Review Paper

Evolution of the compositions of commercial glasses 1830 to 1990. Part II. Container glass

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This paper treats container glasses in the same way as an earlier paper reviewed flat glass compositions. Data for more than 970 analyses of container glasses manufactured during 150 years in 37 countries have been collected and analysed. The data obviously include containers made by hand as well as by machine from green, white and amber glasses. Compositions developed along two lines, one being green and brown glasses coloured by manganese; such glasses were characterized by low alkali content compensated by increased RO and higher Al_2O_3 contents. The other, younger, line includes white glasses historically descended from old "forest glasses" and closely related amber glasses. The compositions of both lines converged gradually, so that from about 1970 all container glasses have fallen within a narrow range of compositions, differing in little but their colouring oxides. Differences between particular producers and countries have decreased a great deal.

1. Introduction

A previous paper [1] dealt with the evolution of the compositions of flat glasses. The development of container glass compositions ran in parallel but separately. Keppeler [2 and 3] and Keppeler et al. [4 and 5] recorded this development from 1920 to 1945. The compositions of American container glasses from 1932 to 1978 were reviewed by Loesell et al. [6] and Stadler et al. [7]. The history of champagne bottle compositions was discussed by Chopinet [8 and 9] and other authors [10 and 11]. This paper describes the development of compositions of container glass, seeking trends in composition and differences between countries.

Container glasses are traditionally divided according to colour: green glasses, brown glasses coloured by manganese, amber glasses, and flint glasses.

2. Research methodology

The work was based on statistical analysis of nearly 1000 compositions in the way already used for flat glasses [1]. Only glass compositions determined by analysis were included in the data base. The compositions of older glasses (up to 1937) were taken from Thiene [12], their sources often being now difficult to recover.

Analyses of Czech glasses and many from other countries were available from the archives of the Mühlig company [13] and in the last half century from the archives of Czech research institute VÚSU [14]. Data relevant to development of high-alumina container glasses [15 and 16] came also from VÚSU. Analyses of container glass from recent years were taken from the author's work [17 and 18]. Russian compositions were obtained from the data of Kutateladze [19], Andrukhina [20], Katkova [21], and Gvazava [22]. Some analyses were obtained from other sources [23 to 28]. Altogether 972 analyses of container glasses were collected: 361 flint, 37 brown (manganese), 124 amber, and 449 green. When numerous results were available for one glass over a period when its composition was not deliberately changed, only one average result was used.

The analyses were classified by country of origin for each colour and they were ordered chronologically. The set for each country was divided into periods of similar compositions. These groups generally covered 3 to 10 analyses of glasses made in a period of 5 to 10 years. Average composition and standard deviation were calculated for each group for further statistical analysis. This method of so-called "data nests" decreases the effect of extreme samples as well as of analytical errors and inexact glass dating. Tables 1 to 5 show compositions of particular groups; *n* is the number of analyses within the group, which is underlined in the case when the analyses are averages of a sequence of operational data. RO = CaO+MgO+BaO, R_2O = Na_2O+K_2O, and Dietzel's sum is $RO+R_2O$ [29]. All data are given in weight percentages. Where no K_2O content is given, a small amount of potassium may be included in the Na_2O content. In older analyses Al_2O_3 may include Fe_2O_3.

Figures 1 to 3 show the development with time of the basic types of glass. The average composition of each group

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is plotted at the middle of the relevant period and a line drawn to represent the trends of these averages. To assist numerical evaluation of changes with time, averages of all groups from all countries were calculated for each type of glass and also for selected longer periods regardless of country of production. The periods generally chosen were:

a) up to 1835 – Glassworks had not yet become specialized; container glass production was a part of hollow ware production, glass was melted in pots and analyses were not very reliable;

b) 1840 to 1880 – production had become specialized; glass was predominantly melted in pots and analyses had become more complete and more accurate;

c) 1880 to 1915 – hand production still prevailed but the first machines emerged and melting in tank furnaces gradually prevailed; analyses were usually reliable;

d) 1919 to 1939 – production was almost entirely by machines and all glasses melted in tanks; science began to influence selection of raw materials and glassworks practices;

e) 1940 to 1945 – the period of wartime shortages; representative statistical data were not always available;

f) 1946 to 1960 – production became fully automatic; first suction then rotary feeder-fed machines were abandoned;

g) 1971 to 1980 – increasingly production used IS machines, dimensions and outputs of melting furnaces increased further, production became fully automated.

3. Flint container glass

At present about one third of all glass melted in the world and half of all container glass is white flint. It has not always been so; in the early days of container production green glass (coloured only by impurities) prevailed, being melted from cheap local raw materials. Purer raw materials came into use in the 1860s (the oldest analysis found is from 1868) and the products began to be designated as white. Sometimes the glass was called "half-white" or even "three-quarters white".

The statistical analysis included 361 analyses, 37% of all the data. Average compositions of particular groups are shown in table 1, averages for periods in table 8 (see section 10), the evolution of the main constituents is shown in figures 1a and b.

Alkali content varied considerably at first, but from about 1910 all groups had very similar R_2O content which decreased by about 0.3 % R_2O in a decade. German glasses had significantly higher R_2O around 1948. At the end of the period concerned (1980 to 1985) Albanian glasses had the highest alkali content (15.7 % R_2O) and glasses from other developing countries (Poland, Vietnam, Korea, China) also had high R_2O. The lowest alkali content was found in Swiss glasses (12.4 % R_2O). Dietzel's sum (R_2O + RO) had a long-term average of about 25.5 % but the total range was from 21 to 30 %.

White flint container glasses have long had soda as the main alkali component; potash contents above 1 % were infrequent after 1930 but German and British glasses generally had at least 0.4 % K_2O and similar levels were occasionally found elsewhere, often the result of supplying alumina as feldspar.

Before about 1920 RO content was near to 8 % but it increased gradually to about 11.5 %. Significant variations were seen in American and British glasses. Up to 1915, MgO content was less than 0.50 % but one German analysis showed 1.2 % MgO in 1924, and another 3.6 % MgO in 1927. However, in 1946 to 1950 only 10 % of German glasses contained MgO. MgO occurred in American glasses from 1923 and magnesia was generally found after 1930 but some glasses without MgO were reported around 1980. In 1978 to 1987 in England, where pure limestone was readily available, MgO was added to only 40 % of the glasses. MgO was added to only half of French glasses from 1978 to 1987. Around 1985, container glasses from Poland, the Netherlands, Denmark, Norway, Korea, and Mexico were melted without raw materials containing magnesia. Czech white container glass contained 0.2 % MgO in 1923, 0.8 % MgO in 1929 and 4.2 % MgO after 1970; high MgO contents (3.5 to 4 %) were also popular in Russia; these two countries used all dolomitic batch but other countries favoured mixtures of limestone and dolomite. The variability of magnesia contents, see figures 1a and b, gave a standard deviation of ± 1.6 % MgO.

Alumina was at first provided only by impurities in sand; up to 1910 glasses generally had about 1.7 % Al_2O_3. When purer sands became available the alumina content decreased in all glasses, minimum values of about 0.50 % Al_2O_3 being found. Czech glasses remained at that level until 1970 to 1980 but higher values were sometimes found: 1.45 % Al_2O_3 in 1923 and 2.8 % Al_2O_3 in 1929. Alumina contents were subsequently increased to improve chemical durability. In 1950, average alumina content had regained about 1.9 % Al_2O_3. Yet American glasses from 1948 had 5.5 % Al_2O_3 and a similar level was found in Albanian glasses between 1980 and 1990. High alumina contents were also found in Brazil, China, and Russia. Low alumina contents were found in Slovakia and Czechia.

On looking at individual periods, average silica content varied between 70.0 and 73.5 % SiO_2. The sum of $Al_2O_3+SiO_2$ tended to vary only a little, from 73.0 to 75.0 %, averaging 74.0 with standard deviation ±0.7 %.

Iron content everywhere decreased. In 1930 French and British flints had 0.3 % Fe_2O_3 but American glasses then contained 0.10 % Fe_2O_3, whilst only a decade later (1940 to 1945) Czech glasses had only 0.05 % Fe_2O_3. By 1980 iron content in most countries had settled at about 0.05 % Fe_2O_3; higher contents being seen only in countries with still-developing glass industries, such as Albania (0.35 % Fe_2O_3), and Bulgaria (0.2 % Fe_2O_3).

Pyrolusite was long used as decolorizer: German glasses showed 0.46 % MnO in 1871 and about 0.22 % MnO was found up to 1925, Czech glasses produced by Mühlig company contained about 0.28 % MnO in 1923 to 1929, but the glasses produced in Nové Sedlo in 1934 had no manganese. During the Second World War manganese was not available and its use was not resumed after 1950 when selenium was used instead.

Initially alkali was generally provided by saltcake but that was steadily replaced by soda ash during the 20th century. An average SO_3 of 0.6 % in 1923 to 1937 decreased to about 0.2 % SO_3 in 1985. High SO_3 contents in older analyses could be partly attributed to systematic analytical error. Sulphate was always used as a refining agent, but in 1880

Table 1. Average composition of flint container glasses in wt%

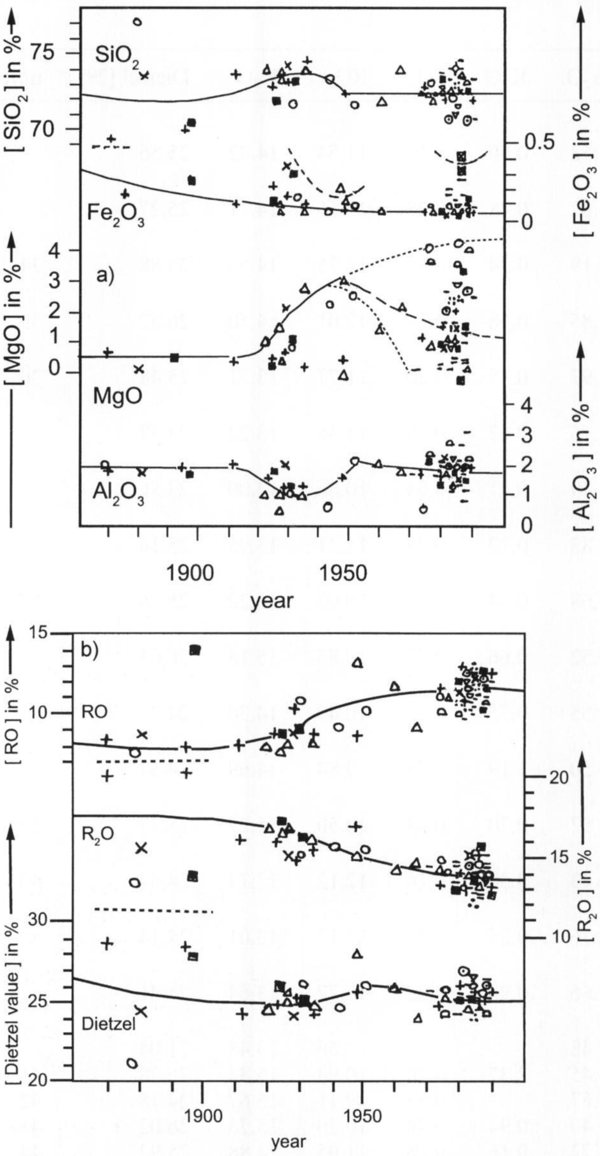
period	*n*	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3	RO	R_2O	Dietzel [29]	note
Germany:													
1868-1871	3	69.34	1.65	0.17	8.24	0.69	19.22	2.98		8.47	20.21	28.68	1
1891-1899	3	69.85	1.71		8.01		17.29	4.77		8.91	20.47	28.48	2
1909-1915	9	73.59	1.85	0.10	7.69	0.34	15.59	0.97		7.99	16.24	24.33	
1923-1926	18	72.80	1.48	0.21	8.33	0.32	15.87	0.83	0.58	8.68	16.13	24.81	3
1927-1931	20	73.17	1.13	0.15	9.66	0.66	14.47	1.40	0.35	10.41	14.96	25.38	4
1933-1937	13	74.34	1.15	0.10	8.48	0.17	13.02	3.02		8.70	15.72	24.41	5
1946-1950	9	72.24	1.42	0.16	8.42	0.37	17.23	1.09	0.63	8.66	17.05	25.71	6
1971-1977	4	72.79	1.61	0.041	10.37	1.07	12.86	0.45	0.29	11.55	13.31	24.86	7
1980-1982	11	72.01	1.55	0.059	10.09	1.77	13.57	0.54	0.26	11.93	14.10	26.04	8
1983-1987	11	72.38	1.62	0.054	9.87	1.99	12.98	0.64	0.20	11.90	13.65	25.53	9
1988-1991	13	71.86	1.91	0.051	9.97	2.53	12.47	0.77	0.14	12.50	13.25	25.75	10
France:													
1879-1882	4	73.42	1.54		8.60	0.10	14.32	1.44		8.62	15.88	24.51	11
1927-1931	3	74.16	1.91	0.36	8.27	2.17	14.96	0.48	0.88	9.00	15.12	24.11	12
1980-1983	4	72.55	2.12	0.032	10.20	0.84	13.15	0.19	0.16	11.18	13.72	24.90	13
1985-1990	5	72.61	1.48	0.052	10.69	1.46	12.91	0.34	0.23	12.17	13.25	25.42	14
Great Britain:													
1875-1919	3	70.48	1.75	0.25	13.75	0.55	12.57	4.50		13.91	14.07	27.98	15
1925-1926	13	71.88	1.69	0.15	8.46	0.32	15.48	4.62		8.75	17.26	26.01	16
1927-1934	11	73.20	1.13	0.32	8.28	1.08	16.40	–	0.64	8.99	16.40	25.39	17
1978-1982	5	73.34	1.30	0.049	11.06	0.96	12.60	0.52	0.30	12.02	13.12	25.14	
1983-1987	8	73.01	1.71	0.045	10.31	0.87	13.37	0.44	0.20	11.18	13.81	24.99	18
1988	6	72.58	1.43	0.052	11.07	1.57	12.30	0.48	0.15	12.63	12.78	25.41	
Italy:													
1980-1984	4	70.52	2.01	0.094	10.30	2.01	13.95	0.66	0.23	12.31	14.61	26.92	19
1986-1989	3	70.47	1.99	0.068	9.53	2.59	12.44	1.37	0.21	12.15	13.81	25.97	20
Russia:													
1972-1980	6	72.28	2.31	0.074	6.91	3.55	14.00	0.62	0.52	10.09	14.21	24.22	21
1982-1984	18	72.13	2.71	0.265	6.52	3.48	14.44	0.58	0.28	9.96	14.64	24.60	22
1985-1990	5	73.01	2.11	0.102	6.36	3.97	13.74	0.22	0.26	10.47	13.87	24.34	
USA:													
1922-1923	5	74.03	0.89		6.99	1.09	16.87			8.02	16.87	24.89	
1927-1929	8	73.85	0.90	0.10	7.03	1.55	17.03			8.23	17.03	25.26	
1933-1936	13	74.02	0.90	0.09	5.12	2.94	16.31	0.04	0.41	8.24	16.31	24.57	23
1946-1950	2	66.10	5.53	0.13	9.70	3.08	13.35	1.50		13.11	15.05	28.16	
1960	3	71.83	1.90	0.053	9.80	1.37	13.97	0.53	0.22	11.47	14.50	25.97	24
1967	4	73.95	1.72		7.57	2.20	14.90			9.15	14.90	24.05	25
1977	4	72.18	2.22	0.053	9.99	0.83	13.70	0.67	9.17	10.95	14.38	25.33	26
1982-1984	3	72.37	1.69	0.052	10.60	1.06	13.64	0.37	0.15	11.73	13.94	25.74	27
Canada:													
1927-1928	2	73.85	0.42	0.06	8.54	0.36	16.74			8.90	16.77	25.67	
1984-1990	2	73.55	1.41	0.054	11.29	0.10	13.10	0.25	0.17	11.38	13.34	24.72	
Austria:													
1981-1990	6	71.51	1.81	0.049	9.89	2.78	12.32	1.06	0.19	12.67	13.38	26.95	28
Hungary:													
1981-1987	8	72.65	1.45	0.065	9.04	2.36	13.64	0.30	0.22	11.73	13.93	25.66	29
Bulgaria:													
1981-1983	4	73.12	1.95	0.20	7.09	2.64	14.12	0.70	0.20	9.73	14.83	24.56	
Poland:													
1979-1984	3	71.55	1.62	0.064	10.39	0.10	14.95	0.16	0.24	10.48	15.11	25.59	30
Holland:													
1980-1990	4	72.77	1.68	0.039	11.26	0.45	12.99	0.34	0.19	11.70	13.33	25.03	
Switzerland:													
1984-1986	3	73.10	1.42	0.051	10.53	2.40	11.56	0.80	0.11	12.93	12.37	25.30	
Yugoslavia:													
1975-1988	4	72.94	1.39	0.114	8.46	2.76	13.57	0.17	0.25	11.39	13.84	25.12	31
Romania:													
1981-1982	2	72.15	2.07	0.172	8.95	1.65	14.53	0.10	0.19	10.60	14.64	25.25	32
Albania:													
1983-1989	3	68.24	4.94	0.35	7.17	3.30	14.60	1.12	0.10	10.47	15.72	25.20	33
Finland:													
1981-1990	4	72.30	1.57	0.047	10.73	0.79	13.48	0.81	0.25	11.51	14.29	25.82	

Table 1. Continued

period	*n*	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3	RO	R_2O	Dietzel [29]	note
Sweden:													
1973-1990	3	72.31	1.57	0.088	10.10	1.44	13.52	0.50	0.16	11.54	14.02	25.56	
Denmark:													
1981-1988	2	72.49	1.96	0.056	10.86	0.30	13.62	0.48	0.28	11.16	14.11	25.27	
Norway:													
1971	1	71.80	1.60	0.052	10.56	0.13	14.19	0.34	0.25	11.35	14.53	25.88	34
Spain:													
1980-1988	2	70.72	2.05	0.072	11.22	0.80	13.85	0.66	0.26	12.01	14.50	26.52	35
Portugal:													
1988-1990	2	72.98	1.10	0.076	9.49	1.49	12.97	0.75	0.20	11.77	13.71	25.48	36
Belgium:													
1984	1	74.00	1.50	0.046	10.46	0.89	12.75	0.47	0.16	11.35	13.22	24.57	
Greece:													
1985	1	74.38	1.42	0.048	9.19	1.67	12.59	0.41	0.04	10.86	13.00	23.86	37
Cuba:													
1985-1989	5	72.46	1.82	0.159	10.26	1.07	13.83	0.12	0.24	11.21	13.95	25.16	
Vietnam:													
1983-1988	4	71.45	1.90	0.27	8.42	1.56	14.68	0.54	0.20	10.03	15.22	25.26	38
Korea:													
1984-1985	2	72.26	1.02	0.058	10.85	Tr.	14.52	0.66	0.35	10.85	15.18	26.03	
Mexico:													
1988-1990	2	72.85	1.71	0.068	10.49	Tr.	13.55	0.75	Tr.	10.49	14.30	24.79	
Brazil:													
1989	1	71.95	3.13	0.100	9.71	0.13	13.50	1.19	0.23	9.84	14.69	24.57	
China:													
1988	1	70.63	2.66	0.145	10.13	Tr.	14.57	0.70	0.38	10.50	15.27	25.77	39
Colombia:													
1988	1	72.54	2.06	0.098	11.11	0.32	12.43	0.30	0.16	12.12	12.73	24.84	40
Australia:													
1988	1	73.59	1.54	0.066	10.75	0.37	12.76	0.25	0.23	11.12	13.01	24.14	
Japan:													
1980	1	73.73	1.55	0.048	10.57	0.20	12.66	0.98	0.22	10.77	13.64	24.41	
Czechia:													
1879	1	77.13	1.83		7.56		13.48			7.56	13.48	21.04	
1926-1937	8	71.67	1.21	0.15	9.92	0.81	14.45	2.37	0.70	10.94	15.34	26.28	41
1941-1945	6	73.42	0.57	0.05	6.64	2.27	15.67		0.55	9.11	15.67	24.78	42
1947-1956	6	71.64	2.03	0.10	7.41	2.56	15.40	0.94	0.46	10.30	15.73	26.02	43
1971-1980	4	73.00	0.45	0.051	6.91	4.11	14.72	0.16	0.18	11.05	14.88	25.92	44
1981-1990	5	72.46	1.33	0.069	7.19	4.26	13.53	0.71	0.19	11.45	14.25	25.70	
Slovakia:													
1987-1989	2	72.29	9.78	0.066	7.35	4.62	14.34	0.28	0.33	11.98	14.46	26.45	

Notes:

1: + 0.46% MnO
2: + 0.20% MnO; 1 · 0.10% Sb_2O_3
3: + 0.23% MnO; 1 · 2.41% BaO; 1 · 5.18% B_2O_3
4: 3 · BaO (2.77 ± 1.72)%; 2 · B_2O_3 (2.25 ± 0.57)%
5: 4 · BaO (0.63 ± 0.78)%; 2 · B_2O_3 (2.50 ± 0.50)%
6: + 0.02% MnO; 2 · BaO (1.23 ± 1.03)%
7: 1 · 0.42% BaO; 1 · 0.73% B_2O_3
8: 1 · 0.81% BaO; 1 · 0.21% B_2O_3
9: 5 · BaO (0.19 ± 0.11)%; 1 · 0.65% B_2O_3
10: 1 · 0.38% BaO
11: 1 · 2.70% PbO
12: + 0.11% MnO; 1 · 0.19% Sb_2O_3; 1 · 0.11% As_2O_3
13: 1 · 0.56% BaO; 1 · 0.65% B_2O_3
14: 1 · 0.13% BaO
15: + 0.15% MnO; 2 · AsO_3 (0.38 ± 0.12)%
16: + 0.97% MnO; 1 · 1.50% BaO; 1 · 0.32% B_2O_3
17: 1 · 0.25% B_2O_3
18: 1 · 0.36% BaO; 1 · 0.48% B_2O_3
19: 1 · 0.52% BaO
20: 1 · 0.09% BaO; 1 · 1.43% PbO; 2 · 1.00% B_2O_3
21: 1 · 0.74% ZrO_2; 1 · 2.87% B_2O_3
22: 1 · 1.00% B_2O_3
23: 5 · BaO (0.52 ± 0.22)%; 3 · B_2O_3; (0.58 ± 0.23)%
24: 3 · BaO (0.30 ± 0.17)%; 2 · F (0.13 ± 0.05)%
25: 1 · 0.50% BaO; 1 · 0.70% B_2O_3; 1 · 0.30% F
26: 4 · BaO (0.10 ± 0.05)%; 0.05% B_2O_3
27: 1 · 0.18% BaO
28: 2 · BaO (0.18 ± 0.22)%; 1 · 0.005% Cr_2O_3
29: 2 · BaO (0.25 ± 0.08)%
30: 2 · BaO (0.61 ± 0.01)%
31: 1 · 0.67% BaO
32: 1 · 0.29% BaO
33: 1 · 0.07% Cr_2O_3
34: 1 · 0.66% BaO
35: 1 · 0.54% B_2O_3
36: 1 · 1.59% PbO
37: 1 · 0.15% BaO
38: 1 · 0.20% BaO
39: 1 · 0.17% BaO
40: 1 · 0.69% BaO
41: + 0.16% MnO; 1 · 2.51% BaO
42: + 0.09% MnO
43: + 0.16% MnO
44: 1 · 0.10% BaO



Figures 1a and b. Evolution of the compositions of flint container glass 1830 to 1990; average of partial groups is plotted: + Germany, × France, ■ England, ⁘ Hungary, △ USA, Russia, ⊙ Italy, ○ Czechia, Austria, Poland, I Yugoslavia, Sweden, Cuba, Brazil, Slovakia.

to 1915 As_2O_3 or Sb_2O_3 were found in one third of glasses, arsenic was also found in three glasses from 1923 to 1937.

The most frequently used other minor constituents were BaO and B_2O_3; see table 2 which gives the number of analyses for each period in which each of those oxides were found, as well as the proportion of all analyses containing it for each period. Those oxides were not used in flints up to 1923 and they were not found in Czech glasses during the Second World War. In other periods, BaO was added to 15 to 30 % of glasses, but the level continuously decreased – from an average of 1.32 to 0.33 % BaO after 1971. Boric oxide was used from the 1930s in no more than 9 % of all glasses and its level decreased gradually but more than 0.5 % was used chiefly in pharmaceutical glasses, probably for better chemical durability. 1.0 to 1.5 % PbO was seen three times towards the end of the 19th century, perhaps for a better-looking glass. In 1975 0.7 % of ZrO_2 was used in Russia, but did not become accepted. American glasses from 1960 to 1967 included 0.05 to 0.3 % F_2.

4. Brown container glass coloured by manganese

Brown glasses coloured by manganese and iron are rather rare, comprising only 37 analyses (4 %) of all container glasses, they came from Germany and Czechia, the oldest ones being from 1887. These glasses were not melted during World War II when manganese was unobtainable. Most manufacturers, above all those in the USA and England, preferred to melt cheaper amber glasses but both types were made in Germany [25]. After 1945 glasses coloured with manganese were probably melted only in Czechia where such bottles were made on Owens machines until 1980 or 1990 for small containers. An overview of compositions is shown in table 3.

The main advantage of such glasses was the possibility of using high-alumina compositions so that alkali could be supplied by minerals (phonolite); thus compensating for the cost of the manganese. German glasses contained 6 to 9 % Al_2O_3. Czech glasses were developed by König [25] in 1929 and were successfully melted by the Mühlig company [30] for a considerable time. Alumina content was increased to 15 % Al_2O_3 but all these glasses had lower than usual RO + R_2O (24.5 % on average) which was necessary to maintain chemical durability. Melting was not difficult because MnO is a good fluxing agent; it was even possible to melt brown glass entirely without soda (but using saltcake as refining agent) and work it on Owens machines. The batch used was (in kg): 600 phonolite, 200 sand, 200 limestone, 18 sulphate, 35 pyrolusite from Java [11].

Introduction of alumina into manganese glasses led to lower silica and the sum of Al_2O_3+SiO_2 moved about 70 %. Alkali content was distinctly low, from 8 to 12 % R_2O, de-

Table 2. Additions of BaO and B_2O_3 in flint container glass in wt%

period	BaO				B_2O_3			
	number of analysis	in wt% of all	content in wt%	mean content in wt%	number of analysis	in wt% of all	content in wt%	mean content in wt%
1923-1937	15	13	0.3-3.0	1.32	10	9	0.3 -5.2	1.62
1941-1945	–	–	–	–	–	–	–	–
1946-1960	6	25	0.2-2.0	0.64	2	8	0.15-0.7	0.43
1971-1980	7	28	0.1-0.7	0.33	2	8	0.6 -2.9	1.80
1981-1990	25	15	0.1-0.8	0.34	8	5	0.2 -1.0	0.69

Table 3. Average composition of brown bottles coloured by manganese in wt%

period	n	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3	MnO	RO	R_2O	Dietzel [29]
Germany:													
1887-1891	3	67.55	9.37	1.46	14.00	0.35	10.02	2.65		7.13	14.12	11.90	26.02
1909-1915	13	60.27	8.94	1.44	14.21	6.83	6.82	1.28	0.55	5.95	16.07	7.99	24.10
1925-1932	7	63.11	5.81	1.58	12.34	2.72	7.82	1.37	0.58	5.13	15.07	8.99	24.06
Czechia:													
1930-1937	5	56.95	11.67	1.49	13.02	0.58	10.33	2.20	0.66	4.78	13.60	10.81	24.41
1939-1941	2	56.10	15.32	1.54	11.79	1.08	11.35		0.46	4.28	11.86	11.35	23.21
1953	1	57.10	13.95		12.38	2.75	8.92			4.70	15.13	8.92	24.95
1971-1990	6	67.57	3.26	0.76	7.27	2.70	14.16	0.72	0.32	3.56	9.97	14.88	24.85
average composition:													
1887-1915	16	63.91	9.15	1.45	14.10	3.59	8.42	1.97	0.55	6.54	15.10	9.95	25.06
$\pm s$		5.15	0.30	0.02	0.15	4.58	2.26	0.97		0.83	1.38	2.76	1.36
1925-1937	12	60.03	8.74	1.53	12.68	4.15	9.08	1.78	0.62	4.96	14.33	9.90	24.23
$\pm s$		4.35	4.14	0.06	0.48	5.05	1.77	0.59	0.06	0.25	1.04	1.29	0.25
1939-1941	2	56.10	15.32	1.54	11.79	1.08	11.35		0.46	4.28	11.86	11.35	23.21

pending on the mineral used. Such raw materials also introduced up to 2.2 % K_2O. MgO was seen quite early, 5.1 % MgO being found in 1909, and gradually became common. Until 1970 manganese glasses contained on average 1.5 % Fe_2O_3 but lower values were found subsequently. German glasses were coloured by 3 to 8 % MnO, Czech ones by about 4.5 % MnO. Other constituents were rarely found but in 1910 small black bottles were produced with 3.75 % MnO and 0.9 % Cr_2O_3.

6. Amber glasses

It is not known when these glasses were first melted. The oldest analyses of amber bottles from France and Hungary were published by Benrath in 1879, see [12], and the first from Germany in 1913. By 1950 such bottle glasses were made throughout the world and accounted for about a quarter of all container glass. The data cover 124 compositions, that is 13 % of all container glasses. The compositions of the groups are given in table 4 and the averages for the various periods in table 8 (section 10). Figure 2 shows the trends of the main constituents.

Amber glasses are characterized by low alumina content: they are said to have a dull brown colour when alumina and iron oxide are high. Compositions from 1879 had around 7 % at most 10 % Al_2O_3, but alumina content decreased quickly and from about 1930 fluctuated around 2.0 % Al_2O_3. American glasses from 1930 had very low alumina. The sum of alumina and silica averaged 73.7 %.

Dietzel's sum was higher in early times but after 1915 fluctuated around 26.2 %. At first alkali content increased to about 17 % in 1930 but then decreased to 14.4 % R_2O in 1985. RO varied in the opposite direction. Most of glasses contained 1 to 3 % MgO; only glasses from Britain, Poland and Korea were without MgO in 1985. Czech glasses at that time contained nearly 4 % MgO.

Iron content fluctuated somewhat around 0.27 % Fe_2O_3. It is of interest that in Britain, the USA, and Germany the colour was emphasized by addition of 0.11 to 0.34 % MnO. BaO was found in 12 glasses (10 % amber glasses) in amounts from 0.1 to 4.5 %, averaging 1.0 % BaO. Boric oxide additions were rare, being found in only 5 analyses (4 %), contents ranging from 0.07 to 0.42 % with an average of 0.8 % B_2O_3. Chromium (0.01 to 0.03 % Cr_2O_3, average 0.024 %) was found in only five glasses (4 %) dating from about 1985, perhaps from recycled cullet. American glasses from 1960 contained (0.14 ± 0.03) % F_2 from melting accelerators.

7. Green glasses

This is the largest group of container glasses: 449 analyses or 46 % of the total. The groups are given in table 5 and average compositions by periods in table 8 (section 10). Figures 3a and b show the trends. The glasses are characterized by significant but gradually decreasing variability. Since 1960 these may be divided into two groups, olive-green coloured by Fe_2O_3+MnO, and emerald green coloured by chromium.

The oldest hand-made glasses were melted with large proportions of minerals which yielded higher Al_2O_3 contents. Average alumina content increased from early times until around 1940 when it reached 7 % Al_2O_3. The introduction of machines appears not to have influenced the alumina contents significantly. But the introduction of newer feeder-fed machines after 1950 was accompanied by a decrease of alumina content (to about 2.3 % Al_2O_3).

Alkali contents varied significantly beginning at about 5 % R_2O (1830) and rising to nearly 15 % around 1965; then decreasing slightly to about 14.1 % R_2O. The lowest content, 2.86 % R_2O, was found in a French glass dated 1881 and only 3.45 % R_2O in an English glass dated 1884. Contents of about 5 to 6 % R_2O were fairly common in early times (e.g. Germany 1899, 1927; France 1926). The only glasses with more potash than soda were French ones from 1835.

RO content changed significantly and irregularly; from 1835 to 1930 two different groups were found. Glasses from Germany, Czechia, Hungary, and the USA had low RO (8 to 11 %) but that subsequently continually increased. Glasses from Italy, France, and Britain at first contained about 25 % RO but that then decreased. In 1920 to 1930 both types had converged to values of 10 to 11 % RO. After that the variability decreased and RO content increased

Table 4. Average composition of amber bottles in wt%

period	*n*	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3	RO	R_2O	Dietzel [29]	note
Germany:													
1913	4	70.32	3.38		8.60	1.33	16.59			9.35	16.58	26.50	
1925-1937	4	73.01	0.68	0.20	6.99	1.51	16.76	0.90		8.87	16.76	25.63	1
1946-1950	8	71.81	1.61	0.24	7.77	0.22	16.49	1.03		8.68	17.51	26.19	2
1972-1983	10	72.17	1.85	0.27	10.28	1.42	13.17	0.58	0.05	11.72	13.80	25.52	3
1984-1990	15	71.07	2.26	0.33	10.37	2.30	12.44	0.86	0.02	12.79	13.17	25.95	4
France:													
1879	3	62.11	7.89		19.54		9.87			19.54	9.87	29.41	
1978-1988	3	72.11	1.98	0.15	11.01	1.09	12.86	0.39	0.05	12.10	13.25	25.35	5
Great Britain:													
1926-1930	5	71.72	1.80	0.37	9.92	0.69	15.35			10.73	15.35	26.08	6
1980-1988	4	72.73	1.88	0.25	10.96	0.19	12.84	1.06	0.04	11.15	13.90	25.05	
Austria:													
1976-1985	7	70.81	2.63	0.26	9.27	2.36	13.50	0.95	traces	11.72	14.45	26.17	7
1987-1990	7	70.96	2.15	0.25	10.44	2.41	12.73	0.88	0.02	12.87	13.60	26.47	8
Hungary:													
1879	2	68.80	7.40		8.94		14.92			8.94	14.92	23.85	
1987	2	72.35	1.80	0.16	8.72	2.40	13.89	0.37	traces	11.19	14.25	25.44	9
Italy:													
1968-1989	8	70.92	2.53	0.21	9.33	2.02	13.58	1.13	0.02	11.35	14.71	26.06	10
Russia:													
1983-1988	5	72.17	2.04	0.32	7.16	2.90	14.68	0.32	0.04	10.20	14.88	25.08	
Bulgaria:													
1976-1984	4	72.18	1.84	0.32	8.91	2.29	12.83	0.78	0.02	11.20	14.60	25.80	
Cuba:													
1984-1987	5	71.37	1.61	0.23	9.46	1.56	15.34	0.28	0.05	11.02	15.62	26.64	
Yugoslavia:													
1982-1990	3	71.25	1.95	0.16	9.51	2.41	14.34	0.12	traces	11.59	14.46	26.05	11
Poland:													
1980	1	72.10	1.30	0.20	1.38	0.15	13.90	0.42	0.01	11.53	14.32	25.85	12
Romania:													
1988	1	71.85	2.61	0.51	8.40	1.55	14.39	0.55	traces	10.03	14.94	25.02	13
Belgium:													
1988	1	71.65	1.76	0.20	8.45	2.49	14.47	0.42	0.01	10.94	14.89	25.83	
Denmark:													
1983	1	71.82	2.38	0.23	10.35	2.26	12.59	0.80	traces	12.61	13.39	26.00	14
Switzerland:													
1982	1	71.00	1.95	0.24	10.59	2.27	13.02	0.65		12.86	13.67	26.53	
Sweden:													
1990	1	72.64	1.78	0.22	9.86	2.09	12.64	0.44	traces	11.95	13.08	25.03	
Brazil:													
1985	1	71.53	1.80	0.26	7.50	2.86	15.19	0.53	traces	10.36	15.72	26.08	
Vietnam:													
1986	1	71.65	1.91	0.31	9.99	1.21	13.80	0.56	traces	11.20	14.36	25.56	15
Korea:													
1989	1	70.05	3.13	0.34	9.30	0.06	15.62	1.11	0.02	9.36	16.73	26.09	
Spain:													
1879	3	63.76	7.37		10.98		17.88			10.98	17.88	28.86	
USA:													
1927-1933	5	72.87	0.61	0.30	7.20	1.46	17.11	0.35		8.66	17.11	25.77	16
1960	4	71.93	2.18						0.07	11.25	14.55	25.80	17
1977	1	71.54	2.47	0.21	10.15	0.74	13.99	0.71	0.06	10.97	14.70	25.67	
Czechia:													
1971-1989	4	72.80	1.42	0.23	7.33	3.94	13.51	0.49	0.04	11.27	13.99	25.36	18

Notes:

1: + 0.38% MnO; 1 · 4.50% BaO
2: + 0.11% MnO; 2 · BaO (2.78 ± 0.81)%
3: 1 · 0.28% BaO
4: 3 · 0.03% Cr_2O_3
5: 1 · 0.02% Cr_2O_3
6: + 0.35% MnO
7: 3 · BaO (0.22 ± 0.14)%
8: 1 · 0.10% BaO
9: 1 · 0.15% BaO
10: 3 · B_2O_3 (0.42 ± 0.1)%
11: 1 · 0.07% B_2O_3
12: 1 · 0.25% BaO
13: 1 · 0.13% BaO
14: 1 · 0.01% Cr_2O_3
15: 1 · 0.08% B_2O_3
16: + 0.44% MnO
17: + (0.14 ± 0.03)% F
18 : + 0.11% BaO

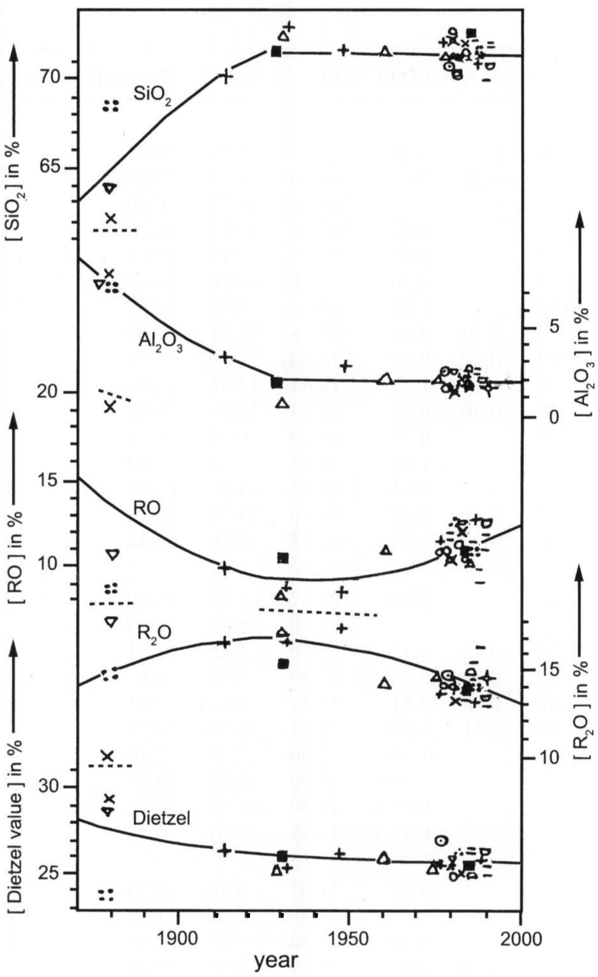


Figure 2. Evolution of the compositions of amber container glass = average of partial groups is plotted:
+ Germany, × France, ■ England, ∷ Hungary, △ USA, ✧ Belgium, ⌓ Russia, ⊙ Italy, ○ Czechia, ▽ Austria, ⌘ Poland, ∴ Switzerland, — Bulgaria, Yugoslavia, Romania, Sweden, Cuba, Denmark, Vietnam, Australia, Brazil, Korea (less number of samples).

somewhat to average 11.5 % RO in 1965. RO + R_2O consequently varied considerably. Up to 1915 it averaged 29 to 30 % but then decreased to a level of about 26.5 % and remained there.

It seems that MgO was used in green bottle glasses from the beginning of their production. In 1830 glasses with 7 % MgO occurred in France, where 8.0 % MgO was found in 1897. Contents above 1.0 % MgO, showing deliberate addition of magnesia raw materials, were found in 21 % of analyses for 1830 to 1880, in 49 % for 1880 to 1915, and in 42 % for 1919 to 1939. After 1945, MgO was common in green glasses but in 1950 only 22 % of German green glasses contained more than 1 % MgO. American glasses contained only 0.65 % MgO in 1977; glass from Poland only 0.2 % MgO in 1984. Czech glasses had more than 1 % MgO in 1931 and after that MgO content increased to 5.0 % MgO in 1954. Then it decreased to a common level of about 2.2 % MgO.

Higher than usual lime contents were found in Italian and French glasses but lower than average in glasses from Germany, Poland, and America. CaO content fluctuated strongly and a downward trend is evident. High lime contents were seen in Italian glasses (27.2 % CaO in 1879) and French glasses. In recent years Polish and Russian glasses have been below average for lime.

0.10 to 0.40 % As_2O_3 occurred in three German glasses dated 1927 and in one glass dated 1935 but these glasses had sulphates as the main refining agent. SO_3 content fluctuated considerably and it decreased from about 0.5 to 0.6 % SO_3 after 1950 to be about 0.1 % SO_3 in 1980.

Iron content was about 5 % Fe_2O_3 about 1850 but decreased from then; it reached about 1 % Fe_2O_3 in about 1890. Since 1960 there has been a further decrease to about 0.4 % Fe_2O_3. Olive green glass with manganese additions was melted in Germany until 1950 and in France until 1960. Some MnO was found in French champagne bottles (0.05 to 0.10 % in 1960 to 1978), but the glass was coloured by chromium. Also Russian glasses from around 1980 contained 0.57 % MnO, even though coloured by 0.09 % Cr_2O_3. Czech glasses coloured by chromium were found after 1965; waste chromium slag was used as colorant.

Manganese content increased gradually in glasses coloured by manganese and iron, reaching a maximum of around 2 % in 1930; the minimum about 0.2 % MnO. Additions of 0.5 to 1.1 % MnO were used according to the desired tint. Emerald green glasses used (0.15 ± 0.04) % Cr_2O_3. In a few cases green glasses were overcoloured by a trace of cobalt.

Other additions have rarely been found in green glasses: BaO was found in 8 glasses (only 2 % of all glasses) which had from 0.05 and 4.3 % BaO, averaging 1.3 % BaO. German glass dated from 1915 contained 2.65 % F_2, some from 1970 had 0.35 % F_2.

8. Variability of composition

Average variations were calculated for particular periods and for each type of glass. The standard deviations are given in table 6, the last line of which gives the average variation of analyses about the yearly average of composition of container glasses melted in the period of 1970 to 1990 in Czech melting units with 70 to 200 t/day output, according to data from laboratory files [1$].

The variations in a group represent composition ranges in one country over a relatively short period; it may be assumed to include both "natural" variations due to raw materials and inaccurate batch plant operation, as well as changes caused by small composition modifications, such as refining agents, in individual glassworks and incidental differences between the glass works. During the 150 years covered by the data such variations continually decreased. If the range in 1919 to 1938 is expressed as ±100, the average value for all observed oxides changed as follows (table 7).

Before 1914 almost all container glass was hand-blown, so that variations in composition did not matter very much: the glassmaker was able to allow for deviations in viscosity with hand-formed glass. As soon as the change-over to continuous mechanical production occurred (from about 1919), it was necessary to maintain viscosity and composition more precisely, so that variations at once decreased. Data

Table 5. Average composition of green container glasses in wt%

period	*n*	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3	MnO	RO	R_2O	Dietzel [29]	note
Germany:														
1851	2	62.77	2.94	5.62	22.13		4.93	1.96	0.63	0.28	22.13	6.89	29.02	
1870-1874	5	63.45	3.40	5.39	18.15	0.55	6.66	2.71	0.55	Tr.	18.26	9.37	27.63	
1884-1889	12	58.65	8.21	2.05	17.32	1.53	10.10	2.25			18.85	11.75	30.60	
1891-1889	11	62.85	3.40	3.01	15.33	2.23	9.59	3.16		0.82	17.56	12.75	30.30	
1904-1909	10	62.89	7.22	2.15	14.35	2.10	7.77	1.54		0.80	17.31	9.77	27.08	1
1910-1913	14	62.75	8.87	1.68	15.13	2.16	7.67	2.31		0.70	17.28	9.20	26.49	
1915	17	63.70	9.37	1.87	13.95	2.70	7.63	0.95		1.12	15.28	8.78	24.17	2
1924-1925	7	62.97	7.22	1.56	14.64	1.22	9.37	1.17	0.50	0.63	15.86	11.21	27.07	
1926-1927	62	65.28	6.25	2.04	12.98	1.52	10.34	2.15	0.52	0.89	14.50	11.29	25.76	3
1928-1929	29	64.88	6.08	2.04	12.65	0.98	10.58	2.36		1.11	13.64	12.46	26.11	
1930-1932	26	63.73	7.34	2.12	13.71	1.33	8.86	2.39	0.50	0.89	14.84	10.91	25.76	
1934-1937	7	64.00	6.92	2.26	15.70	0.71	8.13	1.73		0.75	16.49	10.05	26.54	
1927-1929	3	71.06	2.37	2.29	7.64	0.53	15.42			1.05	8.17	15.42	23.60	4
1945-1950	18	65.26	5.87	1.04	11.72	0.90	12.56	1.71	0.39	0.85	12.62	14.27	26.89	
1970-1977	6	71.96	2.15	0.37	10.23	1.62	12.54	0.74	0.21	0.19 x	11.86	13.28	25.13	5
1982-1990	10	71.38	2.31	0.35	9.86	2.28	12.45	0.86	0.10	0.19 x	12.15	13.26	25.44	
France:														
1830-1835	9	58.00	8.08	4.04	23.15	4.66	0.40	5.87		0.80	25.57	5.03	30.60	6
1871-1879	10	62.30	5.31	4.51	21.44	3.35	4.42	3.20			22.10	5.92	28.03	
1881-1889	6	61.55	5.58	1.67	21.52	4.14	4.93	0.78	0.12	0.11	25.66	5.45	31.11	
1896-1899	6	60.30	5.87	1.95	20.25	6.30	5.43	2.23	0.46	0.55	22.90	7.98	30.87	
1920-1929	9	64.58	5.24	2.35	17.69	3.69	8.32	1.70	0.64	0.81	18.92	8.69	27.61	
1933	4	69.34	4.40	1.93	8.35	1.30	14.11	1.63	0.51	1.60	9.65	14.93	24.58	
1947-1960	1	68.4		2.25						1.50	9.80	15.30	25.10	
1960-1968	1	69.6		1.00						0.10 x	11.50	15.00	26.70	7
1968-1978	1	69.9		0.45						0.05 x	12.50	14.50	27.00	8
1984-1990	5	71.22	2.49	0.31	10.13	1.86	13.11	0.53	0.04	0.18 x	11.99	13.64	25.63	
Great Britain:														
1845	3	56.60	2.33	8.13	21.19	0.87	7.07	2.74		0.68	21.97	9.76	31.73	
1875-1884	3	63.04	2.72	2.55	22.68	5.90	5.93	1.08			25.25	6.65	31.91	
1919-1926	6	68.27	3.87	1.51	10.20	0.76	13.40	3.42		0.49	10.84	15.11	25.95	
1928-1930	5	67.71	3.67	2.24	9.53	1.01	14.40		0.76	1.87	10.79	14.40	25.19	
1988	1	72.07	1.85	0.31	10.55	1.12	13.37	0.60	0.03	0.20 x	11.57	13.97	25.64	
Belgium:														
1927-1934	4	66.50	4.47	2.16	11.85	0.85	13.33		0.62	0.92	12.70	13.33	26.03	
USA:														
1913	6	71.25	2.18		9.42	1.39	15.74				10.81	15.74	26.55	9
1923-1933	4	72.11	1.19	1.29	8.48	0.66	15.57			2.14	9.14	15.57	24.71	
1977	1	72.10	1.91	0.16	10.36	0.65	13.87	0.55	0.14	0.13 x	11.08	14.42	25.50	
Hungary:														
1879	4	69.61	6.17		8.67		15.45				8.67	15.45	24.12	
1978	3	70.48	1.48	0.11	9.20	2.98	15.15	0.20	0.35	0.12 x	12.18	15.35	27.53	
1982-1987	3	71.17	1.80	0.14	8.98	3.02	13.71	0.63	0.18	0.17 x	12.00	14.34	26.34	
Russia:														
1868	1	55.37	7.48	1.70	20.80	3.30				0.65	24.10	8.68	32.78	
1891	3	68.76	3.49		10.85		19.33	1.50			10.85	17.31	28.16	
1928	1	63.24	10.95	3.14	8.78	1.12	8.84	3.69	0.12	0.19	9.99	12.53	22.52	10
1955	2	68.15	4.72	1.60	7.25	1.28	15.95		0.25		8.53	15.96	24.48	
1978-1982	12	69.87	4.34	1.00	7.37	2.48	14.46	1.28	0.13		9.84	14.68	24.52	11
1984-1990	9	71.32	3.20	0.60	7.21	2.62	14.16	0.63	0.23	0.08 x	10.10	14.44	24.53	
Spain:														
1980-1990	4	71.69	1.89	0.26	8.77	2.78	13.37	0.78	9.17	0.15 x	11.63	14.14	25.77	
Italy:														
1879	3	57.70	9.87		27.18		4.09				27.18	4.09	31.27	
1915	1	63.23	4.23	1.16	17.55	3.09	10.74				20.64	10.74	31.38	
1980-1982	3	71.07	2.95	0.72	8.61	2.68	12.59	0.99	0.09	0.12 x	11.28	13.58	24.86	12
1983-1987	5	69.42	2.98	0.36	9.34	2.56	13.77	1.20	0.11	0.16 x	11.90	14.98	26.87	
Bulgaria:														
1971-1984	3	72.87	1.87	0.28	7.91	2.08	13.60	0.76	0.21	0.24 x	9.99	14.36	24.34	
Yugoslavia:														
1982-1990	3	71.42	1.87	0.30	8.62	2.46	13.86	0.82	0.06	0.19 x	11.19	14.68	25.87	13
Austria:														
1983-1990	3	70.08	3.02	0.36	9.49	2.75	12.54	1.19	0.06	0.20 x	12.26	13.73	26.00	14

Table 5. Continued

period	n	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3	MnO	RO	R_2O	Dietzel [29]	note
Romania:														
1980-1988	2	71.40	2.24	0.37	9.14	1.95	13.80	0.56	0.12	0.12*)	11.09	14.36	25.45	15
Switzerland:														
1985-1986	2	70.90	2.45	0.39	10.30	2.01	12.99	0.70	0.03	0.13*)	12.31	13.70	26.01	
Turkey:														
1984	1	71.60	1.83	0.16	9.10	2.96	13.59	0.29	0.15	0.16*)	12.06	13.88	25.94	
Poland:														
1984	1	70.33	2.33	0.51	11.57	0.21	13.85	0.69	0.14	0.10*)	11.78	14.54	26.32	
Czechia:														
1879	3	57.55	4.36		7.28		20.16				7.29	20.16	27.44	
1921-1937	13	59.50	10.00	1.98	12.76	0.86	11.97	2.58	0.67	1.13	13.55	12.52	26.08	16
1942-1945	11	58.38	13.34	2.01	11.53	1.24	12.50	2.17	0.56	0.24	12.77	12.69	25.47	
1947-1959	9	60.46	11.10	1.79	9.37	3.92	11.60	2.71	0.42	0.99	13.29	11.79	25.08	
1971-1990	1	71.28	2.05	0.49	8.77	2.70	13.52	0.66	0.25	0.18*)	11.47	14.18	25.65	
Slovakia:														
1987	2	71.30	1.55	0.24	8.79	2.53	13.00	0.61	0.29	0.14*)	11.67	13.97	25.64	

Notes:
1: 2 · BaO (4.29 ± 0.61)%
2: 1 · 2.65% F
3: 3 · As_2O_3 (0.25 ± 0.19)%
4: feeder machine O'Neil
5: 1 · 0.36% F
6: 1 · 0.45% P_2O_5; 0.90% BaO
7: average 0.10% MnO + 0.07% Cr_2O_3
8: average 0.05% MnO + 0.11% Cr_2O_3
9: glasses for feeder machines
10: 1 · 0.20% TiO_2
11: 1 · 0.50% ZrO_2
12: 2 · 0.14% BaO
13: 1 · 0.31% BaO
14: 1 · 0.05% BaO
15: 1 · 0.11% BaO
16: 1 · 0.31% As_2O_3
*) MnO replaced by Cr_2O_3.

Table 6. Average variability (± s) inside the group of glasses

period	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	RO	R_2O	Dietzel [29]
average value:										
until 1835	7.26	3.55	1.55	5.34	3.52		2.26	3.24	2.38	5.21
1840-1880	3.38	2.31	1.70	2.88	1.47	2.73	1.14	3.21	3.24	2.06
1880-1915	2.67	2.15	0.50	2.38	1.56	2.70	1.15	2.76	3.01	2.41
1919-1939	2.19	1.43	0.37	2.06	0.87	1.87	0.98	2.18	1.80	1.59
1940-1945	1.76	0.68	0.13	0.73	0.48	1.01	0.04	0.61	0.85	1.25
1946-1960	1.73	1.46	0.16	1.99	1.04	1.14		1.26	1.22	0.73
1971-1980	0.97	0.59	0.08	0.66	0.74	0.59	0.39	0.71	0.68	0.60
1981-1990	0.68	0.41	0.07	0.69	0.66	0.59	0.20	0.67	0.63	0.58
variability (± s) at Czech glassworks:										
1971-1990	0.45	0.20	0.04	0.20	0.14	0.24	0.08			

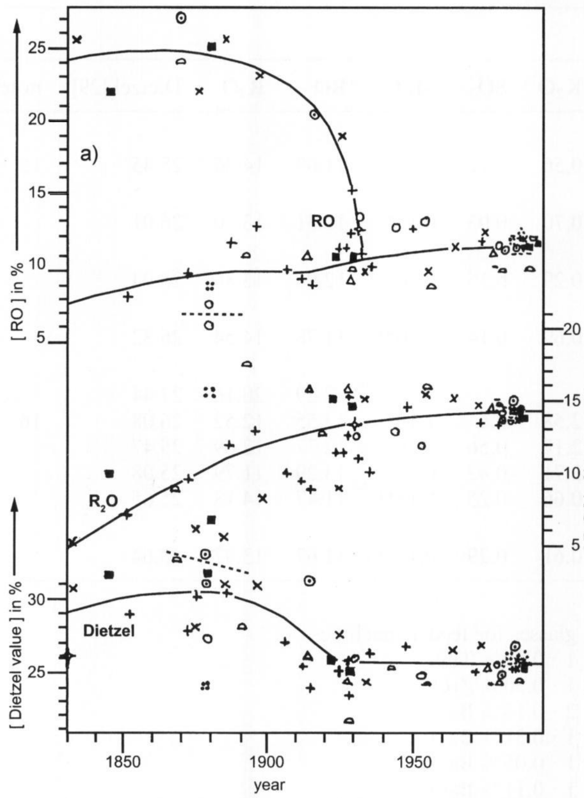
Table 7. Variability of composition for the years 1830 to 1990

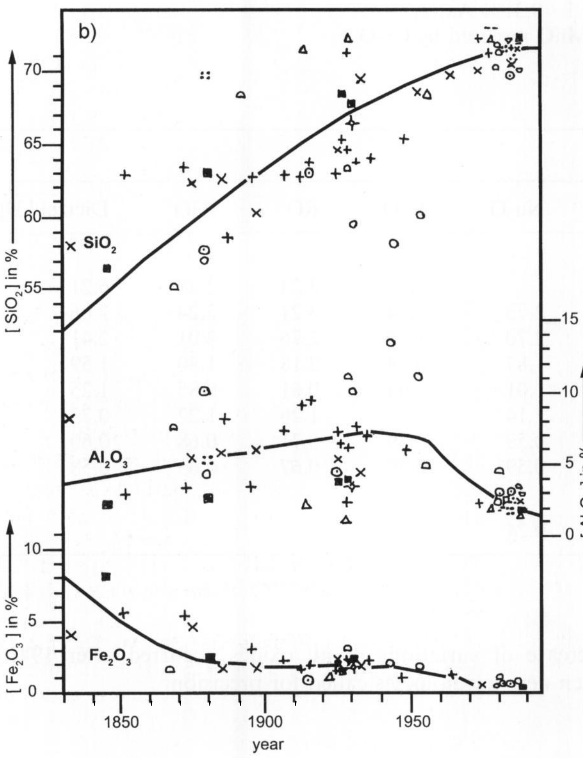
period	variability s
up to 1835	± 278
1840-1915	± 135
1919-1939	± 100
1940-1945	± 46
1946-1960	± 75
1971-1980	± 41
1981-1990	± 34

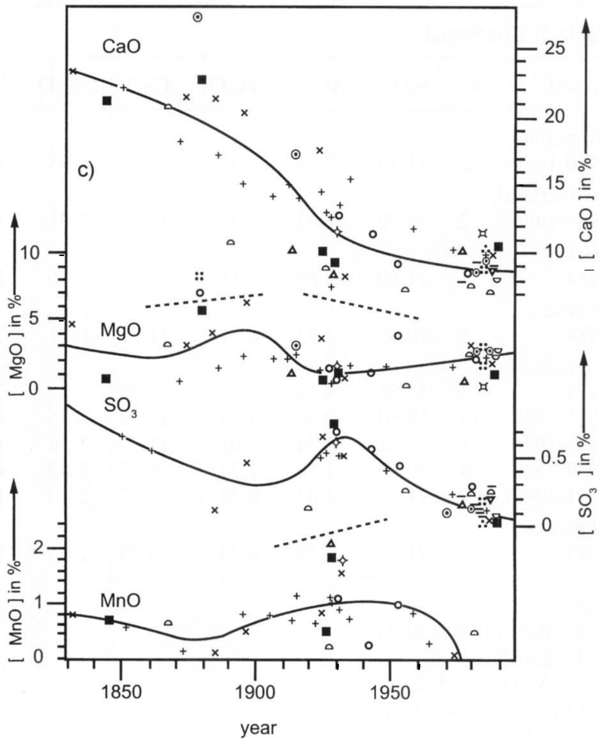
from 1940 to 1945 are not statistically representative, because they were obtained only from few glassworks. Further decrease of variations of all oxides occurred after 1946, when new technologies called for precision.

10. Comparison of container glasses

Table 8 shows average compositions of container glass of all four colours for the various periods. There are differences between colours, but they decrease progressively. The composition of amber glasses is always near to white glasses and manganese coloured glasses near to green glasses. The biggest differences are in alumina content. As soon as production of high alumina glasses was stopped and colouring with chrome green introduced, all compositions converged.







Figures 3a to c. Evolution of the compositions of green container glasses – average of partial groups is plotted: + Germany, × France, ■ England, ⁚⁚ Hungary, △ USA, ▽ Spain, ✧ Belgium, ⌓ Russia, ⊙ Italy, ○ Czechia, ⌓ Austria, D Japan, ⌘ Poland, ∴ Switzerland, — Bulgaria, Yugoslavia, Poland, Romania, Slovakia, Australia, Turkey (less number of samples).

After 1970 the composition of container glasses of all colours differ only in additions of colouring oxides, basic compositions are identical.

11. German green glasses

The composition of containers, especially of coloured ones, was studied by Keppeler around 1930 [2 to 4]. In the early days glasses with about 7.5 % Al_2O_3, high RO content (ca. 26 %) and R_2O of only 77.5 % were used throughout Europe. They were cheap and easily melted but the gathering had to be done at high temperatures and the glass had to be worked quickly to prevent devitrification, their Dietzel's sum was about 32 %. Keppeler described how a "later kind of German hand-blown bottle glass" was developed in the 1890s. It had lower RO and increased alkali partially from mineral raw materials. These glasses contained about

Table 8. Average composition of container glass in wt%

period	*n*	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3	MnO	RO	R_2O	Dietzel [29]
until 1835:													
Green	9	58.00	8.08	4.04	23.15	4.66	0.40	5.87		0.80	25.57	5.03	30.60
1835-1880:													
green	34	60.93	4.95	4.65	18.83	2.79	8.59	2.34	0.59	0.41	19.66	9.66	29.32
amber	8	64.89	7.55		13.15		14.22				13.15	14.22	27.37
flint	8	73.30	1.67	0.17	8.13	0.40	15.67	2.21		0.46	8.22	16.52	24.74
1889-1915:													
green	86	63.55	5.84	1.95	15.57	2.85	9.89	1.84	0.29	0.68	17.72	10.95	28.67
Mn-brown	16	63.91	9.15	1.45	14.10	3.59	8.42	1.97	0.55	6.54	15.10	9.95	25.06
amber	4	70.32	3.18		8.60	1.33	16.59				9.93	16.58	26.50
flint	15	71.31	1.77	0.18	9.81	0.44	15.15	3.41		0.17	9.97	16.93	26.92
1919-1939:													
green	180	65.94	5.71	2.06	11.78	1.18	11.62	2.28	0.54	1.03	12.79	12.74	25.54
Mn-brown	12	60.03	8.74	1.53	12.68	4.15	9.08	1.78	0.62	4.96	14.33	9.90	24.23
amber	14	72.53	1.03	0.29	8.04	1.22	16.40	0.62		0.39	9.42	16.41	25.83
flint	14	73.29	1.16	0.17	8.10	1.04	15.60	1.82	0.59	0.14	8.99	16.17	25.16
1939-1944:													
green	11	58.38	13.34	2.01	11.53	1.24	12.50	2.17	0.56	0.24	12.77	12.69	25.47
Mn-brown	2	56.10	15.32	1.54	11.79	1.08	11.35		0.46	4.28	11.86	11.35	23.21
flint	6	73.42	0.57	0.05	6.64	2.27	15.62		0.55	0.09	9.11	15.67	24.78
1946-1960:													
green	30	65.57	7.23	1.57	9.45	2.03	13.38	2.21	0.35	1.11	11.06	14.33	25.39
amber	12	71.87	1.90	0.14	7.77	0.22	16.49	1.03	0.07	–	9.97	16.03	26.00
flint	20	70.45	2.72	0.11	8.83	1.85	14.74	1.03	0.44	0.09	10.88	15.58	26.46
1970-1979:													
green	41	70.93	2.39	0.43	9.19	2.09	13.91	0.69	0.24		11.49	14.40	25.89
amber	35	71.63	2.10	0.25	9.41	2.15	13.37	0.75	0.03		11.60	14.27	25.87
flint	25	72.73	1.58	9.95	9.48	1.55	13.53	0.53	0.28		11.11	14.01	25.11
1981-1990:													
green	47	71.21	2.28	0.36	9.30	2.27	13.37	0.74	0.13		11.56	14.10	25.66
amber	55	71.75	2.03	0.26	9.30	1.98	13.83	0.57	0.02		11.30	14.39	25.69
flint	67	72.28	1.83	0.09	9.69	1.60	13.47	0.54	0.21		11.36	14.00	25.33

9 % R_2O_3 and 8 % R_2O. The extensive use of these glasses was possible thanks to Siemens regenerative furnaces able to reach melting temperatures 100 °C higher than before. Keppeler [2] wrote in 1930: "Some analyses show even 12 to 14 % Al_2O_3, but these glasses are seldom used and trials using these compositions for working on machines have failed".

We do not know whether specific compositions had been recommended by M. J. Owens to the European Bottle Cartel (the EV) when it was set up in 1907, but to licence his process in Europe he probably did not recommend any changes. Nevertheless, according to Keppeler [3], alkali content was increased to about 12 % R_2O and RO was decreased by 5 % to approximately 15 % RO to get over problems with high viscosity during gathering. Alumina content was held at 7 to 9 % R_2O_3; the use of natural raw materials decreased significantly, economic considerations requiring the maximum possible output.

In Germany from 1920 to 1939 bottles were predominantly produced by Owens machines, semi-automatic Roirant suction machines, and by mouth-blowing. There were no significant differences between analyses of machine-produced and hand-made glasses.

After 1919, softer (longer) American glasses for machines also spread to Europe. High-alumina bottle glasses melted from cheap mineral raw materials had not been widely used in the USA because white-flint bottles were preferred. When feeder-fed machines began to spread that also led to compositions with low Al_2O_3 contents but increased alkali. European manufacturers also appreciated such glasses: Keppeler wrote in 1930 [2] that they could allow the specific melting rate to be doubled, which suited the changeover to feeder-fed machines. As a result the alumina content was decreased but that sometimes impaired chemical durability.

12. Czech high-alumina glasses

The Czech Mühlig company [13] initially went in the opposite direction. The classic "German composition" (6 to 8 % Al_2O_3 and 12 to 15 % R_2O) was for the first time used on Owens machines. Generally, the glass was satisfactory and the general opinion was that alumina content could not be increased. However, as early as 1927 König [25] undertook laboratory trials and he began to increase the alumina content of the glass on two Owens machines. He used sulphate batch (without soda) with high-quality phonolite crushed to pass 6 mm. The high alkali content of the phonolite permitted the use of significantly lower additions of expensive sulphate (only 100 kg/1000 kg glass). Contrary to all expectations nothing untoward was seen in the furnace during trials. Melting temperature was about 1460 °C, the tank output and energy consumption were maintained. The only difference was that batch piles had to be pushed forward. Over three months alumina content increased to 17 % Al_2O_3 and was kept at that level during the whole of 1928. It was found in the following year that such high-alumina glasses had poor resistance to acids. There was a complaint that bottles of lemon juice became cloudy during storage. Laboratory research showed that green glasses with 12 to 13 % of alkalis needed more than 56 % SiO_2 to keep the acid resistance at an acceptable level. Al_2O_3 content was therefore decreased to 13 to 14 % and such glass continued to be melted from 1929 up to 1947. König [24] also claimed that green bottle glasses containing 17 perhaps even 20 % Al_2O_3 could be melted without difficulty; the poor acid resistance was the only problem. The same author [25] also reached similar conclusions about brown manganese glasses. No reports concerning problems during melting or forming have been preserved.

The great saving in batch cost with the high-alumina glass led the Mühlig company's Czech works in the 1930s to dispense with gob-fed machines, which needed more expensive soft glasses. The same composition was melted again from 1945: around 1950 saltcake was replaced by soda ash and limestone by dolomite [15]. When IS machines were first installed after 1946 long American glasses containing 1.5 to 2 % Al_2O_3 and 16 % R_2O were melted. Later alumina content was increased to 4 to 5 % Al_2O_3. In 1952 to 1954, Kotšmíd [16] executed an extensive research on a tank with IS machines and alumina content was increased stepwise to give a glass containing 12 % Al_2O_3, 11.2 % RO and 12.8 % R_2O. This glass was melted with dolomite but no limestone; the melting rate was improved by accelerators (barytes, fluoride). The glass was used on 2 to 3 tanks for 2 to 3 years, being used for both IS and Owens machines. However, it proved to suffer from batch stones and melting temperature was increased to 1490 °C.

Poor quality phonolite (grain size up to 20 mm) as well as operating problems and unskilled operators accounted for the failures. Kříž [30] therefore improved the quality of such glasses in 1958. He introduced a combination of calcite and dolomite for better fusibility, increasing RO to 11.60 % and alkali content to 14.2 %, so that the Dietzel sum reached its optimal value. Such glasses were fusible and worked well for some years but the bad impression caused by previous high-alumina glasses could not be overcome. The glasses were dropped as soon as the availability of soda improved and delivery of phonolite worsened. Glasses with 6 to 7 % of Al_2O_3 and 15 % R_2O were melted and processed on gob machines in the Russian Borzhom glassworks in 1950 to 1953 and perhaps also later, see Kutateladze [19]).

13. Development of container glass in the USA

Data concerning American container glasses collected for 1932 to 1977 are given in the works of Loesell and Lester [6] and Stadler and Cronine [7] from Emhart. SiO_2 content decreased to minimum of 71.8 % in 1960, then increased slightly. Contents of stabilizers, RO and Al_2O_3, increased up to 1960, but were constant from that time. Total fluxes, R_2O, SO_3, F_2 and B_2O_3, decreased slowly to about 14.5 % in 1977 which corresponded with 14.3 % of alkalis. Al_2O_3 ($+Fe_2O_3$) content increased to 2.0 % in 1952 then remained constant. In American glasses alumina was used as a substitute for SiO_2, so that Al_2O_3 + SiO_2 = (74.3 ± 0.5) %. Batches often used both limestone and dolomite.

SO_3 content was lower than in European glasses, at first it was 0.2 %, then it decreased to 0.15 % by 1977. BaO additions were typical for American glasses (refining by barytes), at first averaging 0.7 % BaO. In 1960, 65 % of researched glasses averaged 0.4 % BaO but in 1977, BaO was present in only 20 % of glasses which had an average of 0.08 % BaO. In 1977, approximately 0.04 % SrO coming from admixtures in raw materials was found in some American glasses. In 1937 about 0.5 % B_2O_3 was found but this gradually decreased so that in 1960 only about 20 % of the glasses contained it and after 1972 it disappeared. Also fluorine was present at 0.20 % in 1942 to 1960, when it was present in 95 % of all glasses. Then its content decreased dramatically and it has not been seen in American glasses since 1972.

14. French container glasses – champagne bottles

The compositions of champagne bottles were discussed by Chopinet [8 and 9]. The first glasses for champagne bottles (1860) mostly contained 2 to 3 %, only rarely as much as 5 % Al_2O_3, but they had very high RO contents, 18 to 20 %; the Dietzel sum exceeded 30 %. The glasses melted easily but were inclined to devitrify. Henrivaux [see 12] mentioned analyses of glasses with 4 % MgO addition from 1862. When semi-automatic Boucher machines were introduced, it was found that they could operate with the existing composition used for hand production. Nevertheless Boucher increased alkali and alumina contents slightly and decreased the Dietzel sum to about 26.5 %. When Lynch gob-fed machines were introduced in 1925, difficulties occurred because the forming time was longer and the glass too short: it was necessary to further increase alkali and to decrease CaO content. In the second half of the 20th century the production of champagne bottles was fully mechanized, the composition did not change significantly.

At first champagne bottles were coloured by manganese and iron. After 1956, when Morocco became independent, the price of manganese ore increased significantly and it was replaced by potassium dichromate. Between 1962 and 1968 a combination of $MnO+Fe_2O_3+Cr_2O_3$ was used but use of chromium alone began in 1968. About 1980 melting in reducing conditions was introduced, FeO content was increased although total iron content was decreased; that improved protection against UV rays and Cr^{6+} was eliminated from the glass.

15. Conclusion

Container glasses were developed from commonly used hollow-ware glasses during approximately the first half of the nineteenth century. Initially, low price of raw materials and good fusibility were demanded. The glass blowers were able to accommodate a strong tendency to devitrify and disadvantageous viscosity-temperature relations. The subsequent development of container glass compositions advanced along two lines. The older line was oriented towards cheap products, that is to say using greenish tinted glasses of "natural colour". Good fusibility at low price was attained by means of very high lime contents (up to 20 % CaO), in so-called "French hand-worked glasses". By the end of the nineteenth century, those glasses had been improved by use of mineral raw materials containing almost as much alkali as alumina to give glasses with 5 to 7 % Al_2O_3 and a certain amount of iron which coloured the glass. "German bottle glass for hand production" was developed from about 1860 and both glasses existed side by side until about 1930. High RO content (25 %) was popular in France, England, Italy and also in Russia; low RO content (about 10 %) in Germany, Hungary, Czechia and USA. The alumina content satisfied new requirements for chemical durability. Such glasses were used on the first semi-automatic machines. Nevertheless these glasses evolved becoming longer; between 1902 and 1929 Dietzel's sum decreased by about 3 % to average 25.5 % and RO content by 5 % (to 12.7 %), these changes being to be compensated by an increase of alkali content by 2 to 12.7 %. Such green glasses were used on Owens machines for about three decades [31].

A notable special development was the high-alumina glasses of König [24 and 25] melted in Czechia, beginning in 1929. Thanks to alumina contents up to 17 % Al_2O_3 it was possible to supply alkali almost entirely by phonolite. In glasses coloured brown by manganese it was possible to omit soda or saltcake from the batch. Unfortunately the increased alumina content impaired the acid resistance of the glass and the alumina content had to be decreased to 13 to 14 % Al_2O_3. This type of glass was formed on Owens machines in Czechia between 1929 and 1947. After 1958, a version modified by Kříž [30] having 10.3 % Al_2O_3 and 14.2 % R_2O was worked for some years on IS machines. It appears that such high-alumina glasses are one of the rarely used possibilities for making cheaper green container glass.

Brown glasses coloured by manganese followed the same line of development. The oldest analyses come from 1887; these glasses were melted in Germany, Bohemia, and probably also in France. Imported pyrolusite was expensive, so this type of glass was not widely produced. Shortage of manganese during World War II stopped its production but it was used on suction machines in Czechia up to 1990. The composition of manganese glasses was similar to the others but the favourable effects of iron and manganese oxides on melting allowed the alkali contents to be held at a lower level (9.9 % R_2O).

The world-wide development of cheap green iron-manganese glasses was complete by about 1960. It was characterized by Al_2O_3 contents of 6 to 8 % (except in the high-alumina glasses already discussed). The glasses had Dietzel's sum of 25.5 to 28 %. Iron content decreased step-by-step from 4 to 1.8 % Fe_2O_3, the colour (olive green) was achieved by 0.3 to 1.1 % MnO. About 5 % MgO was found in the oldest French glasses; contents of more than 1 % MgO were found in about half of glasses of this type. Additions of other elements were rare but BaO and F_2 were occasionally seen.

The other main type is colourless flint glass melted from pure raw materials. Its manufacturers generally did not also melt the cheaper green glass, so the two types evolved separately. The oldest analysis come from 1868 and this type of glass continued with only minor changes up to about 1960. Even the oldest of these glasses had soda as the main alkali, K_2O contents not exceeding 3.4 %. These glasses have been characterized by 1 to 2 % Al_2O_3, $RO+R_2O$ 25.5 to 26 % and by high alkali content, about 16 %. The oldest analysis of white container glass containing magnesia was from 1924 but some white flints were still melted without MgO after 1970. From the earliest days up to about 1950 white container glasses were often decolourized by manganese (additions of about 0.07 to 0.25 % MnO). Selenium was also used. White container glasses also show their relationship to colourless glasses in more numerous additions of other supposedly beneficial constituents such as BaO and B_2O_3. Older glasses were refined with arsenic or Sb_2O_3, the others by sulphate.

Longer white glasses for machine production spread to Europe from the USA together with new feeder machines after 1920. These longer or softer American glasses had high alkali contents, up to 16 to 17 % R_2O in 1936. Their R_2O contents later decreased but were still higher than in European flints. Alumina content was about 0.9 % Al_2O_3, always with 1 to 3 % MgO. Other parameters corresponded with European white glasses. Additions of BaO (65 % of glasses in 1960, averaging 0.7 % BaO) and of B_2O_3 (up to 0.5 %) were common in American glasses.

Amber glass has generally been a white glass melted in reducing conditions. It has been melted for a long time, the first analyses being from France and Hungary in 1879, but it became widespread only after 1913, when the compositions of amber glass and white flint were almost the same.

Both types of container compositions converged after 1970. When production became entirely by machine, iron olive green was replaced by clear emerald green coloured by chromium. White flint became the dominant type, the rest being divided between amber and green. From 1970 to 1990 container glasses of all kinds have had Dietzel's sum 25.60 $\pm$ 0.30 % and alkali content (11.40 $\pm$ 0.20) % R_2O. Alumina content has averaged (2.05 $\pm$ 0.30) % Al_2O_3 . Green glasses have been coloured by an average of 0.15 % Cr_2O_3, white glasses have been decolourized by selenium and reduced by carbon. Green and amber glasses have had rather higher iron content than white glasses. The usual practice is to melt from a combination of limestone and dolomite. The glasses have constant Dietzel's sum and constant sum of $Al_2O_3+SiO_2$.

Up to 1939 all groups of container glasses (except of ambers) contained 0.50 to 0.60 % SO_3. Since 1946 SO_3 decreased to about 0.18 % SO_3 in 1990. The reasons for this are increased melting temperatures, reducing refining, and environmental constraints.

The trend for compositions to converge is evident between both the different colours and individual producers. Variations of compositions have decreased step-by-step and both average compositions and ranges of variations have become similar throughout the world. Convergence of compositions significantly helps glass recycling but high recycling rates limit the possibility of individual manufacturers changing compositions. Environmental factors now have important effects on glass melting; for example, even small additions of fluorine have been eliminated, and even lower chlorine contents have to be decreased. For commercial reasons other container glass colours have been introduced, based on both various dark green and amber glasses and their combinations, also blue glasses. To achieve long shelf lives for the contents low UV-transmissivity is prescribed; chemical durability standards have increased. The trend is always to decrease alkali content and to make shorter glasses but the tendency for compositions to converge also continues.

A similar review of pressed glass compositions is being prepared.

*

The author expesses his appreciation to an unknown reviewer for editorial help especially in respect of the usage of English.

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