Dielectric Properties of High Alumina Glass

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Abstract

High Alumina Glasses "Alumina Silicate Glasses" was prepared by utilizing powder technology technique. The starting materials are wasted soda–lime glass from an industrial site, Iraqi-Duekhla raw kaolin and small amounts of potassium carbonates.

X-ray Diffraction and FTIR analysis show complete vitreous glasses are obtained. Increasing Alumina content plays an obvious role in improving physical and mechanical properties of the prepared high alumina glass. In addition, the increased alumina content enhanced the dielectric constant and reduced dielectric loss. These results may be interpreted as due to the bridging bonding of alumina with both the silica and the alkali oxides; which impedes the alternating electric field.

Introduction

The effect of the applied frequency on dielectric constant and loss tangent for selected commercial glasses were studied by Babcock (1977)(1). The dielectric constant of soda lime glass at 10^6 Hz is found to be 6.9 and the loss tangent at frequency 10^6Hz is 0.009. The Dielectric glasses have applications across the entire spectrum of electronic circuits and their past success on a variety of manned and unmanned space missions continues to interest the defense and aerospace industries. Other applications where dielectric glass is currently utilized are shown by Demeko (2005)(2) as high temperature circuitry (Transistor Biasing), radar systems voltage controlled oscillators and amplifier coupling etc.

The aim of this study is to prepare "High Alumina Glasses", also known as "Alumina Silicate Glasses", utilizing cheep source of raw materials. In addition, the study aimed to improve the glass properties by finding better composition, melting temperature and

additives. Finally, this study includes the analysis of the resultant properties of the prepared glasses.

Experimental part

The starting materials were commercial K_2CO_3, Duekhla Kaolin (from Ministry of Industry, Office of Mining and Geology) and Soda Lime Glass S.L.G. (from Ministry of Industry, Al-Taji Glass Manufacturing Site). The chemical analysis for the Kaolin and S.L.G. is shown in the following tables;

The glass was milled to a fine powder by blast method. Then powder batches are prepared by the weighing of the constituents as listed in table (3). The batches were mixed in a porcelain jar laboratory blender for about 30 min to achieve homogeneity. The mixed powders were melted at 1100°C by using alumina crucibles. Afterward, the glass batches were crashed and milled by ball milling and sieved by using laboratory sieve (size 53 micron). Units of 5g of each batch were pressed to disks of 1cm diameter for 3 min and pressure of 3 tons. The glass samples were annealed at temperature of 750°C for 2h to obtain glass bulks.

Grinded and polished samples are utilized for measurements of the physical properties; namely, the Bulk density B.D (the ratio of the mass of material to it's bulk volume i.e, volume of solid component +open pores +sealed pores), Apparent Porosity A.P(expresses as a percent, is the ratio of the volume of the open pores of the specimen to its exterior volume), and Water Absorption W.A(expresses as a percent, is the relationship of the mass of water absorbed to the mass of dry specimen). These physical properties are measured according to ASTM C373-88 (3). In additions, Diametrical Strength test (Brazilian test) is carried by using PHEWE test machine located at Nahrian university. The average of five measurements for each prepared glass is considered.

X-ray (Cu-Kα) diffractograms for selected glass samples are measured by seeking for any residual crystalline phase. FTIR is also used to prove the amorphous state of the prepared glass.

The dielectric properties were measured by using Precision Impedance Analyzer type Agilen technologies 42942A located at the Ministry of Science and Technology. Dielectric constant (relative permittivity ε'), loss tangent (dissipation factor tanδ), loss index (Loss factor ε''), have been measured(4). The frequency range extends from 40Hz to 10MHz.

Results and Discussion

Figure (1) shows the bulk density for the glasses prepared with different Potassium carbonate content. This figure show that the bulk density increases with increasing Kaolin content. It is observed that the bulk density data show no apparent contribution of K_2CO_3 content to their values. Figure (2) shows the apparent porosity and water absorption for the prepared glasses. Both the apparent porosity and water absorption are very close to zero for the prepared glasses. The increased densities with increasing kaolin contents may reflect the new compositions of the glasses. The small values of the apparent porosity and water absorption suggest microstructure with little open and close pore population.

X-ray diffractograms for selected samples are shown in figures (3-5). These figures show no evidence of the existence of any crystalline phase. FTIR spectrograms for kaolin, soda lime glass and selected prepared glass samples are shown in figure (6). The FTIR spectrogram shows absorption peaks in the vicinity of peaks positions dedicated for kaolinite (3694, 3650, and 3620 cm^{-1}). The peaks positions around 1000 cm^{-1} cannot be used for judgment .And because of that, the absorption peaks for kaolinte, quartz and vitreous silica are overlapped there. The peak position at 800 cm^{-1} is again common to quartz and vitreous silica and the peak at 462 cm^{-1} of the quartz cannot be distinguished from the peak of 464 cm^{-1} of the vitreous silica. These peaks are obvious in spectrograms of soda lime glass and the prepared glasses. However, the absorption peaks dedicated to quartz at 695 and 514 cm^{-1} are not noticed on the spectrograms. The peaks at 936 and 912 cm^{-1} that distinguish mullite from other phases are also not noticed. On the contrary, the peaks which clearly indicated the vitreous silica (1636 and 958 cm^{-1}) exist in the spectrograms.

The above analysis of the absorption peaks is relied on the FTIR analysis and absorption peak positions published by Simon (1953)[5] and Ojima(2003)(6). The conclusion is that the prepared glass recipes are totally melted and vitrified which are consistent with the XRD results. This demonstrates that the kaolin and the alkali oxide additives are completely melted and vitrified. Accordingly, the physical, mechanical and electric properties of the prepared glasses

should be attributed exclusively to the vitreous (glassy) state of the prepared glasses.

The diametrical tests shown in figure (7) show an obvious correlation of kaolin and alkali oxide content with strength σ_D. The diametrical strength increases with kaolin content for all the prepared glasses. This is consistent with the increase of alumina content of kaolin source. The increase of the strength σ_D with potassium carbonate content may be due to the new glassy phases originated from the existence of potassium oxide.

Figure (8) shows the dielectric constant versus the applied frequency for glasses prepared with different wt% of K_2CO_3 and wt% of Kaolin. The dielectric losses are presented in figure (9) and the loss tangents are presented in figure (10). A common behavior of the dielectric constant against the frequency is the trend to decrease as the applied frequency increase. Nevertheless, a minimum is recorded for the dielectric constant value for all the prepared classes around 200Hz. This can be attributed to one of two types of polarization or both of them. The first polarization type is the space charge polarization in view of the possible existence of pores-glass medium interfaces. The second polarization type is the dipolar polarization which is common for vitreous material phases(7-8).

As the applied frequencies cross 10^4Hz, the dielectric constant is found in a plateau region. This is common for all materials in view of that the frequency of field becomes larger then what the dipoles can follow. The effect of varying wt% of K_2CO_3 is not clearly shown at 10 and 20 wt% of kaolin. However, for glasses prepared with 30% of Kaolin, the effect of the variation of K_2CO_3 wt% is revealed. The increase of K_2CO_3 in glass batches shows a decrease in the dielectric constant. This may indicate that the probable silica-alumina-alkali oxide phases contribute more to polarization, hence, it would reduce the dielectric constant.

The dielectric losses, as expected, show their primary peak values at the same position where the dielectric constants show their minimum (200 Hz). This is again applied to all the prepared glasses irrespective of the kaolin or the alkali oxide content. Other dielectric loss peaks values are smaller than the primary peak which possibly refers to the same above mentioned mechanisms of polarization. The loss tangents have the same behavior of the dielectric losses. The significance of these plots is that they all show relatively low loss

tangents. Their values are around 0.7 at peak values and much less when the applied frequency increases. This indicates the capability to invest the prepared glasses as cheap dielectrics.

The above figures also show that the glasses prepared with 30 wt% of kaolin have generally higher dielectric constants and lower losses. This indicates that the increase in alumina (that comes from the kaolin) helps in reducing the polarization. The alumina usually forms bridging bonds between the silica and the alkali oxides as discussed by Babcock (1994)[9]. The role of alumina in reducing the polarization can be understood in view that the bridging alumina would hinder, to some extent, the polarization.

Conclusions

1- The dielectric properties of the prepared High Alumina Glasses indicates the capability to invest it as cheap dielectrics.

2- The role of Alumina in the prepared Glasse is that they show higher dielectric contants and lower losses as the Alumina contents increases.

References

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Table (1): Chemical analysis of Duekhla Kaolin.

Composition	Weight %	Composition	Weight %
SiO_2	47.26	CaO	0.15
Al_2O_3	34.84	MgO	0.38
Fe_2O_3	1.32	NaO_2	0.25
TiO_2	1.4	K_2O	0.61
L.O.I: (Loss of Ignition) = 12.91%			

Table (2): Chemical analysis of the Soda Lime Glass.

Composition	Weight %	Composition	Weight %
Na_2O	16.4	Fe_2O	0.6
CaO	5.0	SO_3	0.15
K_2O	0.35	Cl	0.05
MgO	3.45	TiO_2	0.01
Al_2O_3	1.45	SiO_2	72.29
Softening point: 693-699 °C, Melting point: 854 °C.			

Table (3): Batch content of the prepared glasses.

Batch No.	S.L.G. wt %	Kaolin wt %	K_2CO_3 wt %
1	89.25	10	0.75
2	88.50	10	1.50
3	87.80	10	2.20
4	78.50	20	1.50
5	77.80	20	2.20
6	77.00	20	3.00
7	68.50	30	1.50
8	67.00	30	3.00
9	65.50	30	4.50

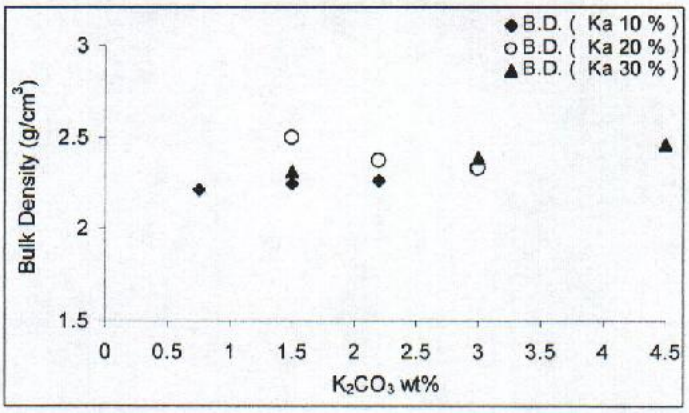
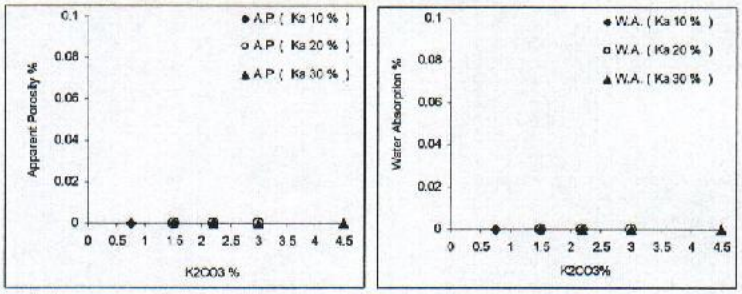


Fig. (1): Bulk Density versus K_2CO_3 percentage at different Kaolin content.



(a) (b)

Fig. (2): (a) Apparent Porosity and (b) Water Absorption versus K_2CO_3 wt% at different Kaolin content.

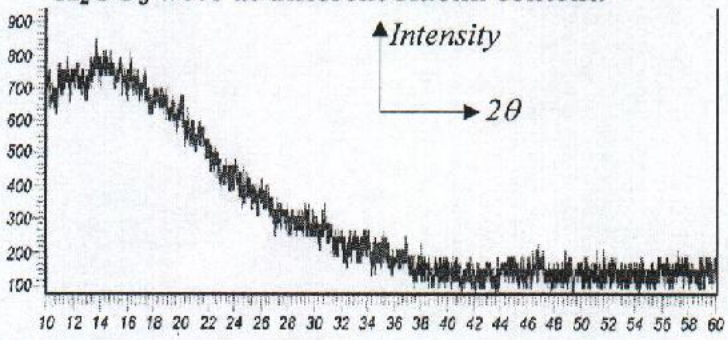


Fig. (3): XRD diffractogram of glass prepared with 10% Kaolin and 1.5% K_2CO_3.

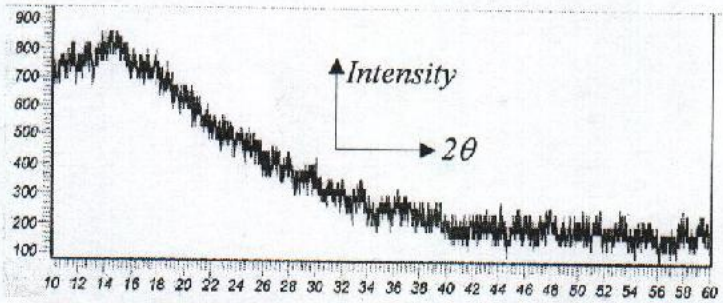


Fig. (4): XRD diffractogram of glass prepared with 20% Kaolin and 2.20% K_2CO_3.

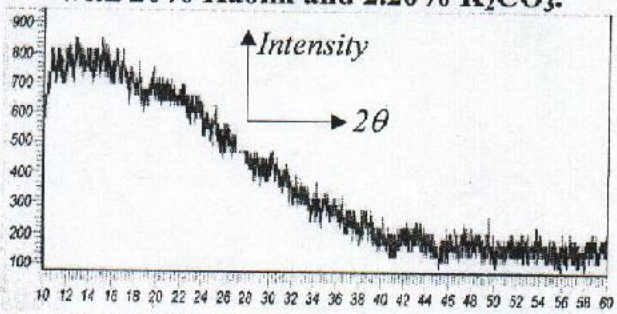


Fig. (5): XRD diffractogram of glass prepared with 30%Kaolin and 3.0% K_2CO_3.

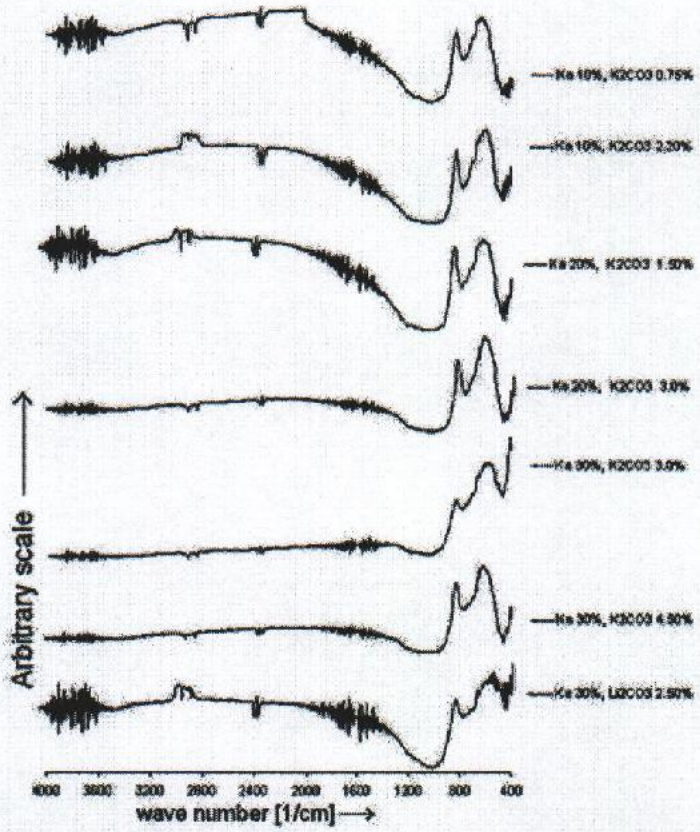


Fig. (6): FTIR spectrograms for selected samples showing dominant vitreous silica absorption peaks for the prepared glasses.

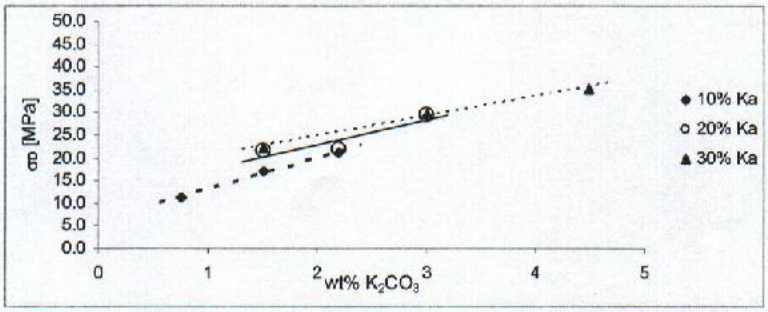
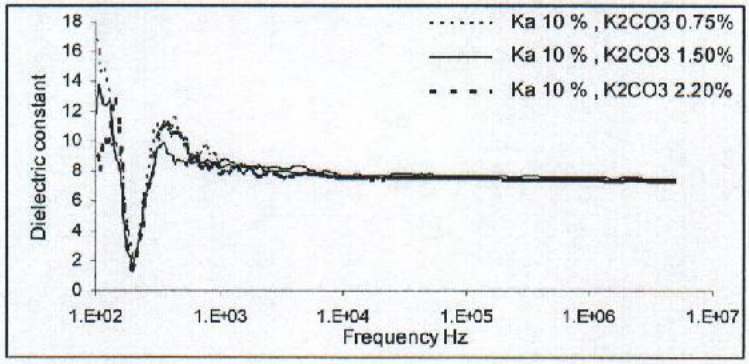
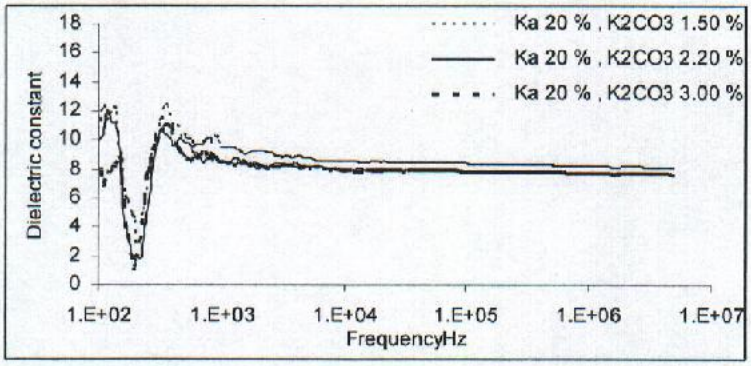


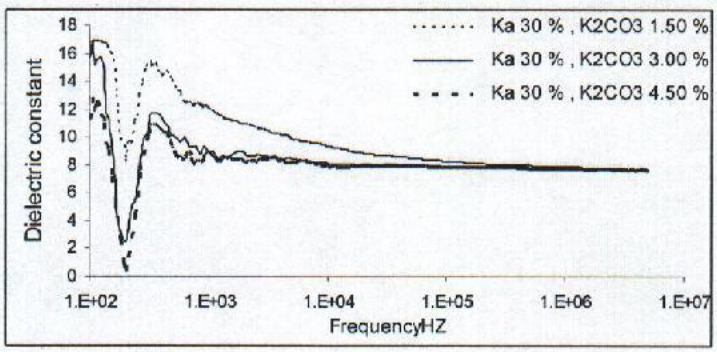
Fig. (7): Diametrical strength versus wt% of K_2CO_3 for glasses prepared with different wt% of kaolin.



(a)

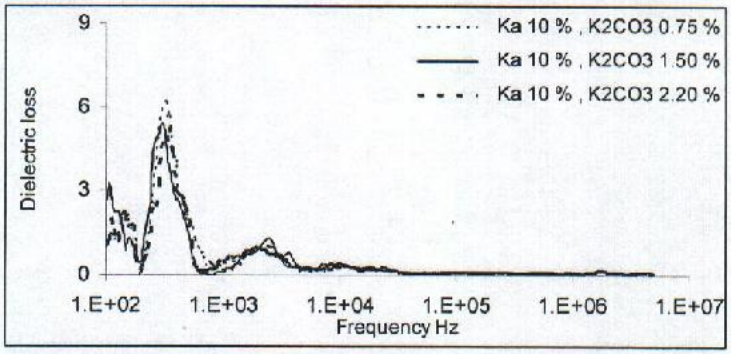


(b)

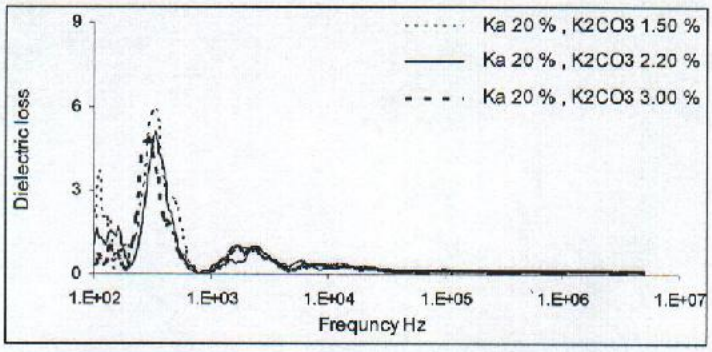


(c)

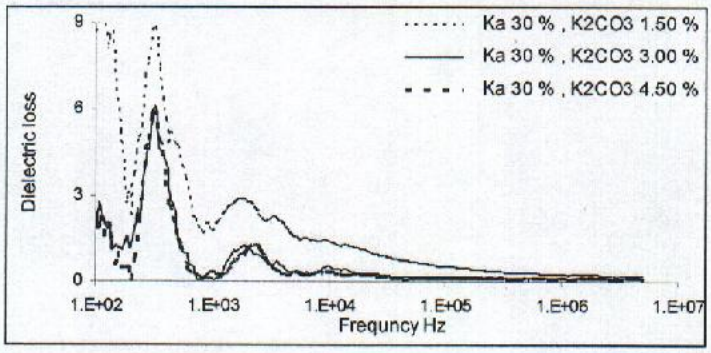
Fig (8): Dielectric constant versus the applied frequency for glasses prepared with different wt% of K2CO3 and (a) 10 wt% of kaolin (b) 20 wt% of kaolin (c) 30 wt% of kaolin -



(a)



(b)



(c)

Fig. (9): Dielectric loss versus the applied frequency for glasses prepared with different wt% of K2CO3 and (a) 10 wt% of kaolin (b) 20 wt% of kaolin (c) 30 wt% of kaolin

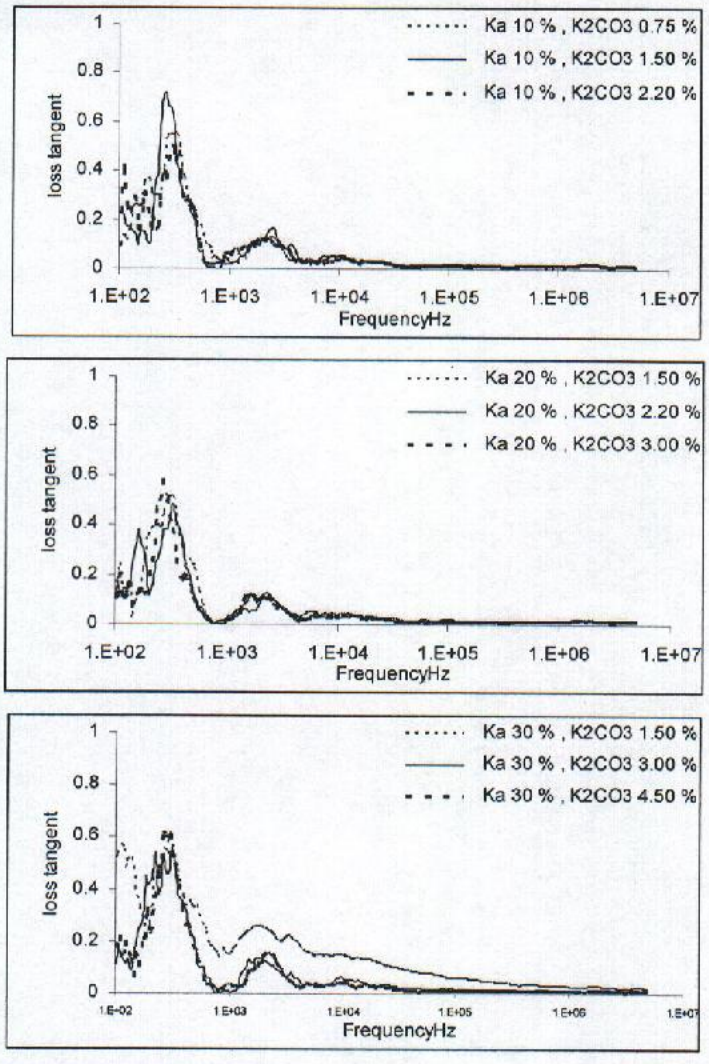


Fig. (10): Loss tangent versus the applied frequency for glasses prepared with different wt% of K_2CO_3 and (a) 10wt% of kaolin (b) 20wt% of kaolin (c) 30wt% of kaolin

الخواص العزلية للزجاج عالي الالومينا

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الخلاصـــــة

حضرت انواع من الزجاج عالي الاومينا "زجاج الالومينا-سليكيا" باستخدام تقنية تكنولوجيا المساحيق. وكانت المواد الاولية هي الزجاج الصودا-لايم المتخلف من منشأة صناعية ،وكاؤلين دويخلة العراقي، ومقادير قليلة من كاربونات البوتاسيوم.

بينت نتائج حيود الاشعة السينية وفحص الاشعة تحت الحمراء ان الزجاج المحضّر هو في حالة لابلورية تماما. وقد أدى زيادة محتوى الالومينا دورا واضحا في تحسين المواصفات الفيزيائية والميكانيكية. فضلا عن ذلك، فان زيادة محتوى الالومينا قد زاد من قيم ثابت العزل وقلل من الفقدان العزلي. يمكن تفسير هذه النتائج من خلال ترابط جسري محتمل للالومينا مع السيليكا من جهة والاكاسيد القاعدية من الجهة الاخرى، وهذا يؤدي الى مقاومة الحقل الكهربائي المتذبذب المسلط.