

Received:
19 September 2018
Revised:
17 January 2019
Accepted:
13 February 2019

Cite as: G. A. Khater,
Mohamed M. Gomaa,
Junfeng Kang,
M. A. Mahmoud. Effect of
CaO/SiO₂ molar ratio on the
electrical and physical
properties of basaltic glass
materials.
Heliyon 5 (2019) e01248.
doi: [10.1016/j.heliyon.2019.e01248](https://doi.org/10.1016/j.heliyon.2019.e01248)



Effect of CaO/SiO₂ molar ratio on the electrical and physical properties of basaltic glass materials

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Abstract

The effect of CaO/SiO₂ molar ratio on the electrical properties of some glass materials was examined, using the impedance spectroscopy, at different frequency ranges, from 100 Hz up to 5 MHz. Also, a trial was accomplished to study the influence of CaO/SiO₂ molar ratio on some physical properties such as density, micro-hardness and bending strength. Six glass batches, based on Sinai basaltic rocks and bypass cement dust, were prepared with different CaO/SiO₂ molar ratios (0.2–0.93 mol %). Accordingly, the electrical properties (conductivity dielectric constant and dielectric loss) of these samples show noticeable change. Electrical results show that the samples with relatively low (CaO/SiO₂) molar ratio have relatively higher electrical conductivity, compared to the other samples with higher (CaO/SiO₂) molar ratios, at different frequencies. Also, at the same time, the bending strength and Vickers micro-hardness show a gradual increase from 56 to 118 MPa and from 4020 to 6120 MPa, respectively, with decreasing CaO/SiO₂ molar ratio. The density of the samples shows a successive increase from 2.79 to 2.96 gm/cm³ with the successive additions of bypass cement dust.

Keywords: Condensed matter physics, Materials science

1. Introduction

The presence of external electrical field changes greatly the electrical properties of glass due to the transfer of cations from its original places to another one under the effect of that field. The structure of the glass material is very important factors in the electrical properties. Traditional glasses are poor conductors at room temperature. The conduction at the network of glasses is the result of the ions movements. Movements of these ions within the samples lead to the variation of the electrical conduction. Also, there is another factor that affects greatly the electrical properties to a great extent, it is the impurities at the glass, even if it is present of small quantities.

The mobility of ions under the effect of an external electric field can be calculated as follows

$$\mu = \frac{\sigma}{nq} \quad (1)$$

σ (Eq. (1)) is the electrical conductivity, μ is the ionic mobility, n is the number of charge carriers (per cm^3), q is the transporter electrical charge. Usually, the charge carriers have low concentrations and are attached weakly to the surface of the glass. If these charge carriers are connected in a series between the two electrodes (when measured), then, it will make a direct current (DC). Free charge carriers, usually, have low concentrations and they are not effective to make a current between the electrodes in the sample. Accordingly, the samples need to be motivated thermally or electrically to be able to feel that current (even with high ion concentrations) [1]. The investigation of the electrical properties in the silicate glass as function of different calcium silica ratio is of great interest due to its importance in technological applications.

In this study silicate glass materials based on basaltic rocks from South Sinai governorate, Egypt, and cement waste from Masr Bani Suif pilot plant were combined with different proportions. The major constituents of basalt are some oxides of (Silica, alumina, iron, calcia, magnesia) in addition to some other minor alkali constituents (soda, potassia, titania and manganese) [2]. Low viscosity basaltic rocks produce polycrystalline components at low temperatures with short production cycles [3, 4]. Also, basaltic rocks have a high oxidation resistance, low thermal conductivity, and high softening and melting temperatures. Basaltic rocks are, usually, used as resistant materials for fires [5, 6, 7]. The wastes from cement manufacturing industries produce cement which, nearly, represents $\sim 7-10\%$ of the output ability of cement manufacture. Calcium oxide and silica are the major components of the cement wastes [8, 9, 10].

By following the changes of concentration between calcium oxide and silica (from basaltic rocks), it is possible to produce various and different glass compositions. Also, it is possible to study its influence and effects on the electrical and physical properties of the glass materials.

Vitreous silica has an electrical conductivity of the order of 5×10^{-12} S/cm at 300 °C. With the increase of sodium oxide concentration, the electrical conductivity values, accordingly, increase up to 3×10^{-6} S/cm for 10% mole until it reaches 3×10^{-3} S/cm for 50% mole and the glass structure are changed [11].

Dutta et al. [12] measured electrical conductivity for some samples of sodium oxide glasses. Samples have concentrations in the range from 20% to 27.5% mol. The sodium oxide concentrations were replaced by calcium oxide (silica oxide constant $\sim 70\%$ mol). With increasing sodium oxide concentration, the conductivity changed from 2×10^{-9} S/cm to 6×10^{-8} S/cm (at 110 °C). Therefore, the increase of calcium oxide increases the conductivity values, due to the presence of Na⁺ and Na⁺ ions [12].

Studying electrical properties of silicate glasses (dielectric constant or permittivity, dielectric loss, impedance and conductivity), at different frequency ranges, is very important to reveal comprehensive information about the nature and structures of the losses that occur at these materials. Also, it is a good opportunity to study the conduction mechanisms at these glass samples and support the information on the structural aspects of these silicate glasses [13, 14]. The electrical properties of these silicate glasses can be expressed by the real and imaginary components of the complex permittivity or the complex impedance.

The permittivity of these silicate glasses indicates the molecular relaxation and transport processes of the material which based on many parameters such as the concentration of minor and major elements, frequency, temperature, and pressure [15].

Magnesium phosphate glasses [16] were studied to check the influence of the electrical properties of ternary zinc on magnesium oxide. The increase of magnesium oxide (MgO) concentration, leads to the participation of Mg²⁺ or Ca²⁺ ions in the interior structure of the glass. Accordingly, the concentrations of the free (Mg²⁺) ions will decrease. Accordingly, the zinc ions will obstruct the strength of the conduction paths (present at the gaps of the glass matrix). Also, the presence of the temperature factor is a very effective parameter that contributes to increase the conductivity of the samples. This is due to the thermally-activated migration of free ions in the main glass structure. If the thermal energy is improved in the glass samples, then many charge carriers can overcome easily the attraction and diffuse long distances in the sample [16].

Using electrical conductivity over wide frequency ranges (100 Hz–5 MHz), is our goal to determine how ionic mobility in silicate glasses and melts behave when

calcium/silica ratio changes. Specifically, to ascertain the influence of CaO/SiO₂ molar ratio on the electrical and physical properties of the obtained silicate glass matrix.

2. Materials & methods

2.1. Glass preparation and melting

The main raw materials used in this study are basaltic rocks and bypass cement dust. Firstly, samples of basaltic rocks have been collected from South Sinai governorate, and then samples were crushed and milled for a while in an agate mill till reaching very fine powder, where they were sieved to obtain particles sizes less than 75 μm, the obtained powder samples were then mixed up to obtain a homogeneous sample which is highly representative of the chemical composition of the Sinai basaltic rocks.

Secondly, bypass cement dust samples have been collected from Masr Bani Suif factory, and then it was milled for a while, until it reaches the same particle size with basalt powder.

Chemical analyses of basalt samples and bypass cement dust have been accomplished by using X-ray Fluorescence (XRF), then six glass batches were prepared from basalt powder with successive additions of different proportions of cement waste from 10 to 50%. The calculated glass batches are entitled as (G0, G10, G20, G30, G40 and G50) where the number refers to the percentage of cement waste on the expense of the basalt powder, as shown in Table 1.

The obtained batches were mixed-up then melted in an alumina crucible at 1350 °C up to 1450 °C for 2 hours to be homogeneous and bubble-free glass. After that, the bubble-free melt was casted onto a steel plate in the form of very thin disc samples (4.0 mm thickness and 2 × 2 cm dimensions of two parallel surfaces to avoid

Table 1. Chemical composition (mol %) of investigated glass samples.

Glass no	Nominal composition		Calculated composition (mol %)								CaO/SiO ₂	TS
	Bypass	Basalt	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	TiO ₂	CaO	MgO	Na ₂ O	K ₂ O		
G0	0	100	54.86	11.11	4.86	1.39	11.11	12.50	3.47	0.69	0.20	685
G10	10	90	52.11	10.56	4.23	1.41	16.20	11.27	3.44	0.70	0.31	682
G20	20	80	48.95	9.79	4.20	0.70	20.98	10.49	3.41	1.40	0.43	680
G30	30	70	46.43	8.57	3.57	0.71	26.43	9.29	3.39	1.43	0.57	680
G40	40	60	43.17	7.91	3.60	0.72	31.65	8.63	2.88	1.44	0.73	678
G50	50	50	39.86	7.25	2.90	0.72	36.96	7.25	2.90	2.17	0.93	675

irregularity in thickness), and then transferred to a preheated muffle furnace adjusted at 550 °C, which was then switched off to cool gradually to room temperature to prevent rupturing of the glass samples.

2.2. Electrical properties

Impedance measurements were carried out on samples (about 25–26 mm diameter and 5–6 mm thick) using an impedance analyzer (Hioki 3522- 50 LCR Hitester) in the frequency range 100 Hz–5 MHz at room temperatures (20 °C). All samples were sandwiched between two (Cu/CuSO₄) electrodes inside an isolated chamber. Four copper wires connected to the electrodes act as electric leads. The impedance analyzer was connected to a personal computer and the recorded data was directly stored on it. The frequency-dependent complex impedance, $Z^*(\omega)$, is

$$Z^*(\omega) = Z'(\omega) - iZ''(\omega) \quad (2)$$

where $Z'(\omega)$ and $Z''(\omega)$ (Eq. (2)) are the real and imaginary parts of the impedance, respectively and ω is the angular frequency ($\omega = 2\pi f$; and f is the frequency of applied field). The d. c. conductivity was extracted from the a. c. conductivity. The measured parameters were the parallel capacitance (C_p) and conductance (G_p) and the series impedance Z . The complex relative dielectric constant ϵ^* can be written as

$$\epsilon^* = \epsilon' - i\epsilon'' \quad (3)$$

The real (ϵ') and imaginary (ϵ'') (Eq. (3)) part of the complex relative permittivity is

$$\epsilon' = \frac{C_p d}{\epsilon_0 A}, \quad \epsilon'' = \frac{G_p d}{\omega \epsilon_0 A} \quad (4)$$

where; A is the cross-sectional area of the sample (Eq. (4)), d is the thickness of the sample, ϵ_0 is the permittivity of air (8.85×10^{-12} F/m). The conductivity was calculated from the following equations:

$$\sigma' = G_p \frac{d}{A} = \epsilon'' \omega \epsilon_0 \quad (5)$$

σ' (Eq. (5)) is the real part of the conductivity.

2.3. Physical properties

2.3.1. Micro-hardness

Indentation micro-hardness was estimated for the samples. The Vicker's hardness (HV) values were investigated using a Shimadzu Micro-hardness tester of M-type (Japan) under a load of 300 g using the following equation

$$HV = A(p/d^2) \text{kg/mm}^2 \quad (6)$$

where A (Eq. (6)) is a constant (1854.5), p is the applied load (g) and d is the average diagonal length (μm). The loading time for all samples is (20 s).

2.3.2. Bending strength

Bending strength was measured by four points bending of the obtained glass, in order to achieve the strength data from glass samples, similar to those that actually used in the building materials we must use unpolished test pieces, using a Shimadzu Autograph DCS- R-10TS universal testing machine at a crosshead speed of 0.5 mm/min.

2.3.3. Chemical durability and density

The chemical durability of the glass-ceramics was estimated by a chemical etching method. The samples were corroded in 1 N HCl and 1 N NaOH solution at 100 °C for 24 h and then the so-called residual rate and weight loss were calculated. The density of the samples was obtained by Archimedes using ethylene glycol as the immersion fluid.

3. Results & discussion

The combination of basalt powder and bypass cement dust (successive concentration from 10-50 %) was used for the preparation six glass batches. We try to follow the change of calcium silica molar ratio and the possibility to study its influence on both the electrical and physical properties of the obtained glass.

It's clear from the Table 1 that there is a gradual increase of CaO/SiO₂ molar ratio from 0.20 to 0.93 corresponding to different glass samples from G0 to G50. The main composition of the cement waste is CaO and with the successive additions of different proportions (10, 20, 30, 40 and 50 %), the percentage of CaO molar ratio is increased at the expense of silica molar ratio.

Fig. 1 shows the resistivity versus frequency for six glass sample compositions with a successive increase of the amount of (CaO) at the expense of the amount of basalt as mentioned before (Table 1). It is obvious from the curves that the conductivity decreases with the increase of the amount of (CaO) at the expense of the amount of basalt. At these basaltic samples there are some oxide elements that are supposed to be relatively semi-conductor elements (Al₂O₃, Fe₂O₃, TiO₂ and MgO) and there are some other oxide elements that are supposed to be relatively insulator oxides (SiO₂, CaO, Na₂O and K₂O) [17, 18, 19, 20, 21]. The decrease of the relatively semi-conductor elements (Al₂O₃, Fe₂O₃, TiO₂, MgO) in the samples is supposed (not always but generally) to decrease the conductivity. Of the other side, the decrease of the relatively insulator elements (SiO₂, CaO, Na₂O, K₂O) at samples

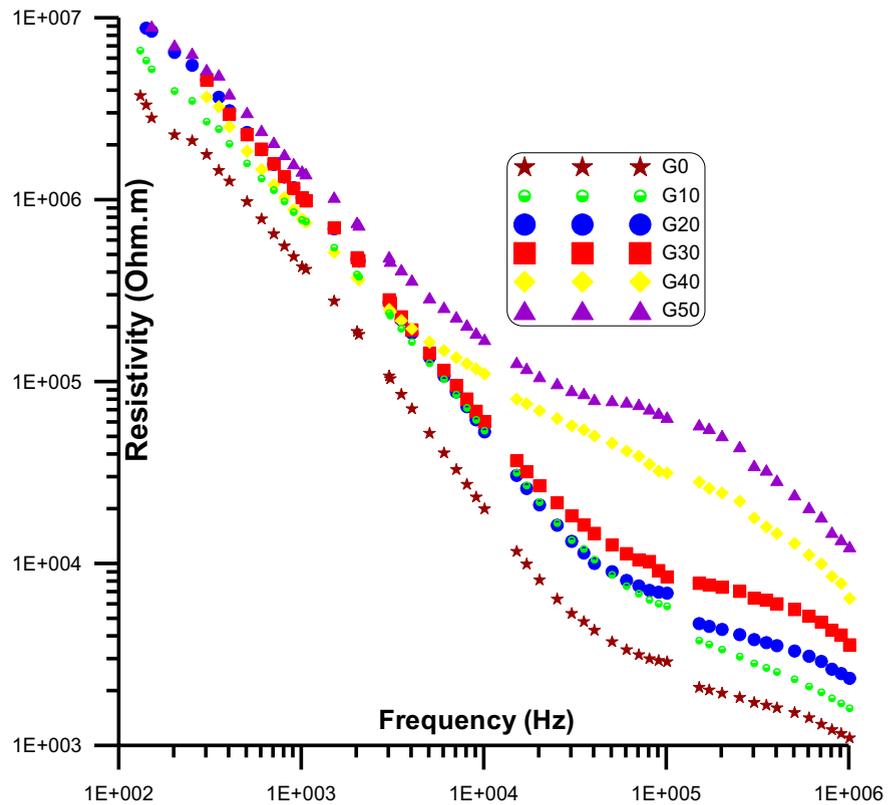


Fig. 1. Shows the resistivity versus frequency for glass sample compositions with different amounts of (CaO) and basalt.

is supposed (not always but generally) to increase the conductivity. The increase of mobility and diffusion of ions in the samples leads consequently to increase the conductivity. The different variations and combinations of concentrations of elements in the samples are responsible directly for the variation of the electrical properties in the sample [22, 23]. The highest conductivity values are found in the basalt sample (G0 with 100% of basalt) and the lowest conductivity values are found at sample G50 (with 50% of basalt). So, the decrease of the basalt in the sample decreases the conductivity.

According to samples (from G0 to G50), the main change of concentration of SiO_2 is gradually substituted or decreased (from ~ 47 to ~ 33) by increasing concentration of CaO (from ~ 8.7 to ~ 28.7), that indirectly leads to the decrease of electrical conductivity. On the other hand, the Fe_2O_3 oxide decreased from (~ 10.8 to ~ 7), Al_2O_3 oxide decreased from ~ 16 to ~ 10.5 and MgO oxide decreased from (~ 7.2 to ~ 4). The additions of those major elements in the glass structure reduce the number of cationic charge carriers that are able to move between adjacent energy levels under the effect of the electric field [24]. Generally, the addition of CaO at the expense of the basalt composition leads to decrease of the electrical properties. The different combinations of the minor and major elements at the samples may be called the

texture [25]. Each group of major and minor elements in the samples has many combinations of connections and clusters that may lead to the different electrical properties. Also, the homogeneity or heterogeneity of the sample is a very important factor after the concentration of the minor or major elements in the samples. If the sample is homogeneous, then measuring different samples of the same concentration may lead to the same electrical properties. In heterogeneous samples, samples with the same concentration do not give the same electrical properties [26]. This is a big issue in measuring electrical properties (due to the effect of texture), especially when samples are measured in the frequency range (not in the DC).

At a certain concentration, the conductivity increases with the increase of frequency. The conductivity frequency dependence is attributed to the increase of energy of particles to overcome barriers and accordingly the number of charge carriers [27].

The conductivity increases nearly with a power law dependence. All the samples have, generally, one slope (~ 0.89). Sample G0 has the highest values of conductivity; while, sample G50 has the lowest values of conductivity, see Fig. 1.

The conductivity increase with the increase of frequency until it saturates at high frequencies (from 10 kHz to 100 kHz for sample G50 and from 50 kHz to 10 MHz for sample G0). The definition of conductivity saturation value is the value when the increase of conductivity starts to be nearly zero at a certain frequency. At relatively higher frequencies (above 100 kHz for sample G50 and above 10 MHz for sample G0) the conductivity increase and saturate (Fig. 1). The conductivity dependence on frequency can be explained in terms of the increase of conducting paths that increase with the increase of frequency and with the increase of successive additions of the concentration of (CaO) at the expense of the amount of basalt. Up to a certain frequency, the conductivity will not change significantly (below 100 kHz for sample G50 and below 10 MHz for sample G0). At a certain frequency, above the previously mentioned frequencies, some of the electrons can overcome the energy barrier to make conduction that increases the conductivity again, so the damaged conducting paths become continuous. As the frequency increases, the current passing through each broken conducting path increases resulting at a general increase of conductivity values.

The substitution of Si by other Ca cation affects the mobility of the Na⁺ ions and blocks the diffusion of Na⁺ ions and the glass becomes more electrical resistive by increasing calcium silica molar ratio from G0 to G50, in addition decreasing Fe⁺², Al⁺³ and Mg⁺² cations also participating in decreasing electrical conductivity of the glass samples.

Fig. 2 shows the dielectric constant versus frequency for six glass sample compositions with a successive increase of the amount of (CaO) at the expense of the amount of basalt (Table 1). There is a general gradual decrease at the dielectric constant with

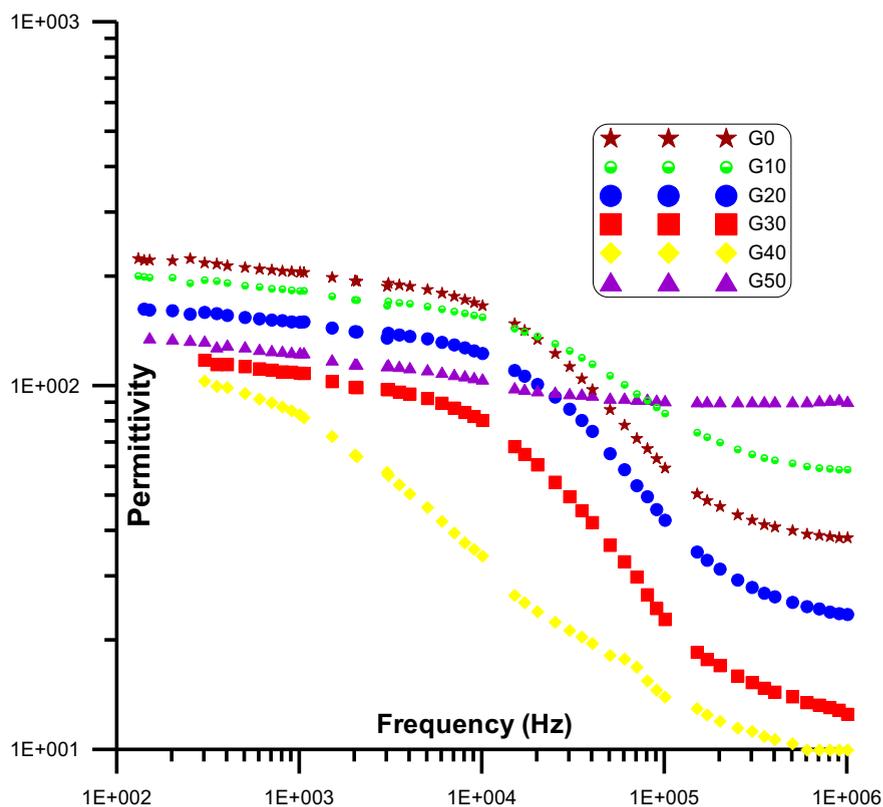


Fig. 2. Shows the dielectric constant versus frequency for glass sample compositions with different amounts of (CaO) and basalt.

the increase of frequency values. In the low-frequency region, the dielectric constant of samples has a gentle decrease response with frequency, indicating that they follow the same power law (Jonscher power law). At high-frequency region, the response of the different samples has a steep slope. All the samples nearly show the same trend or the same behavior, except sample (G50) that shows the gentle slope all over the frequency range. The difference at the behavior of the dielectric constant of sample (G50) may be resulted from the fact that the sample has reached the percolation threshold. There is no interface effect between the samples and the electrodes; that is clear from the dielectric constant slopes of the samples at relatively low-frequencies. For all the samples, the general response of the dielectric constant (at relatively high frequency) may be due to the bulk material of the sample constituents. Relative low frequencies are related directly to the interface effect between grains and between grains and electrode while, relative high frequencies are directly related to bulk material properties. The decrease of the dielectric constant values with increasing frequency can be referred to the inability of heavy charge carriers to rotate rapidly (or to follow the electric field) leading to a delay between the frequency of oscillating dipole and that of the applied field.

Fig. 3 shows the dielectric loss versus frequency for six glass sample compositions with the successive increase of the amount of CaO at the expense of the amount of basalt (Table 1). There is a general increase of the dielectric loss value with increasing frequency. In the low-frequency region, the dielectric loss of samples has a high increase response with frequency (except sample G50), indicating that the stored energy is high. Sample G50 has a low increase response with frequency due to the high dissipation energy. At higher frequency region, the response of the different samples has, nearly, the general same slope. Table 2 shows the parameters of the conductivity, dielectric constant, dielectric loss and complex impedance at some selected frequencies for the different glass samples.

Fig. 4 shows the complex impedance for six glass sample compositions with the successive increase of the amount of (CaO) at the expense of the amount of basalt (Table 1). Each point on the given curve corresponds to one frequency value, with the frequency increasing in the direction right to left. The impedance values decreases as the concentration of (CaO) is increased. All Al_2O_3 , Fe_2O_3 , TiO_2 and MgO ions are moving and contribute to the real impedance values. It is clear from the curves that the complex impedance, of all the samples, shows an insulator behavior. Nearly, there is no D. C. at all the samples. All the samples show an arc, near the imaginary

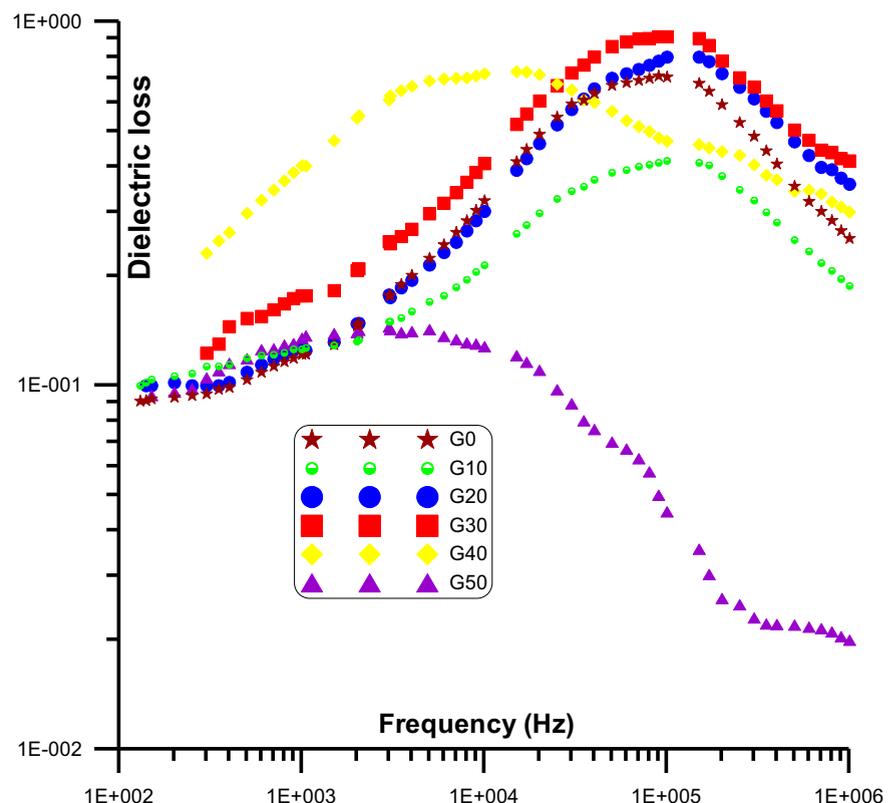


Fig. 3. Shows the dielectric loss versus frequency for glass sample compositions with different amounts of (CaO) and basalt.

Table 2. Shows the parameters of the conductivity, dielectric constant, dielectric loss and complex impedance at some selected frequencies for the different glass samples.

Glass no.	Frequency (Hz)	Real impedance (Ohm)	Imaginary impedance (Ohm)	Dielectric constant	Conductivity (S/m)	Tan δ
G0	500	4620000	4.5E+07	212.12	1.0161E-06	0.104
G0	1000	2780000	2.3E+07	205.83	2.31557E-06	0.122
G0	10000	836000	2592100	166.58	4.95453E-05	0.322
G0	100000	359000	576650	59.622	0.000343726	0.705
G0	1000000	29600	116170	38.35	0.00090157	0.254
G0	5000000	34600	14085	38.35	0.012112963	0.172
G10	500	5920000	5E+07	189.05	6.2536E-07	0.119
G10	1000	3230000	2.6E+07	182.76	1.2809E-06	0.126
G10	10000	631000	2941900	154.9	0.000018413	0.214
G10	100000	181000	498920	84.189	0.00016997	0.415
G10	1000000	14600	78017	59.022	0.00061807	0.188
G10	5000000	19600	12196	59.022	0.009285371	0.114
G20	500	6630000	6.1E+07	154.6	4.22794E-07	0.109
G20	1000	3990000	3.1E+07	150.1	9.64953E-07	0.128
G20	10000	1070000	3544700	123.14	1.86953E-05	0.301
G20	100000	513000	768870	42.844	0.000144156	0.819
G20	1000000	64100	179030	23.609	0.000423534	0.358
G20	5000000	80700	477.94	23.02	0.00384104	0.273
G30	500	1.3E+07	8.2E+07	113.25	4.35086E-07	0.153
G30	1000	7500000	4.2E+07	108.76	9.64953E-07	0.177
G30	10000	2070000	5068600	80.594	1.65031E-05	0.408
G30	100000	945000	1487400	22.86	0.000117325	0.912
G30	1000000	140000	318310	12.554	0.000276968	0.415
G30	5000000	19200	91468	12.121	0.0012635	0.365
G40	500	2.7E+07	9.2E+07	95.574	5.36152E-07	0.298
G40	1000	2E+07	4.9E+07	83.59	1.26489E-06	0.403
G40	10000	6550000	9362100	34.155	8.9808E-06	0.722
G40	100000	47000	3408000	13.992	3.15238E-05	0.471
G40	1000000	176000	452150	10.007	0.000154261	0.302
G40	5000000	18000	99784	10.007	0.000580336	0.211
G50	500	8680000	7.3E+07	128.53	3.28224E-07	0.118
G50	1000	4880000	3.8E+07	123.74	6.88278E-07	0.135

(continued on next page)

Table 2. (Continued)

Glass no.	Frequency (Hz)	Real impedance (Ohm)	Imaginary impedance (Ohm)	Dielectric constant	Conductivity (S/m)	Tan δ
G50	10000	572000	4470600	104.86	5.79711E-06	0.128
G50	100000	19700	521820	91.38	1.55229E-05	0.045
G50	1000000	1030	51506	91.021	7.97644E-05	0.021
G50	5000000	954	10471	87.22	0.00042688	0.019

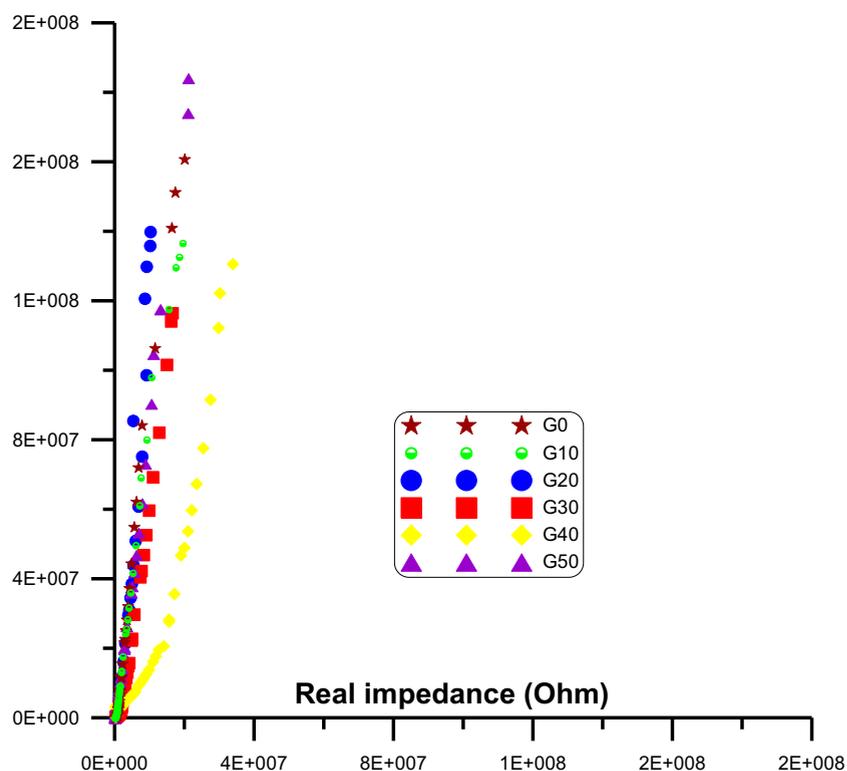


Fig. 4. Shows the complex impedance versus frequency for glass sample compositions with different amounts of (CaO) and basalt.

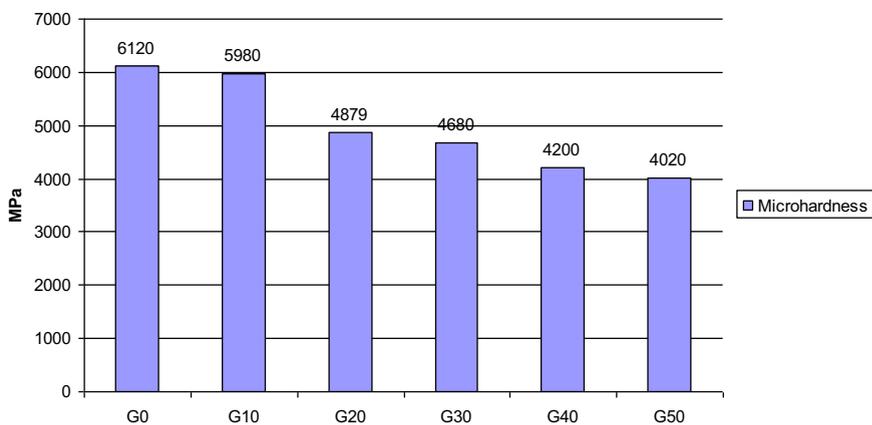
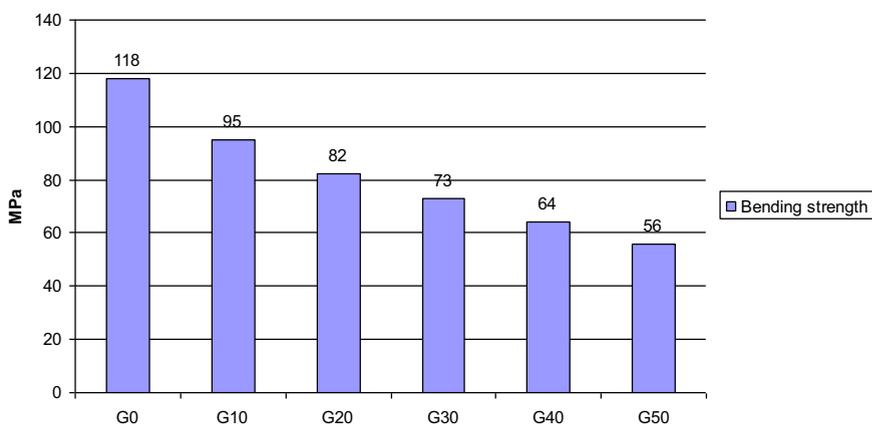
impedance axis ($\text{Im } Z$) that means that the samples are mainly insulator. The typical behavior of the samples is an arc or a part of that arc attached to a semicircle. The different variations and combinations of concentrations (CaO at the expense of the amount of basalt) in the samples did not make much change for the insulation behavior of the samples.

On the other hand, the effects of Calcia Silica molar ratio at the physical properties of the glass samples, shown in Table 3, are presented. The decreasing of calcium silica molar ratio from (0.93–0.22) corresponding to glass samples from G50 to

Table 3. Physical properties of investigated glass sample.

Glass no.	Hardness (MPa)	Bending strength (MPa)	Density (gm/cm ³)	Chemical resistance	
				HCl Weight loss	NaOH Weight loss
G0	6120	118	2.7908	zero	Zero
G10	5980	95	2.8219	0.01	Zero
G20	4879	82	2.8507	0.02	0.02
G30	4680	73	2.8788	0.07	0.04
G40	4200	64	2.9315	0.08	0.04
G50	4020	56	2.9677	0.08	0.05

G0, the bending strength and Vickers micro-hardness (shown at Figs. 5 and 6) increased from 56 to 118 MPa and from 4020 to 6120 MPa respectively and this may be attributed to presence of appropriate silica ratio in glass batches, is the main factor for keeping glass matrix has more rigidity. The density increased

**Fig. 5.** Micro-hardness of investigated glass samples.**Fig. 6.** Bending strength of investigated glass samples.

from 2.79 to 2.96 gm/cm³, due to increasing the calcia at the expense of silica in the glass batch from G0 to G50, where the density of pure calcia and silica are 3.34 and 2.64 gm/cm³, moreover increasing the weight loss of glass samples during its reaction with HCL and NaOH by increasing calcia silica molar ratio from G0 to G50.

4. Conclusion

Successive addition of bypass cement dust to basalt glass batches was applied for changing calcia silica molar ratio in the glass samples and hence its influence on electrical and physical properties of these glass samples was studied. The substitution of Si by Ca cation was found to affect the mobility of the Si ions and blocks the diffusion of Ca ions so that the glass becomes more electrical resistive by increasing calcium silica molar ratio from G0 to G50. Moreover, the additional decreasing of Fe⁺², AL⁺³ and Mg⁺² cations also participates in decreasing the electrical conductivity of these glass samples. On the other hand, bending strength and Vickers micro-hardness were affected directly by the change of calcium silica molar ratio from G0 to G50. It was found that as calcium silica molar ratio increase the mechanical properties of the glass samples decrease.

Declarations

Author contribution statement

G.A. Khater, Mohamed M. Gomaa, Junfeng Kang, M. A. Mahmoud: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Funding statement

This work was supported by the National Research Centre (No. 10060401 - "Utilization of Sinai Basaltic Rocks for the Production of Construction Materials").

Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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