Electron Beam-Induced Modifications in Dielectric and AC Electrical Properties of Gelatin-Acrylic Acid Blends

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Abstract
The main aim of this work is to investigate the dielectric and electrical properties of synthesized acrylic acid–gelatin blends of different acrylic acid weight concentrations with gelatin. An electron beam (EB) with different doses of 50–150 kGy irradiated the synthesized blend while it was also subjected to an applied electric field in the range of 70 Hz–5 MHz to determine the variation in the dielectric parameters and electrical conductivity. The lowest kHz gives the highest dielectric parameters and lowest AC electrical conductivity ($\sigma_{ac}$) values for both the blank and EB-irradiated blends. The lowest dielectric parameters and $\sigma_{ac}$ values were at 0.4 g acrylic acid concentration, and the highest values were for the 0.2 g acrylic acid concentration. Comparing the different EB doses for each acrylic acid concentration, it was found that the order of the lowest dielectric constant, $\varepsilon$, was 0.579 for (0.2 g + 100 kGy) > 0.475 for (0.6 g + 50 kGy) > 0.412 for (1 g + 50 kGy) > 0.315 for (0.4 g + 100 kGy) at 0.5 kHz. However, the order of the highest $\sigma_{ac}$ was $1.71 \times 10^{-4}$ S/m for (0.2 g + 150 kGy) > $1.39 \times 10^{-4}$ S/m for (0.6 g + 75 kGy) > $1.08 \times 10^{-4}$ S/m for (1 g + 75 kGy) > $8.56 \times 10^{-5}$ S/m for (0.4 g + 150 kGy) at 5 MHz. A modified equation was deduced for the AC conductivity as a function of the frequency of the applied field and the concentration of acrylic acid mixed with gelatin at room temperature.

Keywords Electron irradiation · electron accelerator · dielectric permittivity · AC conductivity · gelatin–acrylic acid blend

Introduction
The treatment of different materials and products by exposure to ionizing radiation can change their chemical, physical, and/or biological characteristics. This ionizing process minimizes the effect of materials on the environment or increases their utility in many applications. Radiation is divided into non-ionizing and ionizing. Ionizing radiation is defined as radiation with sufficient energy to separate the electrons of atoms and hence convert them into charged ions. It is classified into $\alpha$-particles, $\beta$-particles, X-rays, $\gamma$-rays, accelerated electrons and ions beam. Non-ionizing radiation is electromagnetic (EM) radiation with no sufficient energy to produce ionization. The accelerated electrons are produced by particle accelerators and have different applications such as material surface curing, wire and cable cross-linking, heat-shrinkable materials, and sterilization of food and medical products. The ionization and excitation of electrons in an atom are two principles for chemical reactions due to the radiation effect. Therefore, atoms displaced from their original sites to adjacent lattice sites become loosely bonded to some trapping centers or free as the chain scission (main-chain degradation, free radical formation) or the cross-linking causing the degradation occurs. Electron beam (EB) irradiation is a photochemical process that improves the physical properties of several polymers, such as structural, thermal, electrical, and dielectric properties. It not only cancels the chemical structure of the polymer but also ameliorates the presence of trapped charges in the polymer matrix (creating defects). The modification in the polymer properties is mainly dependent on the irradiation dose rate and polymer characteristics. The
absorbed dose is defined as the ionizing energy delivered per unit mass of material with the international gray (Gy) unit (as the absorption of (J/kg)). In general, EB irradiation is a quick and efficient method for producing modified polymers that are widely used in many industrial applications, such as electrical cables to extend their life and make them more resistant to dangerous solvents and fire, electronic devices, smart glass, and so on.\textsuperscript{8,14–16}

Natural gelatin is a natural multipurpose biomaterial that is obtained through partial alkaline/acid hydrolysis of the insoluble fibrous collagen that exists in animal tissues.\textsuperscript{17–19} It is widely used in the industrial and pharmaceutical fields such as phantom applications, wound dressing, tissue engineering, drug delivery, gene delivery, food packing materials, and health care.\textsuperscript{20–32} In electrochemical biosensors, various materials are used as suitable matrices for physical entrapment, such as cellulose, polyvinyl alcohol, gelatin, chitosan, and others.\textsuperscript{33–38} Gelatin is used as an electrode substrate for biosensor systems as it is an excellent biocompatible and biodegradable polymer.\textsuperscript{39,40} Also, it acts as a matrix for implants, stabilizers in vaccines, and targeted drug delivery systems and enables the design of several different drug carrier systems.\textsuperscript{41–44} Acrylic acid is a compound that can be used in different applications such as the water treatment industry, textile industry, personal care products, and micro-sensors.\textsuperscript{45–47} Previous reports studies on glucose electrochemical biosensors, chitosan-based electrochemical sensors and biosensors, bio sensor properties of glucose oxidase immobilized within SiO\textsubscript{2} gels, development of a glucose biosensor using gelatin and gelatin-polyacrylamide supporting systems, an alginate immobilization matrix in the construction of an amperometric biosensor, electrochemical study of gelatin as a matrix for the immobilization of a horse heart, and electrochemical glucose biosensing by pyranose oxidase immobilized in a gold nanoparticle-polyaniline/AgCl/gelatin nanocomposite matrix. With respect to acrylic acid, they include a selection of acrylate esters and/or 1-vinyl imidazole, polyaniline/poly(acrylonitrile-co-acrylic acid) composite film, a poly(acrylonitrile-co-acrylic acid) film-based glucose biosensor, and acrylic acid onto polytetrafluoroethylene films for glucose oxidase immobilization.\textsuperscript{36–52} There are few studies in the literature that used gelatin as an electrochemical biosensor and acrylic acid.\textsuperscript{53–61} Moreover, there is no literature reporting on the acrylic acid–gelatin blends used in electrochemical biosensors.

Previously, it was deduced that an acrylic acid–polymer blend can improve the electrical behaviour of the polymer itself to form a modified polymer suitable for micro-sensor applications.\textsuperscript{62} Recently, one requirement was noted for the materials that can be used in flexible electronic devices, which is that they have a low dielectric value with better performance and efficiency, such as polymers with excellent leakage current.\textsuperscript{63} We studied the electrical and dielectric properties of the synthesized blends formed from the addition of acrylic acid with different concentrations to the constant concentration of gelatin to be suitable for flexible electronics such as biosensors. Moreover, we use the different concentrations of acrylic acid to obtain the lowest dielectric value for gelatin–acrylic acid blends. Also, the effect of EB irradiation with different doses on these properties of the blends was studied. Finally, a modified equation of AC electrical conductivity as a function of the frequency of the applied field (in the kHz/MHz range) and the concentration of acrylic acid mixed with gelatin at room temperature only was deduced.

Many studies have been directed towards the improvement of different polymers with high and low dielectric constants.\textsuperscript{8,64,65} Polymers with a high dielectric constant can be used as artificial muscles and film capacitors, while those with a low dielectric constant can be used in the capacitor and dielectric fields.

When comparing the obtained results to the literature, we did not find any results related to acrylic acid–gelatin blends.\textsuperscript{48–51} By adding acrylic acid to gelatin to form a blend, the electrical properties of gelatin are improved. Also, the ratio between gelatin and acrylic acid to form a blend is not found. Hence, we synthesized the different weight concentrations of acrylic acid of 0.2 g, 0.4 g, 0.6 g, and 1 g with 1 g gelatin. The aim of using different weight concentrations of acrylic acid is to improve the electrical properties of the formed blends.

**Experimental Details**

**Preparation of Gelatin–Acrylic Acid Blends**

The gelatin solution was prepared by swelling 1 g of gelatin powder in 50 ml of distilled water with continuous stirring and heating at 50°C for 30 min. Acrylic acid of different weight concentrations equal to 0.2 g, 0.4 g, 0.6 g, and 1 g was dissolved in 20 ml of distilled water for each concentration. Then a 1 g gelatin solution was mixed with an acrylic acid concentration (e.g., 0.2 g) to form the gelatin–acrylic acid blend. This step was repeated for each acrylic acid concentration. Each mixture was stirred for 1 h in order to solubilize, then cast on a clean Petri dish and left for 48 h to dry. Finally, the formed blend was cut into films with dimensions of 1 cm × 1 cm to be suitable for EB irradiation at the National Center for Research and Radiation Technology, Egyptian Atomic Energy Authority.

**Irradiation Source**

A Vivirad (France) industrial linear electron accelerator of 3 MeV/3 mA EB was used to irradiate the films in the air.
The minimum dose was 3.0 kGy with a conveyor speed of 16 m/min and scanning area of 8 cm × 70 cm. The operating conditions of the gelatin–acrylic acid blends were 25 kGy dose, 10 mA current, 5.59 m/min conveyor speed, and 2.7 MeV of EB energy. The EB irradiation doses used were 50 kGy, 75 kGy, 100 kGy, and 150 kGy for all acrylic acid–gelatin blends.

**Dielectric and AC Electrical Property Measurement**

The dielectric permittivity and AC electrical conductivity of the films were measured using a computer-controlled precision impedance analyzer (Model EUCOL-U2826) LCR meter over a frequency range up to 5 MHz at room temperature in the Cyclotron Facility of the Nuclear Research Center, Egyptian Atomic Energy Authority.

The dielectric study determines several properties such as the dielectric storage or constant (\(\varepsilon\)), dielectric loss (\(\varepsilon''\)), dissipation factor (\(\tan \delta\)) and the AC electrical conductivity (\(\sigma_{ac}\)) over a range of frequencies. The two important factors for determining the polymer's suitability as an electrolyte for energy storage applications are \(\varepsilon\) and \(\varepsilon''\). The dielectric response of any material is characterized by two parameters: the real (\(\varepsilon\)) and imaginary (\(\varepsilon''\)) parts of the complex dielectric permittivity (\(\varepsilon^*\)). The complex dielectric permittivity characterizes the electrical polarization degree of a material that experiences the effect of an externally applied electric field. The dielectric permittivity, \(\varepsilon\) in general, is independent of electric field strength below a certain critical level or above which carrier injection into the material is essential.\(^6\) Otherwise, it depends strongly on the frequency of the applied AC electric field (the time rate of change of the varying field). When an alternating field of a given frequency is incident on a dielectric material, it will take some time to polarize. This time changes with respect to the incident field's frequency, and the time difference can be represented by a phase term, \(\delta\), called the loss angle. \(\varepsilon\) measures dipole polarization and alignment and was calculated from capacitance (computer-measured values) using the following equations:\(^66,67\)

\[
\varepsilon' = \frac{Cd}{\epsilon_r \epsilon_0 A}
\]

\[\varepsilon'' = \frac{Gd}{\epsilon_r \omega \epsilon_0 A} \tag{2}\]

where \(\epsilon_r\) is the permittivity of free space \((8.85 \times 10^{-12} \text{ F/m})\), \(C\) and \(G\) are the capacitance and conductance, respectively, and \(d\) and \(A\) are the thickness and cross-section area of the sample, respectively. Hence, the dissipation factor or loss tangent, \(\tan (\delta)\), can be given by the relation\(^68\)

\[
\tan \delta = \frac{\varepsilon''}{\varepsilon'}
\]

The dissipation factor explains the amount of energy which is dispersed in the material during the application of an electric field and describes the ratio between the total ohmic and capacitive reactance.\(^69,70\) \(\varepsilon''\) illustrates the required energy necessary to align the dipoles and measures the energy loss through heat of the dielectric material due to the applied electric field.\(^70\) The AC electrical conductivity (\(\sigma_{ac}\)) can be given by Eq. 4

\[
\sigma_{ac} = \omega \varepsilon_0 \varepsilon'' = \omega \varepsilon_0 \varepsilon' \tan \delta
\]

The AC electrical conductivity is a tool for determining hopping dynamics for ions and providing information about transport in materials. It depends on the material conductor length, cross-section area, and temperature. Here, \(\varepsilon\), \(\varepsilon''\), and \(\sigma_{ac}\) of the blends have been measured by impedance spectroscopy through the application of a small AC signal across the sample that is sandwiched between two copper electrodes under spring pressure. The impedance parameters were measured with a computer-controlled impedance analyzer.

**Results and Discussion**

**Dielectric Studies and Dependence on Frequency**

**Dielectric Constant**

Figure 1a–d shows the \(\varepsilon\) dependence on the frequency of the applied AC electric field in the range from 70 Hz to 900 Hz for the gelatin blended with different weight concentrations (0.2 g, 0.4 g, 0.6 g, and 1 g) of acrylic acid films irradiated by a 50 kGy EB dose and compared with the blank ones, respectively. The decrease in \(\varepsilon\) values with increasing frequency is due to the inability of induced dipoles in the blends to change the direction of orientation with increased applied field. At low frequencies, there are high values of \(\varepsilon\) because of the charges accumulated at the interface between the film and the electrodes (space charge polarization)\(^8\). Also, it is not related to the hopping dynamics of the mobile ions because the polarization process occurs rapidly.\(^7\) It is recognized that the first appearance of a dielectric in an electric field causes its polarization. Moreover, it was clear that \(\varepsilon\) of blank blends with 0.2 g, 0.6 g, and 1 g acrylic acid was higher than that of the blends irradiated by a 50-kGy EB dose, except for the blend with 0.4 g acrylic acid. At about the half weight concentration of acrylic acid (0.4 g) with respect to gelatin (1 g), there was a high degree of
cross-linking due to EB irradiation, which led to an increase in the $\varepsilon'$ value.

As illustrated in Fig. 2a–d the $\varepsilon'$ dependence on the frequency of the applied AC electric field shows that at high frequencies the induced dipoles begin to lag behind the applied field and/or the dipoles' alignment is gradually failing to follow the applied field, hence $\varepsilon'$ is constant.\textsuperscript{6} Moreover, it was observed that $\varepsilon'$ of blank blends with 0.2 g, 0.6 g, and 1 g acrylic acid was higher than $\varepsilon'$ of the blends irradiated by 50 kGy EB, except for the blend with 0.4 g acrylic acid.

**Dielectric Loss**

Figure 3a–d show the $\varepsilon''$ dependence on the frequency of the applied AC electric field in the range from 70 Hz to 900 Hz for gelatin blends irradiated by a 50-kGy EB dose as compared to the blank blends. The decrease in $\varepsilon''$ with increased frequency is possibly because the dipoles are aligned directly to the electric field (indicating the dielectric polarization).

At very low frequencies, the increase in the $\varepsilon''$ values is due to the interfacial polarization.\textsuperscript{72} Moreover, it was clear that $\varepsilon''$ of blank blends with 0.2 g, 0.6 g, and 1 g acrylic acid was higher than $\varepsilon''$ of the blends irradiated by a 50-kGy EB dose, except for the blend with 0.4 g acrylic acid.

Figure 4a–d shows the $\varepsilon''$ dependence on the frequency of the applied AC electric field in the range from 1.1 kHz to 5 MHz for the blends as compared to the blank blends. At high frequencies, the decrease in the values of $\varepsilon''$ is due to the phase lag of dipole orientation against the changing electric field. At very high frequencies, $\varepsilon''$ is constant as a result of the dipoles' alignment with the applied oscillating field, and gradually the orientation process stops.\textsuperscript{73}

From the previous figures, it is obvious that the irradiated blends were smaller than the blanks except for the acrylic acid concentration of 0.4 g. At about the half weight concentration of acrylic acid (0.4 g) with respect to gelatin weight (1 g), there is a high degree of cross-linking due to the EB irradiation, which leads to an increase in $\varepsilon'$ and $\varepsilon''$ for irradiated blends. At high frequencies (1.1 kHz–5 MHz).
MHz), there was an essential difference in $\varepsilon$ and $\varepsilon''$ values of gelatin–acrylic acid blank blends as compared to those irradiated by 50 kGy EB. During EB irradiation, the $\varepsilon$ and $\varepsilon''$ values decreased more for the irradiated blends than for the blanks for all acrylic acid concentrations except 0.4 g. This indicates that the effect of EB irradiation cross-linking is notable in the high frequency region. The reduction in the $\varepsilon$ and $\varepsilon''$ values for the blends after EB irradiation cross-linking is explained as the interfacial cross-links in the acrylic acid–gelatin blend that cause the formation of a three-dimensional network, which limits the polarization.74

Dielectric Loss Factor (Dissipation Factor)

Figure 5a–d shows the tan$\delta$ dependence on the frequency of the applied AC electric field in the range from 70 Hz to 900 Hz for gelatin blends irradiated by 50 kGy EB as compared to the blank blends. At low frequencies, the predominance is the ohmic active components as compared to the capacitive reactance ones.70 Also, the space charges are able to follow the applied field. The tan$\delta$ dependence on the frequency of the applied AC electric field is shown in Fig. 6a–d in the range from 1.1 kHz to 5 MHz for gelatin blends irradiated by a 50-kGy EB dose as compared to the blanks. At higher frequencies, the space charges do not have enough time to build up and undergo relaxation. Because the charge carriers are delocalized and change the ohmic component into a frequency-independent factor, there is an obvious decrease in dissipation factor.70

Dependence of AC Electrical Conductivity on Frequency

$\sigma_{ac}$ dependence on the frequency of the applied AC electric field in the range from 70 Hz to 900 Hz is shown in Fig. 7a–d for gelatin blends irradiated by a 50-kGy EB dose as compared to the blanks, while Fig. 8a–d shows the $\sigma_{ac}$ dependence on the frequency of the applied AC electric field in the
range from 1.1 kHz to 5 MHz for gelatin blends irradiated by a 50-kGy EB dose as compared to the blanks. At very low frequencies, it was clear that the relationship between conductivity and frequency was independent due to the free charge carriers’ drift mobility. As the frequency increases (at a definite frequency called the hopping frequency \( \omega_p \)), \( \sigma_{ac} \) is increased linearly as a result of the bound charge with dielectric relaxation. At 0.4 g weight concentration of acrylic acid, \( \sigma_{ac} \) of the EB irradiated blend was higher than the blank, which means it has more defects. The same behaviour is observed in the frequency ranges from 70 Hz to 900 Hz and 1.1 kHz to 5 MHz. The increase in \( \sigma_{ac} \) by increasing the applied frequency indicates that there may be more free charges involved in the hopping process through the defective sites along the blend chains. This is a common characteristic of disordered materials. It was obvious that \( \sigma_{ac} \) increases linearly with increasing frequency according to the Jonscher power law.

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**Fig. 3** Dielectric loss dependence on frequency from 70 Hz to 900 Hz at room temperature: (a) 0.2 g, (b) 0.4 g, (c) 0.6 g and (d) 1 g of acrylic acid.
Comparison between \( \varepsilon \) and \( \sigma_{ac} \) for the blank blends irradiated by various EB doses of 50, 75, 100 and 150 kGy at specific frequencies of 0.5 kHz, 3 kHz, 0.1 MHz and 1 MHz are displayed in Figs. 9a–d and 10a–d, respectively. EB irradiation reduces \( \varepsilon \) at higher irradiation doses because of the formation of inter- and intra-bonds as interfacial cross-links in the blends during irradiation at 100 kGy. Otherwise, the increase in \( \varepsilon \) values (at 75 kGy) is due to the irradiation in air that is attributed to the increase in the number of dipoles as a result of increasing the double bonds. The other reason is attributed to the formation of new defect sites in the band gaps of the blends as a result of chain scission and rearrangement of linking bonds. These defects tap the charge carriers that exist in the band gap of the blends. The irradiation increases the ability of blends to store charges. At the highest EB dose (150 kGy), the irradiation starts to break the branched chains for all blends due to the formation of more defects, except for the 1 g blend. For the blend of 1 g acrylic acid–gelatin at 150 kGy, the increase in EB irradiation dose may increase the number of free radicals that may react to decrease the number of dipoles in the blends. 77

It was found that for the acrylic acid concentrations of 0.2 g and 0.4 g
• The lowest frequency (0.5 kHz) has the highest $\varepsilon$ values.
• The irradiated blends with the lowest $\varepsilon$ values were those with an EB dose of 100 kGy.

But with respect to acrylic acid concentrations of 0.6 g and 1 g

• The lowest frequency (0.5 kHz) has the highest $\varepsilon$ values.
• The irradiated blends with the lowest $\varepsilon$ values were those with an EB dose of 50 kGy.

It was found that for all acrylic acid concentrations, the behaviour of $\varepsilon$ values in the MHz frequency range was similar to that in the kHz frequency range. Moreover, it is deduced that

1. The lowest dielectric parameters can be in the case of the low concentrations of acrylic acid (0.2 g and 0.4 g) blends and irradiated by 100 kGy EB.
2. By increasing the concentration to 0.6 g or 1 g, the blends should be irradiated by 50 kGy EB to give the lowest dielectric parameters.

The increase in $\sigma_{ac}$ by increasing EB irradiation doses at 150 kGy for the acrylic acid concentration that was lower than the half weight concentration of gelatin, 0.2 g and 0.4 g, started to break the branched chains and was attributed to the space charge polarization due to the EB irradiation as shown in Fig. 10a–h. Also, the same behaviour occurs for the blends irradiated by 75 kGy of the acrylic acid

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**Fig. 5** Loss factor dependence on frequency from 70 Hz to 900 Hz at room temperature: (a) 0.2 g, (b) 0.4 g, (c) 0.6 g and (d) 1 g of acrylic acid, respectively.
concentration that was larger than the half-weight concentration of gelatin, 0.6 g and 1 g. This means that there is a degradation of gelatin chains in the blend due to chain scission. The decrease in $\sigma_{ac}$ by increasing EB irradiation doses at 75 kGy for the acrylic acid concentration that was lower than the half-weight concentration of gelatin, 0.2 g and 0.4 g, which is due to the formation of some defects in the energy gap. Also, the same behaviour occurs for the blends irradiated by 150 kGy of the acrylic acid concentration that was larger than the half-weight concentration of gelatin, 0.6 g and 1 g. This is related to the formation of defects, also called clusters or traps, leading to the creation of barriers against the mobility of electrons (charge carriers). This means that EB irradiation induced the cross-linking in the acrylic acid–gelatin blend.

It was found that for the acrylic acid concentrations of 0.2 g and 0.4 g

- The lowest frequency (0.5 kHz) has the lowest $\sigma_{ac}$ values.
- The irradiated blends with the highest $\sigma_{ac}$ values were those with an EB dose of 150 kGy.

But with respect to acrylic acid concentrations of 0.6 g and 1 g

\[ \text{Fig. 6 Loss factor dependence on frequency from 1.1 kHz to 5 MHz at room temperature: (a) 0.2 g, (b) 0.4 g, (c) 0.6 g and (d) 1 g of acrylic acid. respectively.} \]
The lowest frequency (0.5 kHz) has the lowest $\sigma_{ac}$ values.

The irradiated blends with the highest $\sigma_{ac}$ values were those than with EB dose of 75 kGy.

It was also found that for all acrylic acid concentrations, the behaviour of $\sigma_{ac}$ values in the MHz frequency range was similar to that in the kHz frequency range. Moreover, it is deduced that

1. The highest AC electrical conductivity can be achieved in the case of the low concentrations of acrylic acid (0.2 g and 0.4 g) blends irradiated by 150 kGy EB.

2. By increasing the concentration to 0.6 g or 1 g, the blends should be irradiated by 75 kGy EB to give the highest AC electrical conductivity.

In a comparison between the different EB doses for each acrylic acid concentration and blank blends at a definite frequency, it was found that the lowest $\epsilon$ values for 0.2 g and 0.4 g acrylic acid blends at 100 kGy EB dose and 0.5 kHz were 0.579 and 0.315, respectively. The lowest $\epsilon$ values for 0.6 g and 1 g acrylic acid blends at 50 kGy EB dose and 0.5 kHz were 0.579 and 0.315, respectively.
kHz were 0.475 and 0.412, respectively. The $\varepsilon$ order for the acrylic acid concentrations was 0.2 g > 0.6 g > 1 g > 0.4 g blends. Figure 11a and b shows the $\varepsilon$ values for blends with different acrylic acid concentrations at (0.5, 3) kHz and (0.1, 1) MHz irradiated by different EB doses, respectively. It was observed that the 0.4 g and 1 g acrylic acid concentrations had the highest and lowest $\varepsilon$ values for the kHz and MHz frequency ranges, respectively. The increase in the $\varepsilon$ value is attributed to the mechanism of space charge polarization that appears obviously at 0.4 g acrylic acid blend. 78 Also, it was found that the lowest $\varepsilon$ values were for gelatin blended with 1 g acrylic acid weight concentration and irradiated with a 50-kGy EB dose.

In a comparison between the different EB doses for each acrylic acid concentration and blank blends at a definite frequency, it was found that the highest $\sigma_{ac}$ values for 0.2 g and 0.4 g of acrylic acid blends at a 150-kGy EB dose and 5 MHz were $1.71 \times 10^{-4}$ S/m and $8.56 \times 10^{-5}$ S/m, respectively. The highest $\sigma_{ac}$ values for 0.6 g and 1 g of acrylic acid blends at a 75-kGy EB dose and 5 MHz were $1.39 \times 10^{-4}$ S/m and $1.08 \times 10^{-4}$ S/m, respectively. Then, the $\sigma_{ac}$ order for the acrylic acid concentration was 0.2 g > 0.6 g > 1 g > 0.4 g. Figure 11c and d shows the $\sigma_{ac}$ values for the different acrylic acid concentrations at (0.5, 3) kHz and (0.1, 1) MHz irradiated at different EB doses, respectively. It was clear that the 0.2 g and 0.4 g acrylic acid...
acid concentrations were the highest and lowest $\sigma_{ac}$ values for the kHz and MHz frequency ranges, respectively. Also, it was found that the highest $\sigma_{ac}$ values were for gelatin blended with 0.2 g of acrylic acid weight concentration and irradiated with a 150 kGy-EB dose. It is clear that the increase or decrease in $\sigma_{ac}$ values depends on the concentration of acrylic acid, EB irradiation dose, and frequency of the applied field. The increase in $\sigma_{ac}$ was due to the irradiation starting to break the branched chains and was attributed to the space charge polarization as shown in Fig. 11c and d. The highest $\sigma_{ac}$ values were for 0.2 g and 0.6 g of acrylic acid blends that were irradiated by 150 kGy and 75 kGy at frequencies of 0.5 kHz, 3 kHz, 0.1 MHz, and 1 MHz, respectively. The decrease in $\sigma_{ac}$ was due to the formation of some defects (clusters or traps) in the energy gap that led to the creation of barriers against the mobility of electrons (charge carriers). This means that EB irradiation induced the cross-linking in the acrylic acid–gelatin blend. The lowest $\sigma_{ac}$ values were for 0.4 g and 1 g acrylic acid blends that were irradiated by 150 kGy and 75 kGy at frequencies of 0.5 kHz, 3 kHz, 0.1 MHz, and 1 MHz, respectively.

Fig. 9 Dielectric constant dependence on various EB doses at kHz and MHz frequencies and at room temperature: (a) 0.2 g, (b) 0.4 g, (c) 0.6 g and (d) 1 g of acrylic acid.
Effect of Various Acrylic Acid Concentrations on $\varepsilon$ and $\sigma_{ac}$ for Blank Blends at Various Frequencies

Figure 12a–d shows the dependence of $\varepsilon$, $\varepsilon''$, and $\tan\delta$ on different acrylic acid concentrations at frequencies of 0.5 kHz, 3 kHz, 0.1 MHz, and 1 MHz, respectively. These figures clearly show that dielectric parameters ($\varepsilon$, $\varepsilon''$, $\tan\delta$) have the highest and lowest values at frequencies of 0.5 kHz and 1 MHz, respectively. The blend with 0.4 g of acrylic acid concentration has the lowest $\varepsilon$, $\varepsilon''$, $\tan\delta$ values for the different frequencies. Figure 12d and e shows the $\sigma_{ac}$ dependence on different acrylic acid concentrations of 0.2 g, 0.4 g, 0.6 g, and 1 g blank blends at frequencies 0.5 kHz, 3 kHz and 0.1 MHz, 1 MHz, respectively. It is clear from these figures that $\sigma_{ac}$ has the highest values at the highest frequency (1 MHz) on the order of $10^{-8}$. On the other hand, the lowest $\sigma_{ac}$ values occurred at the lowest frequency of 0.5 kHz on the order of $10^{-8}$. With respect to the acrylic acid concentrations, the 0.4 g and 0.2 g blends have the lowest and highest $\sigma_{ac}$ values at different frequencies. It was found that the $\varepsilon$, $\varepsilon''$, $\tan\delta$ and $\sigma_{ac}$ values of the 0.8-g acrylic acid concentration were very close as compared with the 1 g concentration of acrylic acid.

In Fig. 12a, the higher value of $\varepsilon$ for the 0.2 g of acrylic acid–gelatin blend is related to the presence of double bonds and the acrylic acid as a filler that increases $\varepsilon$ due to the increased interfacial polarization. An interfacial polarization is created due to the charges moving and then accumulating at the interface in the blended matrix under the applied field. So the field generated polarizes the surrounding matrix. As the content of acrylic acid is increased, the inter-particle distance increases. So the blend of 0.2 g acrylic acid gives the highest value of $\varepsilon$, followed by 0.6 g and 1 g acrylic acid blend, and then the lowest is at 0.4 g. Because the blend of 0.4 g acrylic acid represented nearly the half-weight of gelatin (1 g), that results in the highest degree of inter-particle distance. In polymers, there is a relaxation due to the formation of dipoles on the side chains, i.e., viscoelasticity. Then, there is a shift in the $\varepsilon''$ values for the different blank blends, indicating the influence of acrylic acid addition on the relaxation of gelatin–acrylic acid blends, as shown in Fig. 12b.

Modification of the AC Electrical Conductivity Equation As a Function of Frequency and Acrylic Acid Concentration at Room Temperature

At the highest EB dose (150 kGy), the relationship between AC conductivity as a function of acrylic acid concentration and frequency can be obtained by Eq. 5:

$$\sigma_{ac} = 10^{-8} \times (2 \times f_{value}) \times X^{-0.4}$$  (5)

where $f_{value}$ is the frequency in kHz and $X$ is the acrylic acid concentration in g.

Therefore, we compare the experimental data of $\sigma_{ac}$ (S/m) at 0.5 kHz [Exp. (0.5 kHz)] and 3 kHz [Exp. (3 kHz)] with the fitting data 0.5 kHz [Power (0.5 kHz)], 3 kHz [Power (3 kHz)] and also the data using the modified Eq. 5 for 0.5 kHz [Eq. (0.5 kHz)] and 3 kHz [Eq. (3 kHz)], as shown in Fig. 13a. For example, the term [Exp. (0.5 kHz)] is the experimental data for the ac conductivity measured at 0.5 kHz electric field frequency for the different weight concentrations of acrylic acid in g, [Power (0.5 kHz)] is the fitting for experimental values of ac conductivity at 0.5 kHz electric field frequency for the different weight concentrations of acrylic acid in g, [Eq. (0.5 kHz)] is the fitting for experimental values of ac conductivity at 0.5 kHz electric field frequency for the different weight concentrations of acrylic acid in g, [Eq. (0.5 kHz)] is the calculated values according to the Eq. (5). Also, we repeated that with respect to the experimental data of $\sigma_{ac}$ at frequencies of 0.1 MHz and 1 MHz using Eq. (5), as shown in Fig. 13b.

Conclusions

- For the frequency of the applied field, the lowest kHz (0.5 kHz) gives the highest dielectric parameters than the highest MHz (1 MHz) for blank and EB irradiated blends. In addition, for blank and EB irradiated blends, the lowest kHz (0.5 kHz) has the lowest AC electrical conductivity compared to the highest MHz (1 MHz).
- For the different weight concentrations of acrylic acid and the blank EB blends, the acrylic acid concentration of 0.2 g has the highest dielectric parameters and AC electrical conductivity and the acrylic acid concentration of 0.4 g has the lowest dielectric parameters and AC electrical conductivity.
For the different EB doses, for the irradiated EB blends, the highest dielectric parameters occurred for the lowest concentration (0.2 g) after the irradiation by a high EB dose (100 kGy), and the highest multiplicity concentrations (e.g., 0.6 g and 1 g) should be irradiated by the half value of the EB dose (50 kGy). The highest AC electrical conductivity occurred for the lowest concentration (0.2 g) after the irradiation by a high EB dose (150 kGy), and the highest multiplicity concentration (e.g., 0.6 g and 1 g) should be irradiated by the half value of the EB dose (75 kGy). The lowest dielectric parameters occurred for the concentration (0.4 g) after the irradiation by a high EB dose (100 kGy), and the lowest AC electrical conductivity occurred for the concentration (0.4 g) after the irradiation by a high EB dose (150 kGy).

There is compensation between the EB doses and the concentration of acrylic acid to give the blends of the lowest dielectric parameters and the highest AC electrical conductivity. Also, the EB irradiation can cause cross-linking and/or chain scission reactions that depend on the irradiation dose and appear in the dielectric parameters and AC electrical conductivity. The chain scission causes the degradation in the blends, leading to the formation of free radicals that either improve the cross-linking reaction or accelerate the degradation. At the focus point, the dielectric behaviour of the blends arises due to the presence of an appreciable number of defects in the form of breaking of...
bonds, bending, cross-linking, etc. During EB irradiation, the increase in these defects leads to the formation of a greater number of dipoles that affect the dielectric and AC electrical properties.

Finally, there is a modified equation between the AC conductivity, the frequency of the AC applied field in the kHz and MHz range, and the concentration of acrylic acid mixed with gelatin at room temperature only.
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