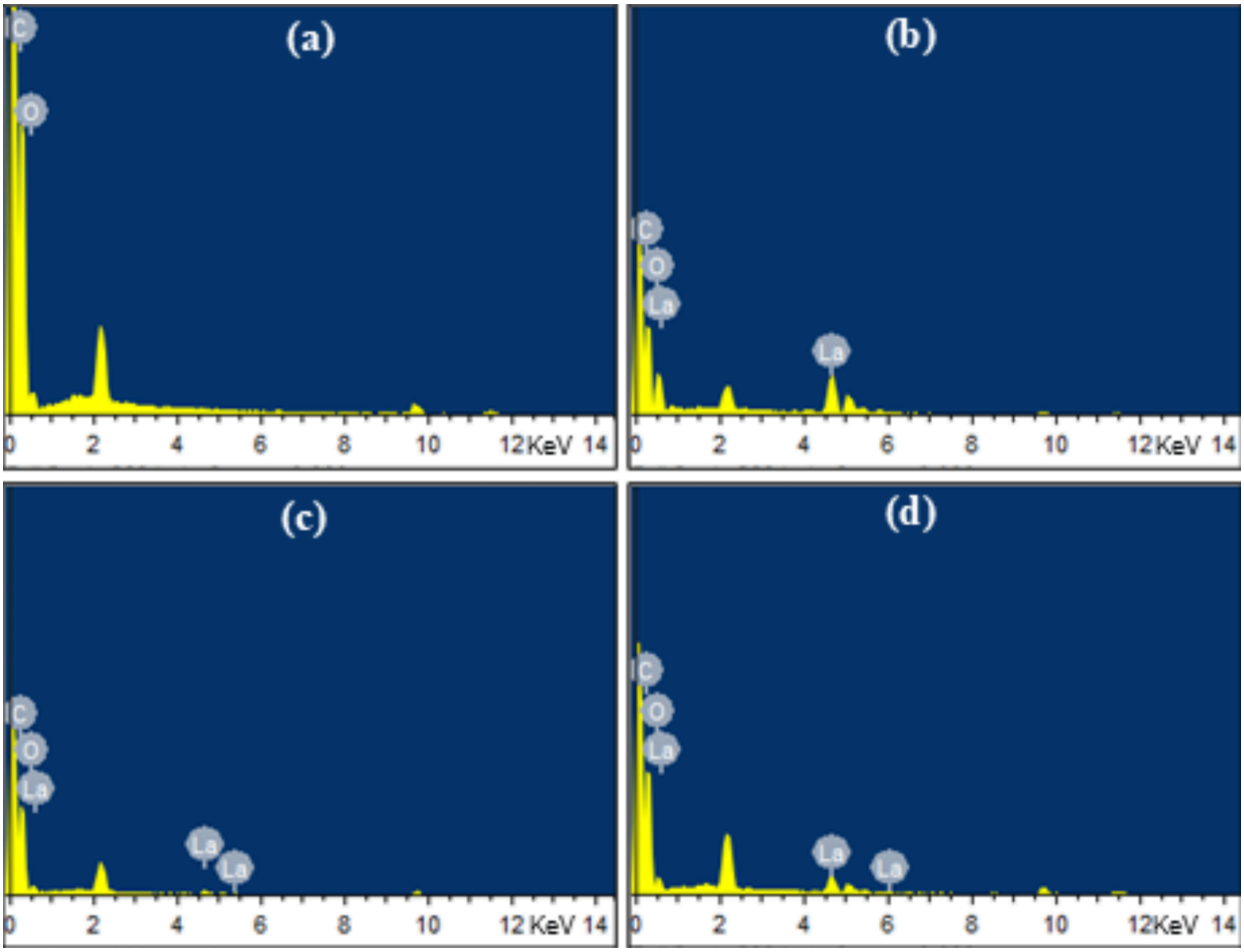

FIGURE 4: EDX Spectrums of (a) Pristine PVP, (b) Pure Lanthanum Oxide, (c) $LO_{1\%}/PVP_{99\%}$ and (d) $LO_{5\%}/PVP_{95\%}$

EDX, energy-dispersive X-ray spectroscopy; PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

Impendence spectroscopy

Dielectric Constant

Figure 5 shows dielectric constant versus frequency response in different frequency ranges for pristine PVP and LO/PVP composites with varying LO filler concentrations (1-5 mwt%). In low frequency regions (log f ~ 1.3 to 2.5), the dielectric constant is high due to dominance of Maxwell-Wagner interfacial polarization, which happens due to accumulation of charge carriers at LO/PVP interfaces [23]. In the intermediate frequency region (log f ~ 2.5 to 5), a rapid drop is observed

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due to dipolar polarization, where dipoles attempt to align with alternating electric fields but fail to follow. In the high-frequency region (log f ~ 5 to 8), dielectric constant curves exhibit nearly constant values, i.e., form a plateau region. In this region only electronic and ionic polarization play a significant role; dipolar and interfacial polarization become ineffective. Because electronic polarization occurs so rapidly that it can follow an electric field even at high frequencies, behavior becomes frequency independent [24].

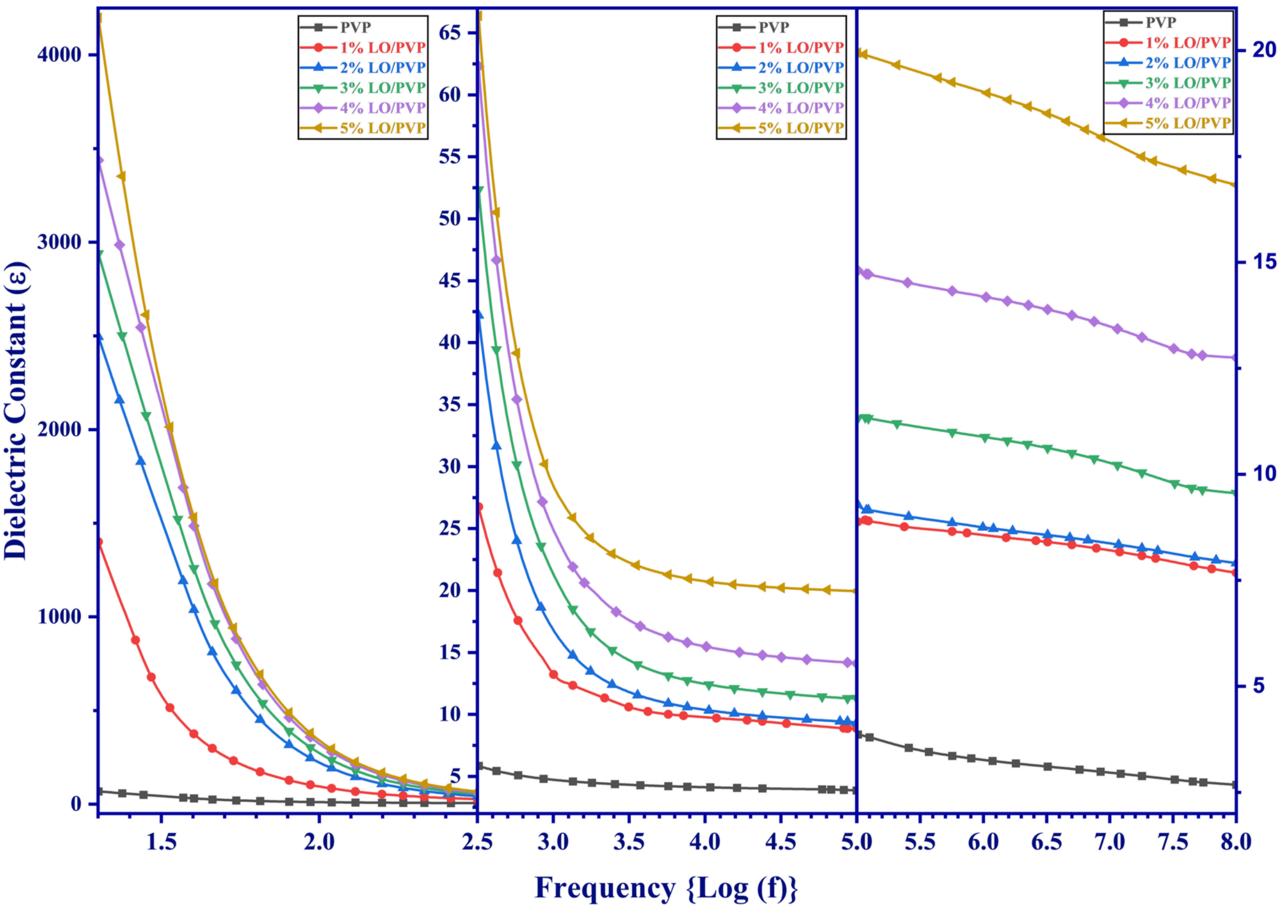

FIGURE 5: Dielectric Constant Versus Frequency Response

PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

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Sample Name	Frequency						
	100 Hz	1 KHz	10 KHz	100 KHz	1 MHz	10 MHz	100 MHz
Pristine PVP	10.99	4.73	4.13	3.87	3.26	2.97	2.68
$LO_{1\%}/PVP_{99\%}$	92.99	13.22	9.77	8.88	8.57	8.21	7.68
$LO_{2\%}/PVP_{98\%}$	217.70	16.77	10.37	9.28	8.75	8.37	7.91
$LO_{3\%}/PVP_{97\%}$	267.60	21.26	12.46	11.32	10.89	10.27	9.56
$LO_{4\%}/PVP_{96\%}$	316.37	24.84	15.48	14.81	13.02	13.49	12.76
$LO_{5\%}/PVP_{95\%}$	333.75	28.38	20.75	19.94	19.02	17.86	16.83

TABLE 1: Dielectric Constant Versus Frequency Response of Pristine PVP and LO/PVP Composites (1-5 mwt%)

PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

According to Table 1 and observations, a gradual increase in dielectric constant values indicates the formation of heterogeneous systems. The dielectric constant increases with increasing LO filler concentration, and the highest dielectric constant value was achieved at $LO_{5\%}/PVP_{95\%}$ due to the Maxwell-Wagner interfacial polarization effect because charges accumulate on heterogeneous interfaces and conductivity also increases at higher concentrations, which supports polarization [25]. Obtained dielectric constant values are comparable with previously reported studies of metal oxide-reinforced polymer composites. Incorporation of LO filler into the PVP matrix significantly enhances dielectric properties of PVP, which makes it suitable for dielectric applications and energy storage applications.

Dissipation Factor

Figure 6 shows frequency dependence of the dissipation factor in different frequency regions for pristine PVP and LO/PVP composites with different filler concentrations. In low-frequency regions (log f ~ 1.3 to 2.5), the dissipation factor shows a high value due to Maxwell-Wagner interfacial polarization, space charge accumulation at interfaces, and electrode polarization effects because charge carriers have sufficient time to accumulate at interfaces [26]. In the intermediate frequency region (log f ~ 2.5 to 5), the dissipation factor drops sharply because dipoles and charge carriers fail to respond synchronously with alternating electric fields and energy dissipation. In high-frequency regions (log f ~ 5 to 8), a slight increase in dissipation factor is observed, which may possibly be due to electronic polarization and conduction losses. The dissipation factor decreases as frequency increases in the low-frequency region and attains a minimum value in the intermediate region, and then slightly increases in the high-frequency region, which is indicating increased conduction and intrinsic polarization losses [27]. Increasing LO filler concentration enhances dielectric losses because of increased interfacial polarization effects and facilitates charge carrier mobility; for example, at 1 MHz, the dissipation factor value for pristine PVP is only 0.03, while for $LO_{5\%}/PVP_{95\%}$, it increases to 0.05 (as shown in Table 2). Observed interfacial polarization effects have been reported in previous studies of metal oxide-incorporated polymer composites.

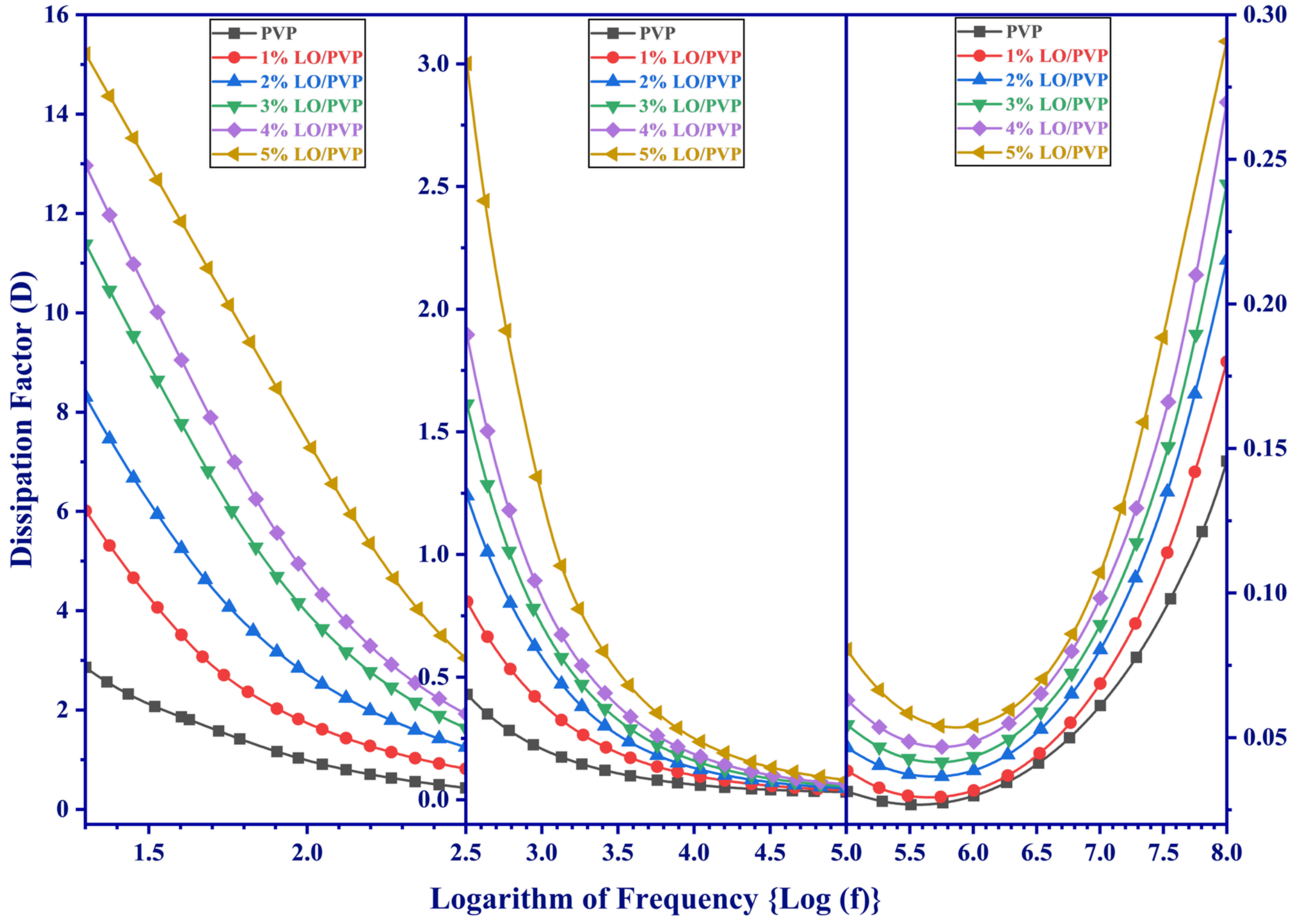

FIGURE 6: Dissipation Factor Versus Frequency Response

PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

Sample Name	Frequency						
	100 Hz	1 KHz	10 KHz	100 KHz	1 MHz	10 MHz	100 MHz
Pristine PVP	0.98	0.21	0.06	0.03	0.03	0.06	0.15
$LO_{1\%}/PVP_{99\%}$	1.72	0.39	0.10	0.04	0.03	0.07	0.18
$LO_{2\%}/PVP_{98\%}$	2.70	0.58	0.13	0.05	0.04	0.08	0.22
$LO_{3\%}/PVP_{97\%}$	3.92	0.71	0.16	0.05	0.04	0.09	0.24
$LO_{4\%}/PVP_{96\%}$	4.66	0.82	0.19	0.06	0.05	0.10	0.27
$LO_{5\%}/PVP_{95\%}$	7.37	1.23	0.25	0.08	0.05	0.11	0.29

TABLE 2: Dissipation Factor Versus Frequency Response of Pristine PVP and LO/PVP Composites (1-5 mwt%)

PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

Quality Factor

Figure *7* shows the frequency dependence of a quality factor for pristine PVP and LO/PVP composites with different filler concentrations. At low frequencies, the quality factor remains very low due to intense dielectric losses. At intermediate frequencies, the quality factor exhibits a relaxation-controlled peak, and energy storage efficiency is maximized with minimum dielectric losses. At high frequencies, after the peak, quality factor decreases monotonically due to dominance of electronic and ionic polarization with increased conduction losses [28]. Quality factor curves show peak behavior with increasing frequencies, which agrees well with previous studies. The maximum value of the quality factor is observed at intermediate frequencies. The graph shows that increasing LO filler concentration in LO/PVP composites reduced the quality factor due to higher energy losses and values of quality factor at different frequencies for all samples are presented in Table *3*.

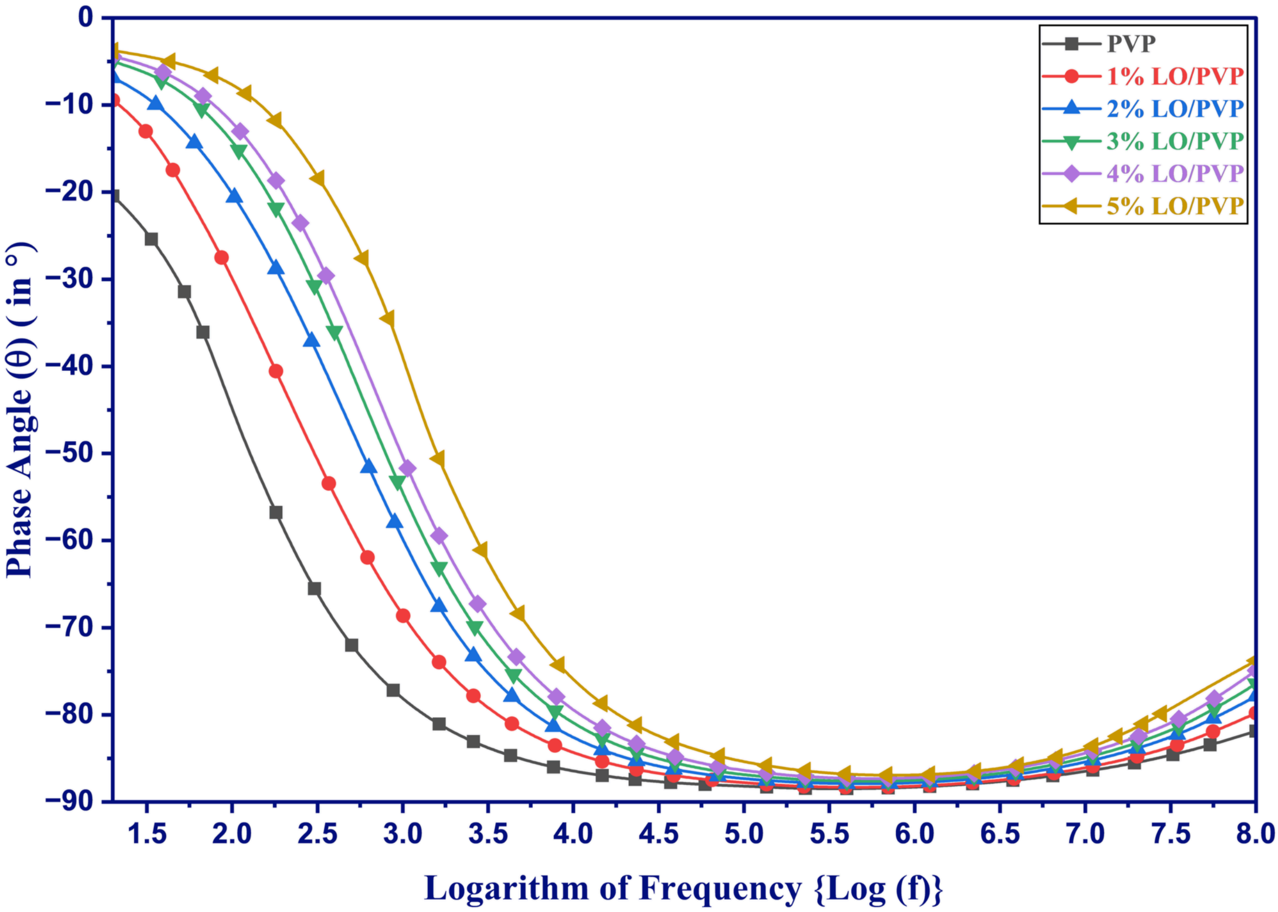

FIGURE 7: Quality Factor Versus Frequency Response

PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

Sample Name	Frequency						
	100 Hz	1 KHz	10 KHz	100 KHz	1 MHz	10 MHz	100 MHz
Pristine PVP	1.210	5.417	17.083	31.895	28.809	14.956	6.814
$LO_{1\%}/PVP_{99\%}$	0.318	1.812	8.085	22.686	27.488	13.212	4.953
$LO_{2\%}/PVP_{98\%}$	0.280	1.549	6.898	20.362	26.344	13.278	5.123
$LO_{3\%}/PVP_{97\%}$	0.257	1.459	6.438	19.179	25.006	12.369	4.524
$LO_{4\%}/PVP_{96\%}$	0.198	1.064	4.912	15.133	22.543	11.857	4.763
$LO_{5\%}/PVP_{95\%}$	0.185	0.991	4.371	13.300	20.240	11.300	4.411

TABLE 3: Quality Factor Versus Frequency Response of Pristine PVP and LO/PVP Composites (1-5 mwt%)

PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

Phase Angle

Figure *8* shows frequency dependence of phase angle in different frequency regions for pristine PVP and LO/PVP composites with different filler concentrations. In the low-frequency region (log f ~ 1.3 to 2.5), values of phase angle are less negative and gradually decrease with increasing frequency. In this region, material exhibits more resistive behavior due to charge accumulation, interfacial polarization, and high dielectric losses. In the intermediate frequency region (log f ~ 2.5 to 5), the phase angle values drop sharply and approach -90°, indicating a dominant capacitive nature as dipoles effectively follow the alternating electric field. Energy storage efficiency is high in this region. In the high-frequency region (log f ~ 5 to 8), a slight increase in phase angle values is observed because it fails to follow rapidly changing alternating electric fields, leading to an increase in dielectric losses that causes a deviation from ideal capacitive behavior. In this overall phase angle analysis, it can be observed that LO/PVP composites transition from resistive to capacitive behavior with increasing frequency. In the intermediary frequency region, the maximum capacitive behavior is observed and corresponding values are presented in Table *4*. With increasing LO filler concentration, the phase angle has a less negative value in comparison to pristine PVP and lower LO filler concentration composites [29], which is indicating enhanced dielectric relaxation behavior due to an increase in charge carrier concentration at LO/PVP interfaces.

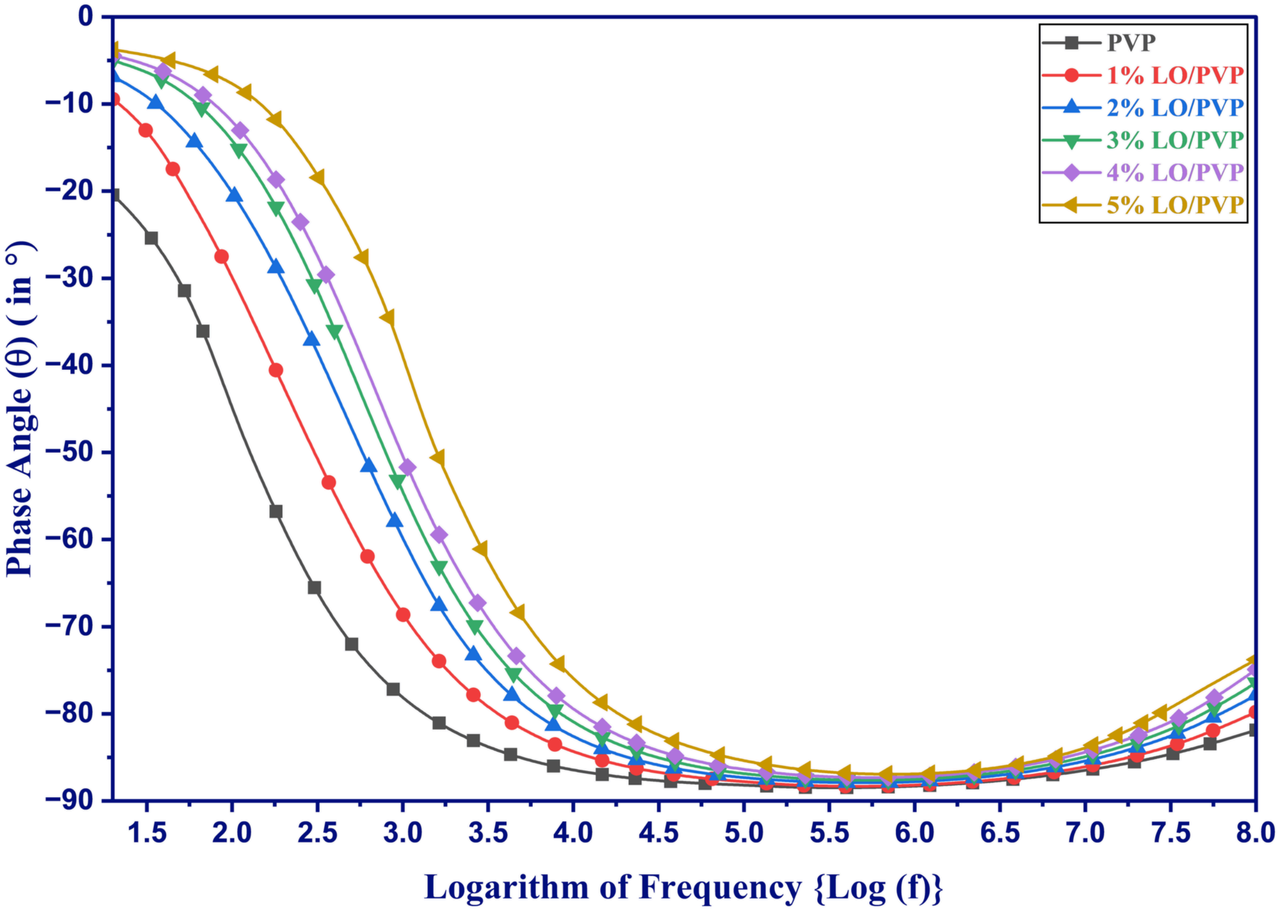

FIGURE 8: Phase Angle Versus Frequency Response

PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

Sample Name	Frequency						
	100 Hz	1 KHz	10 KHz	100 KHz	1 MHz	10 MHz	100 MHz
Pristine PVP	-47.95	-78.88	-86.58	-88.24	-88.02	-86.10	-82.06
$LO_{1\%}/PVP_{99\%}$	-27.73	-65.71	-83.45	-87.43	-87.35	-84.30	-79.72
$LO_{2\%}/PVP_{98\%}$	-19.58	-62.10	-83.10	-87.52	-87.95	-85.72	-78.74
$LO_{3\%}/PVP_{97\%}$	-15.87	-56.67	-81.45	-87.07	-87.74	-85.45	-77.73
$LO_{4\%}/PVP_{96\%}$	-13.52	-50.75	-77.13	-84.87	-87.56	-85.31	-78.11
$LO_{5\%}/PVP_{95\%}$	-14.60	-55.54	-81.30	-87.08	-87.79	-85.64	-78.72

TABLE 4: Phase Angle (in degree) Versus Frequency Response of Pristine PVP and LO/PVP Composites (1-5 mwt%)

PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

Impedance

Figure *9* shows frequency dependence of impedance in different frequency regions for pristine PVP and LO/PVP composites with different filler concentrations. In low-frequency regions (log f ~ 1.3 to 2.5), impedance is observed to be high due to charge accumulation at interfaces and electrode polarization, indicating the material behaves resistively or has resistive behavior. In the intermediate frequency region (log f ~ 2.5 to 5), impedance drops sharply due to dipolar response to the electric field and increased charge mobility. In this region, resistance decreases, and a resistive nature transitions to a capacitive nature. In the high-frequency region (log f ~ 5 to 8), impedance values become very low. This behavior is indicating enhanced charge mobility and reduced resistive effects. In this overall impedance analysis, it is noted that the impedance of LO/PVP composites decreases as frequency increases and transitions from highly resistive behavior at low frequencies to capacitive behavior at high frequencies and values of impedance at different frequencies for all samples are presented in Table *5* [30]. An increase in LO filler concentration enhances charge mobility and reduces impedance, which confirms improved conductivity due to increased charge carrier concentration.

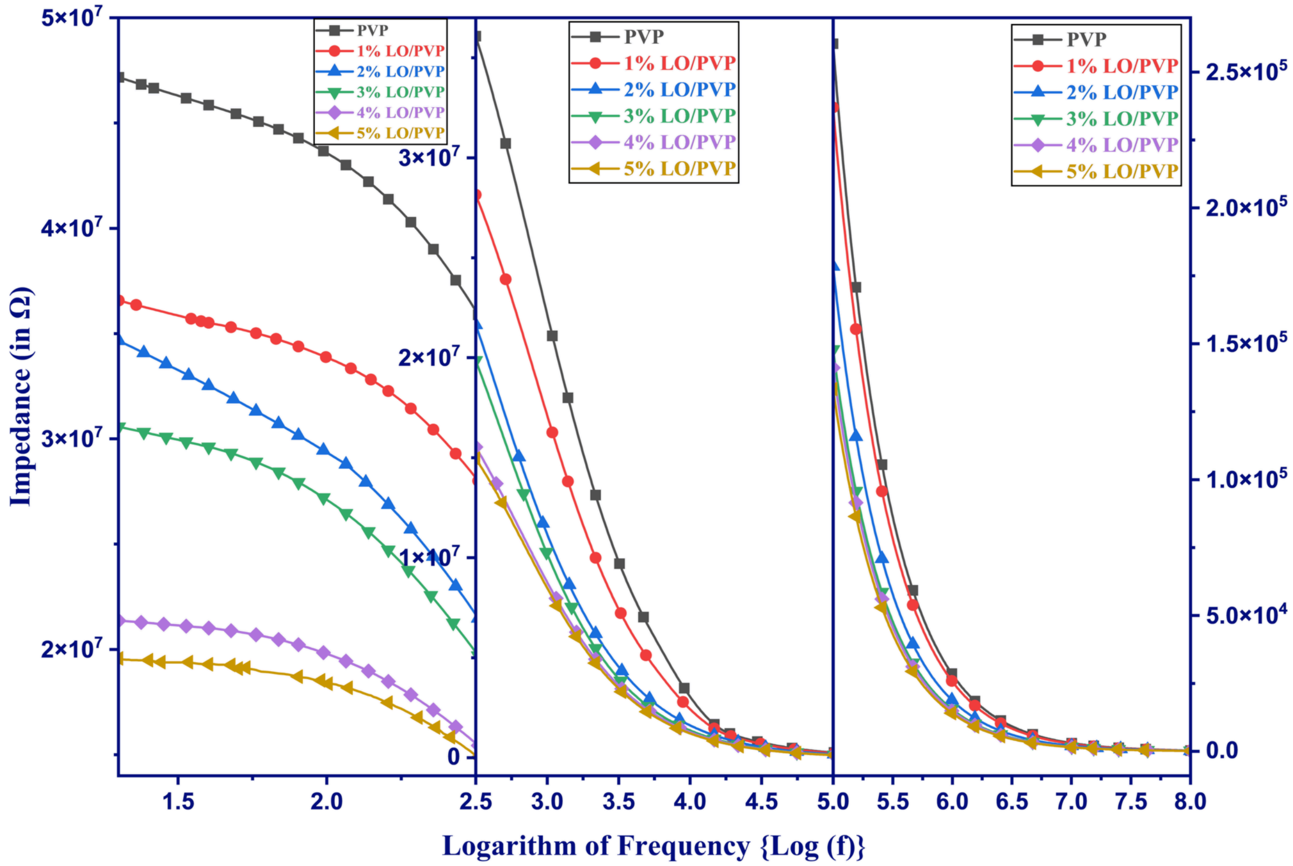

FIGURE 9: Impedance Versus Frequency Response

PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

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Sample Name	Frequency						
	100 Hz	1 KHz	10 KHz	100 KHz	1 MHz	10 MHz	100 MHz
Pristine PVP	4.35E+07	2.21E+07	3037447	255757	28018.86	3001.958	252.7416
$LO_{1\%}/PVP_{99\%}$	3.38E+07	1.70E+07	2419361	232732.6	25405.12	2723.972	244.8702
$LO_{2\%}/PVP_{98\%}$	2.93E+07	1.11E+07	1613103	175177.9	18516.11	2024.704	201.0883
$LO_{3\%}/PVP_{97\%}$	2.71E+07	1.01E+07	1361468	145127.8	15231.91	1623.138	146.0438
$LO_{4\%}/PVP_{96\%}$	1.98E+07	8729951	1277763	138519.3	14619.13	1563.749	134.9616
$LO_{5\%}/PVP_{95\%}$	1.84E+07	8451300	1213416	130851.2	13794.33	1475.014	105.3559

TABLE 5: Impedance (in Ω) versus Frequency Response of Pristine PVP and LO/PVP Composites (1-5 mwt%)

PVP, polyvinyl pyrrolidone; LO, lanthanum oxide

Conclusions

In energy storage devices, the search for advanced materials with high dielectric constant and low dissipation factor for energy storage applications has led to the synthesis of La_2O_3/PVP polymer composites using a conventional ball-milling technique. From XRD analysis, the amorphous nature of PVP and the hexagonal crystal structure of La_2O_3, with lattice parameters a = b = 6.51 Å and c = 14.2 Å, were observed. Using the Debye-Scherrer equation, the average crystallite size of the composites was found to be 132 nm. In composites, a broad hump of polymer and peaks of filler indicate successful composite formation. FTIR spectroscopy was used to analyze bonding interactions in the synthesized composites. Surface morphology was analyzed using SEM. The rough surface of the composites was found to increase the interfacial area and help in trapping charge carriers, which is beneficial for energy storage. From the EDX spectrum, in the study of elements, peaks of PVP and La_2O_3 elements, namely carbon (C), oxygen (O), and lanthanum (La), are obtained. Using impedance spectroscopy, dielectric permittivity, dissipation factor, quality factor, phase angle, and impedance were investigated over a broad frequency range. In composites, with increasing filler concentration, the value of permittivity is found to increase. In comparison to pristine PVP, the dielectric constant of the 5% filler concentration composites is found to increase up to approximately six times. At a frequency of 1 MHz, the dissipation factor for pristine PVP is 0.03, while for 5% LO/PVP composites it is found to be 0.05. The obtained results are consistent with previously reported studies of ceramic-polymer composites. In the future, these polymer composites may prove to be emerging materials for electrodes in energy storage devices.

Additional Information

Author Contributions

All authors have reviewed the final version to be published and agreed to be accountable for all aspects of the work.

Concept and design: Jagdish P. Yadav, Jitendra K. Mishra, Dinesh K. Yadav, Anju Yadav, Suchitra Yadav, Balram Tripathi

Acquisition, analysis, or interpretation of data: Jagdish P. Yadav, Jitendra K. Mishra, Dinesh K. Yadav, Anju Yadav, Suchitra Yadav, Vikas Kumawat, Balram Tripathi

Drafting of the manuscript: Jagdish P. Yadav, Jitendra K. Mishra, Dinesh K. Yadav

Critical review of the manuscript for important intellectual content: Jitendra K. Mishra, Dinesh K. Yadav, Anju Yadav, Suchitra Yadav, Vikas Kumawat, Balram Tripathi

Supervision: Dinesh K. Yadav

Disclosures

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Data Availability Statements

The datasets (and/or code) supporting this study are available from the corresponding author upon reasonable request.

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