Trends in

Applied Sciences Research

ISSN 1819-3579

Academic
Journals Inc.

www.academicjournals.com

Trends in Applied Sciences Research 4 (4): 176-187, 2009
ISSN 1819-3579

Utilization of Local Raw Materials for the Production of Commercial Glasses

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Abstract: The main objective of this study was to prepare transparent soda-lime-silica and borosilicate glasses utilizing locally available raw materials. Major source of oxides for the preparation of different various glass batches are silica sand, limestone, magnesite, clay, feldspar, granite and nepheline syenite. The mean chemical composition of soda-lime-silica glasses consisted of SiO_2 (70.72%), CaO (10.78%) and Na_2O (16.67%) and that of borosilicate glass consisted of SiO_2 (71.83%), Na_2O (5.30%) and B_2O_3 (13.26%). The Coefficient of Thermal Expansion (CTE) ranged from 93.64 to 110.78×10^{-7}°C for soda-lime-silica glass and from 44.85 to 73.25×10^{-7}°C for borosilicate glass in the Temperature range of 25-300°C. A strong correlation was observed between the batch and among other glass parameters such as chemical composition, melting temperature, color, density, microhardness, chemical suitability and coefficient of thermal expansion. The multivariate analysis of major oxides and the different properties of prepared glasses suggest that most of these properties depended on the composition of glasses. The properties of the prepared glasses highlighted the potential of local raw materials for glass industry in the Kingdom.

Key words: Raw materials, glass, density, microhardness, chemical durability, coefficient of thermal expansion, correlation

INTRODUCTION

Glass is a super cooled liquid prepared from the abrupt quench of a mixture of specified inorganic compounds. From the structural point of view, glass is an amorphous non-crystalline solid. Currently, a variety of glasses are available in the market. Among these, soda-lime-silica and borosilicate glasses were considered for investigation. Soda-lime-silica and borosilicate glasses are inexpensive and widely used commercially. These are characterized by versatile properties such as durability, high electrical resistivity, good spectral transmission in the visible region, high and low coefficient of thermal expansion (~100×10^{-7}/°C). These glasses are suitable for the processing plants in the chemical industry, as laboratory apparatus, ampoules and other pharmaceutical containers.

The availability and easy procurement of raw materials for producing different type of glasses are some of the major factors determining the development of economically feasible glass industry. During the present study, seven kinds of raw materials such as silica sand, limestone, magnesite, clay, feldspar, granite and nepheline syenite for producing glasses were selected from different parts of the Riyadh and Hail regions (Fig. 1). The emerging glass industry can play an important role in the economy of a country like the Kingdom of Saudi Arabia.

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The study of literature showed that the composition of the soda-lime-silica and borosilicate glasses was similar to the previously manufactured glasses which were a mixture of inexpensive batch materials such as soda ash (Na_2CO_3), lime ($CaCO_3$), silica sand and B_2O_3 with a melting point of 1400-1500°C (Varshneya, 1990). The ratio of different oxides in conventional containers, flat (soda-lime-silica glass) and other commercial glasses is Na_2O:CaO:6SiO_2 containing about 15-18% (Na_2O), 10% (CaO), 72-75% (SiO_2) and for the borosilicate glass, this ratio contains SiO_2 (55.8-78.8%), Na_2O (2-8%), Al_2O_3 (3-16%) and B_2O_3 (10-15%). The Characteristics of such glasses depend not only on the composition of raw materials but also on the processing parameters like melting condition, temperature and annealing.

In Saudi Arabia, information is very limited on different types of commercial glasses available in the market. Amjad (1998) conducted a study on the evaluation of silica sand for glass production in Riyadh. In another study, Khater and Amjad (1999) showed the effect of batch composition on the efficiency of commercial bottle.

The main objective of this study was to prepare two types of commercial glasses (Soda-lime-silica and borosilicate glasses) using local raw materials and to determine the physical and chemical properties of locally produced glasses.

MATERIALS AND METHODS

The study was carried out at King Abdulaziz City for Science and Technology (KACST), Riyadh during 2004-2007.

Raw Materials

Most of the raw materials are predominantly strata bound sedimentary rock. These are white sandstone, clay and limestone and belong to Biyadh, Dhurma and Tuwaiq Formations and Magnesite from Zarghat area near Hail Region, Saudi Arabia. The feldspar and granite were collected from Ar-Ruwaydah area along both sides of Riyadh-Makkah highway. The nepheline syenite was collected from Jabal Sawada, west of Tabuk city (Fig. 1).

Preparation of Glasses and Test Methods

Five batches of Soda-Lime-Silica Glass (SLSG) with dominant component as major oxides were prepared (Table 1). Similarly, seven batches of borosilicate glass (BSG) were prepared with dominant component as major oxides, feldspar, clay, granite and nepheline syenite using

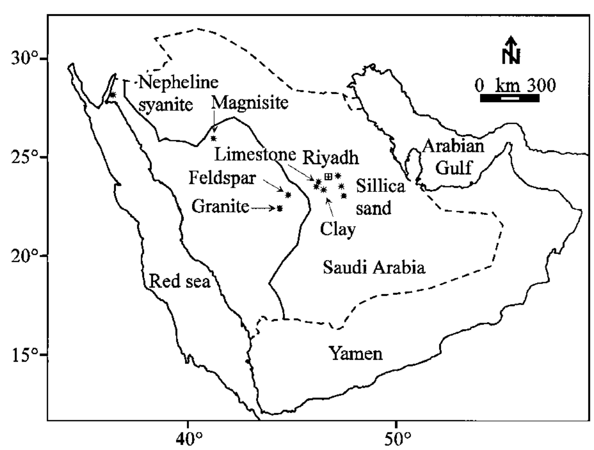

Fig. 1: Location map of raw materials

Table 1: Chemical analysis of raw materials used in soda-lime-silica glass batches

	Silica sand										
Sample No.	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	P_2O_5	LOI	Total
Major oxides (%)											
SLG-1 and 2	98.27	0.31	0.66	0.09	<0.01	0.10	0.02	0.09	0.01	0.26	99.82
SLG-3	97.04	0.55	0.41	0.73	<0.01	0.05	0.02	0.15	0.01	0.90	99.90
SLG-4	98.44	0.08	0.03	0.05	--	--	--	0.09	0.01	0.01	99.14
SLG-5	98.90	0.17	0.25	0.07	<0.01	0.04	0.02	0.07	<0.01	0.15	99.71
Limestone											
SLG-1 and 2	0.83	0.48	0.16	54.11	0.29	0.01	0.04	0.01	--	43.20	99.13
SLG-3	0.67	0.18	0.11	53.86	0.39	0.01	0.02	0.01	--	43.70	99.12
SLG-4	0.37	0.21	0.03	54.79	0.24	0.01	0.03	0.01	--	43.40	99.09
SLG-5	0.76	0.18	0.05	54.70	0.27	0.01	0.09	0.01	--	43.40	99.44
Magnesite											
SLG-2	3.27	0.09	0.03	2.78	47.19	0.03	0.04	<0.05	--	46.24	99.73

Table 2: Chemical analysis of raw materials used in borosilicate glass batches

	Silica sand										
Sample No.	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	P_2O_5	LOI	Total
Major oxides (%)											
BORO-1	99.24	0.17	0.20	0.01	<0.01	0.06	0.02	0.07	0.02	0.17	99.97
BORO-2	99.18	0.08	0.23	0.08	<0.01	0.11	0.02	0.04	<0.01	0.19	99.96
BORO-3	98.89	0.27	0.21	0.02	<0.01	0.05	0.02	0.05	<0.01	0.21	99.77
BORO-4	97.12	1.31	0.43	0.34	0.13	--	0.11	0.10	0.06	0.42	100.00
BORO-5	97.69	0.43	0.03	0.14	0.06	--	0.01	0.09	0.02	0.34	99.16
Feldspar											
BORO-1,3,4	75.20	13.04	1.41	1.19	0.03	3.50	4.75	0.08	--	0.65	99.91
Clay											
BORO-2,6,7	43.29	37.10	0.78	0.19	0.07	0.03	0.03	3.74	--	14.10	99.64
BORO-5	60.51	23.77	1.82	0.22	0.24	0.44	0.30	1.77	--	10.80	100.00
Granite											
BORO-2	64.81	19.29	0.29	0.35	0.07	2.91	11.68	0.02	--	--	99.42
Nepheline syenite											
BORO-4	53.32	20.16	7.04	1.68	0.10	7.32	5.17	0.15	--	3.74	99.08

the locally available raw materials (Table 2). The glasses were prepared by melting each batch material (starting powder) in a sintered alumina crucible in the temperature range of 1350-1550°C. The glass melts were poured into steel mould and patties annealed at 650°C in the pre-heated muffle furnace. The chemical analysis of the raw materials and the prepared glasses samples was carried out in ALS Chemex Lab. (Canada).

The density of glasses was determined by Archimedes methods. The microhardness of glasses was determined using a Vicker's microhardness indenter (Digital Micro hardness Tester; MHV2000). There were three replications of each batch samples. The mean values of three random measurements for each sample with a load of 200 g and a time of 10 sec were presented for evaluation.

The chemical durability of glass was obtained by the attacking rate of water, pH of solution, the total volume of solution in contact with the glass, solution concentrations and the glass composition. The Coefficient of Thermal Expansion (CTE) of glass samples (dimension 0.5×0.5×2.0 cm) was measured using Netzsch dilatometer (DIL 402PC, Germany) at a heating rate of 5°C min^{-1}.

RESULTS AND DISCUSSION

Chemical Composition

The chemical and physical properties such as density, microhardness, chemical durability and coefficient of thermal expansion) of soda-lime-silica and borosilicate glasses varied with the change in batch composition (Table 3, 4).

Table 3: Batch, chemical composition and properties of soda-lime-silica glasses

Sample No.	Batch constituent	Chemical composition of glass (%)	Descriptions	Microhardness (kg mm^{-2})	Density (g cm^{-3})	Chemical durability wt. loss (%) H_2O	Chemical durability wt. loss (%) HCl (0.1 N)	CTE α $\times 10^{-7}$ °C^{-1} (25-300°C)
SLG-1	Silica sand	SiO_2: 71.0	Colour: Colourless	561	2.400	0.19	0.03	98.985
	Limestone	Na_2O: 16.0	Transparency :Transparent					
	Na_2CO_3 *	Al_2O_3: 0.32	Melting Temp. (°C): 1450					
		MgO: 0.01	Mould State: Bubble Free					
		CaO: 10.6	Annealing Temp. (°C): 700					
		Fe_2O_3: 0.34	Devitrification: none					
SLG-2	Silica sand	SiO_2: 70.0	Colour: Light green	473	2.413	0.43	0.03	102.35
	Limestone	Na_2O: 16.45	Transparency :Transparent					
	Magnesite	Al_2O_3: 1.39	Melting Temp. (°C): 1450					
	Na_2CO_3 *	MgO: 2.18	Mould State: fine bubbles					
		CaO: 9.67	Annealing Temp. (°C): 500					
		Fe_2O_3: 0.35	Devitrifiction: none					
SLG-3	Silica sand	SiO_2: 70.0	Colour: Light Blue	502	2.413	0.26	0.10	110.78
	Limestone	Na_2O: 17.2	Transparency :Transparent					
	Na_2CO_3 *	Al_2O_3: 1.71	Melting Temp. (°C): 1350					
		MgO: 0.13	Mould State: no bubbles					
		CaO: 10.75	Annealing Temp. (°C): 540					
		Fe_2O_3: 0.17	Devitrification: none					
SLG-4	Silica sand	SiO_2: 68.10	Colour: Colourless	528	2.456	0.09	0.05	93.64
	Limestone	Na_2O: 17.75	Transparency :Transparent					
	Na_2CO_3	Al_2O_3: 2.48	Melting Temp. (°C): 1350					
		MgO: 0.09	Mould State: no bubbles					
		CaO: 11.65	Annealing Temp. (°C): 540					
		Fe_2O_3: 0.06	Devitrifiction: none					
SLG-5	Silica sand	SiO_2: 74.00	Colour: Colourless	419	2.445			103.287
	Limestone	Na_2O: 15.97	Transparency :Transparent					
	Na_2CO_3 *	Al_2O_3: 3.39	Melting Temp.: (°C): 1350					
		MgO: 0.01	Mould State: no bubbles					
		CaO: 11.22	Annealing Temp.: (°C): 540					
		Fe_2O_3: 0.10	Devitrification: none					

*Soda Ash

The chemical analysis of raw materials for different batches of soda-lime-silica and borosilicate glasses is given in Table 1 and 2. The batch of raw materials for soda-lime-silica glass contained silica sand as SiO_2 (97.04-98.27%), limestone as CaO (53.86-54.79%), limestone as MgO (0.24-0.39%) and magnesite as MgO (47.19%) (Table 1). The batch of borosilicate glasses contained silica sand as SiO_2 (97.12-99.24%); clay as SiO_2 (43.29-60.51%), Al_2O_3 (23.77-37.1%), feldspar as SiO_2 (75.20%), Al_2O_3 (13.04%), Na_2O (3.5%), K_2O (4.75%), granite as SiO_2 (64.81%), Al_2O_3 (19.29%), Na_2O (2.91%), K_2O (11.68%) and Nepheline Syenite [SiO_2 (53.32%), Al_2O_3 (20.16%), Na_2O (7.32%), K_2O (11.68%), Fe_2O_3 (7.04%)] (Table 4). The melting point of glasses varied from 1350 to 1550°C (Table 3, 4). Bourne (1994) stated that the higher melting point of glass is due to the presence of iron and transition elements which impart color. In the present study, most probably iron played a significant role in coloring the glasses. The upper limit of chrome in the colorless glass is 0.0005%.

All the samples of soda-lime-silica and borosilicate glasses were transparent and showed the uniformity of composite batch. The chemical composition of prepared soda-lime-silica glasses was SiO_2 (68.10-74.00% with an average value of 70.62%), CaO (9.67-11.85% with an average value of 10.78%), Na_2O (15.97-17.75% with an average value of 16.67%, Al_2O_3 (0.32-3.39% with an average value of 1.86%), MgO (0.01-2.18% with an average value of 0.48%) and Fe_2O_3 (0.06-0.35 with an average value of 0.20%) (Table 3). The chemical components of borosilicate glasses were SiO_2 (55.8-78.8% with an average value of 71.82%), Na_2O (3.50-8.14% with an average value of 5.30%), Al_2O_3\92.69-15.6% with an average value

Table 4: Batch, chemical composition of glass and properties of borosilicate glasses

Sample No.	Batch composition (Raw materials)	Chemical composition of glass (%)	Descriptions	Microhardness ($kg\ mm^{-2}$)	Density ($g\ cm^{-3}$)	Chemical durability wt. loss (%)		CTE α $\times 10^{-7}$ °C^{-1} (30-300°C)
						H_2O	HCl 0.1 N	
BORO-1	Silica sand	SiO_2: 78.80	Colour: Colourless	499	2.127	0.06	0.10	43.65
	Feldspar	Na_2O: 4.29	Transparency :Transparent					
	Boric acid	Al_2O_3: 3.88	Melting Temp. (°C): 1550					
	Na_2CO_3	K_2O: 2.22	Mould State: no bubbles					
		B_2O_3: 11.90	Annealing Temp. (°C): 450					
			Devitrification: none					
BORO-2	Silica sand	SiO_2: 77.40	Colour: Colourless	542	2.021	0.55	0.09	46.69
	Granite	Na_2O: 3.50	Transparency :Transparent					
	Boric acid	Al_2O_3: 2.69	Melting Temp. (°C): 1550					
	Na_2CO_3	K_2O: 3.02	Mould State: no bubbles					
		B_2O_3: 14.10	Annealing Temp. (°C): 450					
			Devitrification: none					
BORO-3	Silica sand	SiO_2: 74.20	Colour: Light green	438	2.194	0.39	0.19	52.03
	N Syanite:	Na_2O: 5.38	Transparency :Transparent					
	Boric acid	Al_2O_3: 4.04	Melting Temp. (°C): 1550					
	Na_2CO_3	K_2O: 2.63	Mould State: fine bubbles					
		B_2O_3: 14.70	Annealing Temp. (°C): 450					
			Devitrification: none					
BORO4	Silica sand	SiO_2: 77.20	Colour: Light blue	503	2.149	0.26	0.11	40.06
	Feldspar	Na_2O: 4.15	Transparency :Transparent					
	Boric Acid	Al_2O_3: 6.38	Melting Temp. (°C): 1550					
	Na_2CO_3	K_2O: 2.30	Mould State: no bubbles					
		B_2O_3: 11.60	Annealing Temp. (°C): 450					
			Devitrification: none					
BORO-5	Silica sand	SiO_2: 71.40	Colour: Colourless	521	2.303	0.15	0.10	53.93
	Clay	Na_2O: 5.30	Transparency :Transparent					
	Boric Acid	Al_2O_3: 7.02	Melting Temp. (°C): 1550					
	Na_2CO_3	K_2O: 2.76	Mould State: fine bubbles					
	KNO_3	B_2O_3: 14.4	Annealing Temp. (°C): 450					
			Devitrification: none					
BORO-6	Silica sand	SiO_2: 68.00	Colour: Colourless	476	2.245	0.23	0.20	55.60
	Clay	Na_2O: 6.36	Transparency :Transparent					
	Boric Acid	Al_2O_3: 8.35	Melting Temp. (°C): 1550					
	Na_2CO_3	K_2O: 3.41	Mould State: no bubbles					
	KNO_3	B_2O_3: 14.40	Annealing Temp. (°C): 450					
			Devitrification: none					
BORO-7	Silica sand	SiO_2: 55.80	Colour: Light smoky	493	2.328	1.54	0.42	69.68
	Clay	Na_2O: 8.14	Transparency :Transparent					
	Boric Acid	Al_2O_3: 15.60	Melting Temp. (°C): 1550					
	Na_2CO_3	K_2O: 2.67	Mould State: with bubbles					
	KNO_3	B_2O_3: 11.70	Annealing Temp. (°C): 450					
			Devitrification: none					

of 6.85%) and B_2O_3 (11.60-14.40% with an average value of 13.26%) as given in Table 4. The percentage of SiO_2 in the prepared and standard commercial borosilicate glass was almost similar. The Na_2O contents showed fluctuation in the borosilicate glasses due to the difference in the amount of pure analytical grade Na_2CO_3 in the batches. The fluctuation in Al_2O_3 contents could be due to different natural source of Al_2O_3 in the batches (feldspar = 13.04% and clay = 23.77-37.1%) as shown in Table 1 and 2. The presence of MgO in batch sample (SLG-2; 2.18%) did not play any significant role on physical properties of the glass as the soda-lime-silica glasses contained high contents of soda than lime. Because this reaction consumed all the lime from limestone and magnesite thus resulting the magnesium carbonate as free to decompose at the Na_2CO_3-$CaCO_3$ eutectic temperature (Roth *et al*., 1981). The chemical analysis of standard soda-lime-silica and borosilicate glasses showed that the composition of prepared glasses was almost identical (Table 3, 4). Although, physical difference between both the standard and prepared glasses was small but it was more

pronounced on other properties of glasses which could be attributed to variation in the chemical composition of raw materials.

Density

Density is the most important property of glass and provides information about the constitution or structure of glass. The density of soda-lime-silica glasses ranged from 2.40-2.45 g cm^{-3} with an average value of 2.42 g cm^{-3} and that of borosilicate glasses ranged from 2.02-2.33 g cm^{-3} with an average value of 2.19 g cm^{-3} (Table 3, 4). These results agree with the results of Morey and Merwin (1932), Konijnendijk (1971) and Imoka *et al.* (1971).

The regression analysis between the density and the chemical composition of borosilicate glass showed negative correlation between SiO_2 and density (r = -0.62), positive correlation between Na_2O and density (r = 0.74) as well as between Al_2O_3 and density (r = 0.63) (Fig. 2) and a positive correlation between density and Na_2O+Al_2O_3 (r = 0.6012) in the borosilicate glasses (Fig. 3). The other published work shows that in borosilicate glass, especially in the binary-component system (B_2O_3-SiO_2), the density increased with decrease in B_2O_3 contents (Jabra *et al.*, 1980). Whereas, in the multi component system (Na_2O-B_2O_3-SiO_2), the effect of composition on density was not very clear. The correlation showed that density increased with the corresponding increase of Na_2O+Al_2O_3 contents in the soda lime glass (Fig. 3). These findings are in agreement with study of Khater and Amjad (1999).

The difference in the density of different type of glasses may be attributed to the water contents in addition to temperature and other factors (Volf, 1990). Bruckner and Navarro (1966) stated that the density increases on the sample cooled from the sample melted at temperature up to 1500°C and decreases with that melted at temperature above 1500°C. But in the present study, the density of soda-lime-silica glasses decreased following the decrease

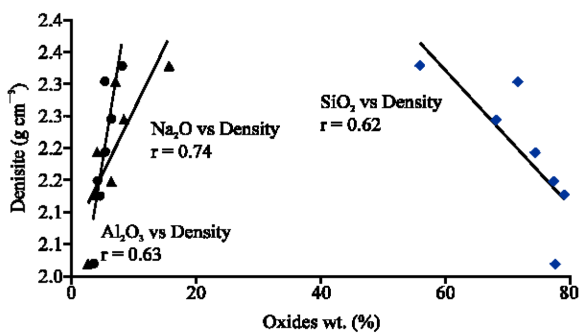

Fig. 2: Relationship between oxides and density of borosilicate

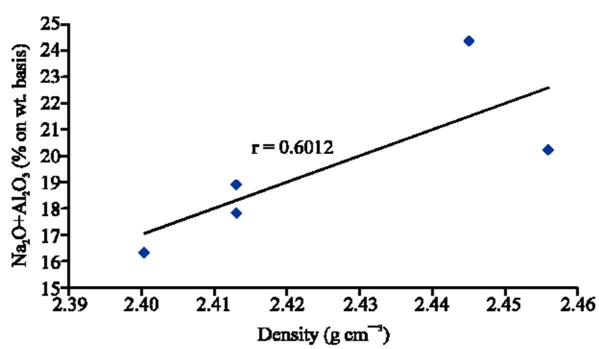

Fig. 3: Relationship between density and oxides of soda lime silica glass

in the melting point (SLG-5, 1350°C, density 419 g cm^{-2}) and increased at a higher melting point (SLG-1, 1450°C, density 561 g cm^{-2}). There was no relationship between melting point and density of borosilicate glasses as the melting point remained constant during the process (1550°C) but a fluctuation in the density was observed which could be subjected to many other unknown factors.

Microhardness V'H

The hardness is a complicate mixture of resistance to fracture and elastic to plastic deformation. Microhardness varies from one type to another type of glass. Microhardness is a bond sensitive property (Zarzycki, 1991) which provides an insight on the nature of the chemical bonding in a material. The soda-lime-silica glass is the hardest. The microhardness of soda-lime-silica glasses ranged from 419-561 kg mm^{-2}, with an average value of 496.6 kg mm^{-2} (Table 3) and that of borosilicate glasses varied between 438 and 542 kg mm^{-2} with an average value of 496 kg mm^{-2} (Table 4). Ainsworth (1954) studied the microhardness of Na_2O-CaO-SiO_2 and Na_2O-B_2O_3-SiO_2 glasses which ranged from 487-620 and 478-591 kg mm^{-2} respectively in different batches. The study of Imoka *et al*. (1971) supported the microhardness values of both types of glasses prepared in the present study. However, Varshneya (1990) showed the microhardness value was 460 kg mm^{-2} for Na_2O-CaO-SiO_2 glass) and was 480 kg mm^{-2} for Na_2O-B_2O_3-SiO_2 glass.

The correlation between microhardness and SiO_2 contents of soda-lime-silica glasses was negative and very poor (r = 0.4258) (Fig. 4), positive between microhardness and CaO (r = 0.640), while it was negative between microhardness and Na_2O (r = -0.621) (Fig. 5). Also, the correlation was negative and poor between microhardness vs Na_2O/CaO+MgO (r = -5497) (Fig. 6).

The V'H value of borosilicate glass is generally controlled by the glass compositions. Any increase in the V'H value of the glasses is followed by an increase in the SiO_2 contents. Nassar and Adawi (1981) stated that Al_2O_3 concentration in the glass composition plays an important role with the microhardness and an increase of Al_2O_3 lead to an increase of non-bridging oxygen ions. However, such an increase in the concentrations of non-bridging oxygen reduces the connectivity of the glassy network and elastic modules of the glass (Shelby, 1997).

Chemical Durability

The chemical durability of soda-lime-silica glasses ranged from 0.03-0.10% on weight basis (Table 3). The chemical durability was highest (0.1%) in SLG-3 glass containing 17.2%

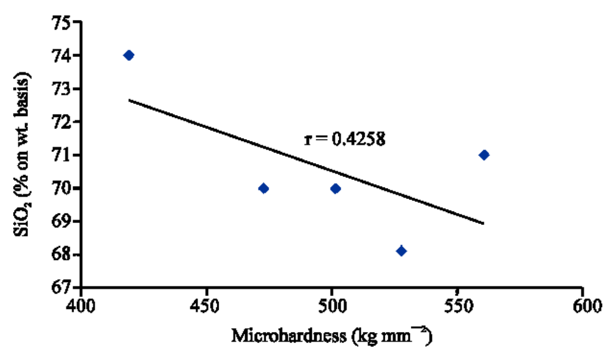

Fig. 4: Relationship between microhardness and SiO_2 of soda lime silica glasses

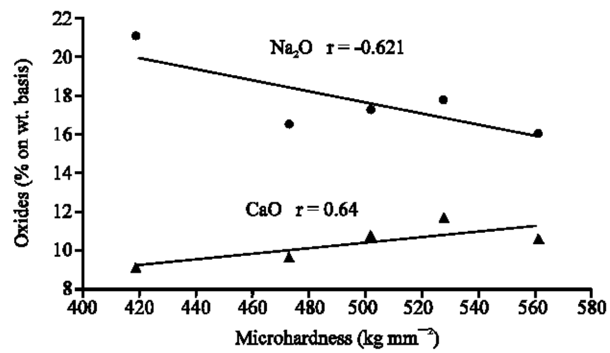

Fig. 5: Relationship between microhardness and Na_2O and CaO of soda lime silica glasses

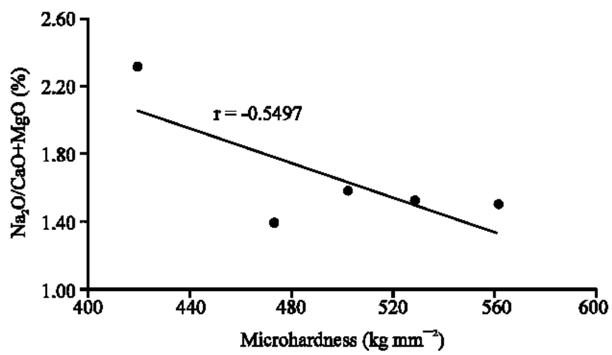

Fig. 6: Relationship between microhardness and Na_2O/CaO+MgO of soda lime silica glass

Na_2O and lowest (0.03%) in SLG-1 and SLG-2 glass containing 16.0 and 16.45% Na_2O, respectively. The high contents of Na_2O decreased glass chemical durability. Whereas, high durability was achieved with low concentration of Na_2O and Al_2O_3 (Table 3). This is in agreement with the results of Sinton *et al.* (2001) who found that Na_2O decreases chemical durability. The chemical durability of the borosilicate glasses varied with the batch composition. The weight losses were 0.06-1.54% (on weight basis) in water and 0.09-0.19% (on weight basis) in 0.1 N HCl. The difference in percent weight loss was more in water than 0.1 N HCl solution. Overall, percent weight loss was higher in borosilicate glasses than soda-lime silica glasses. Similarly, the chemical durability of borosilicate glasses was much lower than soda-lime-silica glasses and the weight loss was higher in water that in 0.1 N HCl solution.

Coefficient of Thermal Expansion (CTE) of Glasses

The values of CTE of the prepared soda-lime-silica glasses ranged from $93.64\text{-}110.78\times10^{-7}$°C^{-1} with an average value of 101.8×10^{-7}°C^{-1} in the temperature range of 25-300°C (Table 3, Fig. 7). The correlation was poor and negative between CTE and CaO (r = -0.25) (Fig. 8) and between CTE and microhardness of the soda-lime-silicate glasses (r = -0.383) (Fig. 9). Comparison between standard, Physical and chemical characteristics of prepared soda-lime-silica glasses is shown in Table 5.

The data showed that majority of samples in soda-lime-silica glasses of multi component system did not show any significant relationship with its batch composition. Other experimental studies on soda-lime-silica glass showed that an increase in alkaline oxides such as CaO and MgO increased the CTE (Rawson, 1980).

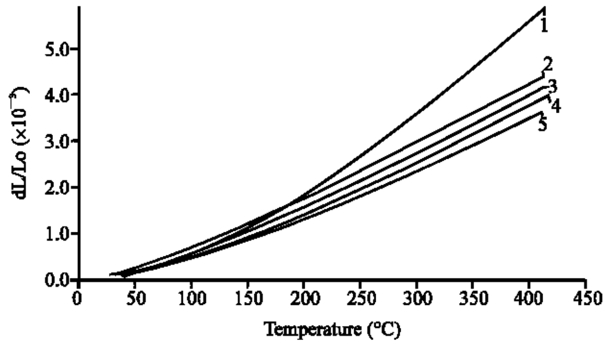

Fig. 7: Thermal expansion curves of soda lime silica glasses. 1: SLG-1, 2: SLG-2, 3: SLG-3, 4: SLG-4, 5: SLG-5

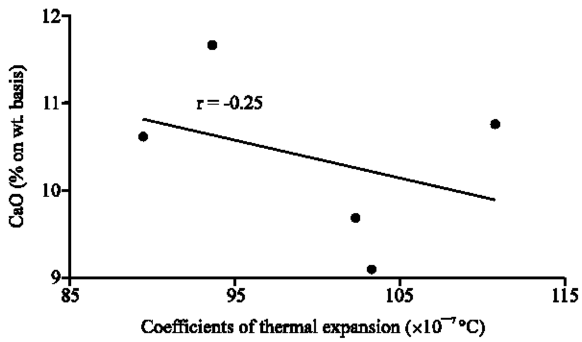

Fig. 8: Relationship between CTE and CaO of soda lime silica glass

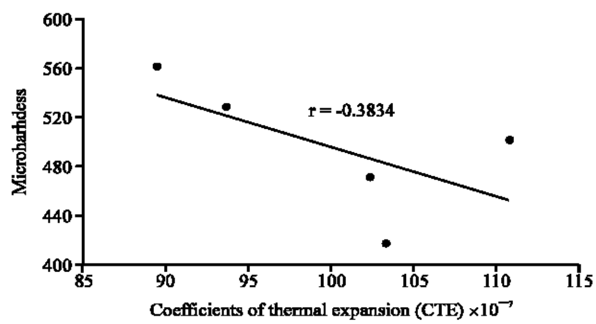

Fig. 9: Relationship between CTE and microhardness of soda lime silica glasses

Table 5: Comparison between standard and physical and chemical characteristics of prepared soda-lime-silica glasses

Properties	Standard	SLG-1	SLG-2	SLG-3	SLG-4	SLG-5
Chemical (wt. %)						
SiO_2	72.0	71.00	70.00	70.00	68.10	74.00
Na_2O	16.0	16.00	16.45	17.20	17.75	15.97
Al_2O_3	2.0	0.32	1.39	1.71	2.48	3.39
CaO	9.3	10.60	9.67	10.75	11.65	11.22
Physical						
CTE $\times 10^{-7}$ cm^2 (°C) (25-300°)	92-100 (50-350°C)	98.98	102.35	110.78	93.64	103.29
Density (g cm^{-3})	2.31-2.52	2.40	2.41	2.41	2.45	2.45
Annealing point (°C)	554°C	700.00	500.00	540.00	540.00	540.00
Microhardness (kg mm^{-2})	500-600	561.00	473.00	502.00	528.00	419.00

*US Patent (2003)

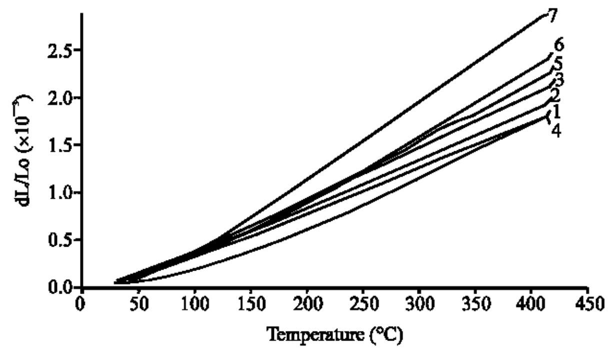

Fig. 10: Thermal expansion curves of borosilicate glasses. 1: Boro-1, 2: Boro-2, 3: Boro-3, 4: Boro-4, 5: Boro-5, 6: Boro-6, 7: Boro-7

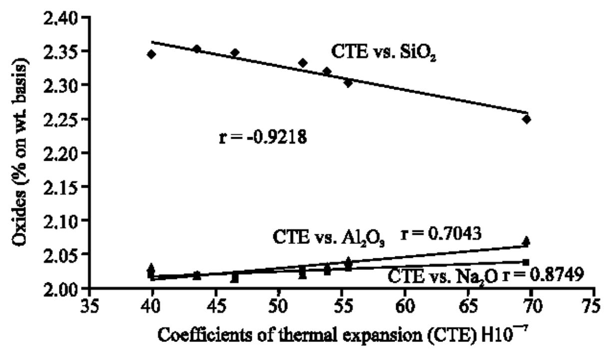

Fig. 11: Relationship between CTE and SiO_2, Al_2O_3 and Na_2O of borosilicate glasses

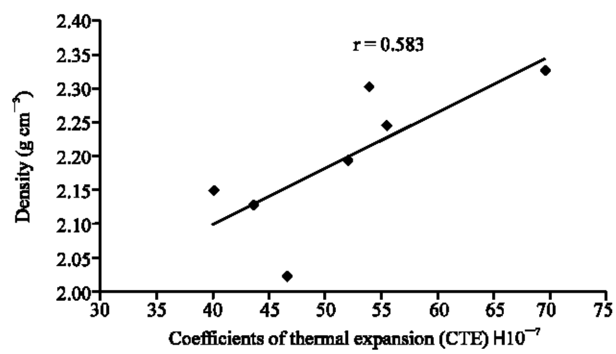

Fig. 12: Relationship between CTE and density of sodium borosilicate glasses

The CTE values of borosilicate glasses ranged from $44.85\text{-}73.25\times10^{-7}$°$C^{-1}$ with an average value of 54.29×10^{-7}-°C^{-1} (Table 4, Fig. 10). Moreover, the CTE curves showed the familiar pattern which are characteristics of normal silicate glasses. The correlation was negative between CTE and SiO_2 (r = -0.9218) and positive between CTE and Na_2O (r = 0.8749) as well as between CTE and Al_2O_3 contents (r = 0.7043) (Fig. 11). The correlation was positive between CTE and density of glasses (r = 0.5838) (Fig. 12). Comparison between standard, Physical and chemical characteristics of prepared borosilicate glasses is shown in Table 6.

Table 6: Comparison between standard and physical and chemical characteristics of prepared borosilicate glasses

Properties	Standard ASTM (2006)	BORO1	BORO2	BORO3	BORO4	BORO5	BORO6	BORO7
Chemical								
SiO_2	81	78.80	77.40	74.20	74.20	71.40	68.00	55.80
B_2O_3	12.5	11.90	14.10	14.70	14.70	14.40	14.40	11.70
Al_2O_3	12.5	11.90	14.10	14.70	14.70	14.40	14.40	11.70
Na_2O	4.5	4.29	3.50	5.38	5.38	5.30	6.36	8.14
Physical properties								
CTE X10^{-}m^{20}C (25-300°)	32-33	44.85	47.88	52.83	45.02	56.01	60.19	73.25
Density g cm^{-3}	2.53	2.13	2.02	2.19	2.149	2.30	2.24	2.33
Annealing point°C	560	450.00	450.00	450.00	450.00	450.00	450.00	450.00
Microhardness kg mm^{-2}	488-512	499.00	542.00	438.00	503.00	521.00	476.00	493.00

In the two component system (B_2O_3-SiO_2) and multicomponent system (Na_2O-B_2O_3-SiO_2), the CTE increases with an increase in B_2O_3 % on weight basis (Cousen and Turner, 1928; Mazurin *et al*., 1969). The above statement supported the present findings because in this case, the sample (BORO-1) contains only 11.09% B_2O_3 and its coefficient of thermal expansion is 44.85×10^{-7}C, but when the B_2O_3 % increased to 14.40 in the sample (BORO-5) then the CTE increased to 56.01×10^{-7}C. The results of the study strongly agree with the findings of Kurkjian (1963) and Mazurin *et al*. (1969) who stated that increasing the contents of Na_2O increases the CTE values of borosilicate glasses. The above stated results showed a great potential for the production of commercially viable soda-lime-silica and borosilicate glasses from the local raw materials. The properties of the prepared glasses were comparable to the properties of known standard glasses. It is further mentioned that the consistency of chemical composition of the raw materials is very important because any change will affect the physical and chemical properties of glasses. Overall, the study findings suggest that local raw materials are suitable for the production of glasses in a local glass industry and can be exported to other countries from the Kingdom. Besides this, standardization of glass batch composition is vital for the production of commercially viable soda-lime-silica and borosilicate glasses in Saudi Arabia.

CONCLUSION

Soda-lime-silica and borosilicate glass batches were prepared from locally available raw materials such as silica sand, limestone, magnesite, clay, feldspar, granite and nepheline syenite. The chemical composition of the final prepared soda-lime-silica glasses composed of SiO_2 (68.10-74.00% with an average value of 70.62%), CaO (9.67-11.85% with an average of 10.78%), Na_2O (15.97-17.75% with an average value of 16.67%, Al_2O_3 (0.32-3.39% with an average value of 1.86%), MgO (0.01-2.18% with an average value of 0.48%) and Fe_2O_3) 0.06-0.35 with an average value of 0.20%) as given in Table 3. The chemical components of borosilicate glasses were SiO_2 (55.8-78.8% with an average value of 71.82%), Na_2O (3.50-8.14% with an average value of 5.30%), Al_2O_3 (92.69-15.6% with an average value of 6.85%) and B_2O_3 (11.60-14.40% with an average value of 13.26%). The slight variation in the standard and prepared glass composition amounted to more than 80% and is related to the composition of raw materials and the glass batch preparation from the natural raw materials. The composition and physical properties of prepared glasses showed mainly positive correlation with the batch composition. In conclusions, the locally prepared borosilicate glasses seem to be commercially more viable than the soda-lime-silica glasses when considering its usefulness.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the financial support and laboratory facilities of the King Abdulaziz City for Science and Technology (KACST) during the research project BM-24-01. We thank Prof. Esmat Hamzawy, National Research Center-Egypt, the Project Consultant) for his valuable comments. Thanks are extended to Ma' aden Company for providing Nepheline Syenite sample.

REFERENCES

Ainsworth, L., 1954. A single crystal X-ray analysis of Mg vermiculite. J. Soc. Glass Technol., 38: 501-501.

Amjad, M.A., 1998. Evaluation of Riyadh white silica sand for glass production. Glass Technol., 39: 105-110.

Bourne, L.H., 1994. Glass raw materials. Ind. Rocks Minerals, 293: 62-62.

Bruckner, R. and J. Navarro, 1966. Physical and chemical investigation in the preparation of high silica glasses. Glastechnolo. Bull., 39: 283-283.

Cousen, A. and W.E.S. Turner, 1928. A study of the glasses boric oxide-silica. J. Soc. Glass Technol., 12: 169-169.

Imoka, M., H. Hasegawa, Y. Hamaguchi and Y. Kurotaki, 1971. Refractive index and Abbe's number of glass of Lanthenum borate system. Yogyo Kyokaishi, 5: 164-164.

Jabra, R., J. Pelous and J.J. Phalippou, 1980. Brillouin scattering measurements of attention and velocity of hyper sounds in SiO_2-B_2O_3 glasses. Non-Crystalline Solids, 37: 349-358.

Khater, G.A. and M.A. Amjad, 1999. Effect of Glass batch composition on the efficiency of bottle production. J. King Saud Univ. Sci., 11: 35-42.

Konijnendijk, W.L., 1971. The structure of borosilicate glasses. Thesis, Eindhoven, Netherlands.

Kurkjian, C.R., 1963. Relaxation of torsional stress in the transfor-mation range of a soda-lime-silica glass. Physics chem. Glasses, 4: 128-136.

Mazurin, O.V., A.S. Tatesh and M.V. Strel'tsina, 1969. Glass formation and stability of inorganic Glasses (in Russian). Sklar Keram, 19: 100-100.

Morey, G.W. and H.E. Merwin, 1932. The relation between the composition and the density and optical properties of glass. J. Optical Soc. Am., 22: 632-662.

Nassar, A.M. and M.A. Adawi, 1981. The role of Al^{3+} ions in alumino borate Glasses as revealed by molar volume, refractive index and micro hardness. J. Crystalline Solids, 50: 155-161.

Rawson, H., 1980. Properties and Applications of Glass. Glass Science and Technology 3, Elsevier, Oxford-New York.

Roth, R.S., T. Negas and L.P. Cook, 1981. In Phase Diagram for Ceramists. 4: The American Ceramic Society, Columbus, Ohio.

Shelby, J.E., 1997. Introduction to Glass Science and Technology. RSC, UK.

Sinton, C.W. and W.C. LaCourse, 2001. Experimental survey of the chemical durability of commercial soda-lime silicate glasses. Materials Res. Bull., 36: 2471-2479.

Varshneya, A.K., 1990. Strength of Glass. In: Chemistry of Glasses, Paul, A.D.E. (Ed.), 2nd Edn., Chapman and Hall, London, New York.

Volf, B.M., 1990. Technical Approach to Glass. Glass Science and Technology, 10, New York.

Zarzycki, J., 1991. Glasses and Amorphous Materials. In: Materials Science and Technology: A Comprehensive Treatment, Cohn, R.W., P. Haasan and E.J. Karmer (Eds.). Vol. 9, VCH, Weinheim, New York.