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Melt viscosities in the system Na-Fe-Si-O-F-Cl: Contrasting effects of F and Cl in alkaline melts

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ABSTRACT

The shear viscosities of melts in the system Na-Fe-Si-O-F-Cl were determined over a wide range of temperatures (400–1200 °C) at 1 atm pressure in air. The compositions are based on the addition of Fe_2O_3, $FeCl_3$, and FeF_3 to a base melt composition corresponding to sodium disilicate ($Na_2Si_2O_5$). Viscosities were determined using concentric cylinder and micropenetration methods and measurements span the range of $10^{0.5}$ to 10^{11} Pa·s. The chemical compositions of these melts were analyzed after the viscometry determinations. The iron is fully oxidized under the conditions of the viscometry. Although F and especially Cl are volatile elements in silicate melts, levels of Cl and F up to over 3 and 4 wt%, respectively, were stabilized in these melts, assisted presumably by the presence of Fe^{3+}. Although some volatilization occurred during the original synthesis of these samples, none occurred during viscometry. The anionic substitutions Cl_2O_{-1} and F_2O_{-1} have very different influences on the viscosity. The F_2O_{-1} substitution causes a drastic decrease in viscosity over the entire investigated range whereas the Cl_2O_{-1} substitution causes a much smaller decrease in viscosity in the high viscosity range and a slight increase in viscosity in the low viscosity range. As a consequence, minor to major element abundance of Cl in strongly peralkaline undersaturated volcanic rocks are not likely to significantly influence melt viscosity.

INTRODUCTION

The large number of elements present as cations in the silicate melts involved in igneous processes stands in stark contrast to the dominant anion being a single element: O. Nevertheless, despite the relatively limited substitutions permitted among the anions, significant deviations from the approximation of a silicate melt as an oxide melt occur in nature as demonstrated by the significant levels of F in topaz rhyolites (i.e., >1 wt% F; Burt et al. 1982) and Cl in pantellerites (e.g., >1 wt% Cl; Lowenstern 1994).

A wealth of data exists on the influence of cationic substitutions, and mixing on the viscosity of silicate melts was extensively investigated by the systematic addition of relatively non-volatile oxide components (see review by Richet and Bottinga 1995). The relatively limited solubility of anionic substitutions in geologically relevant melts makes experimental investigations into the influence of anionic substitutions on melt viscosity difficult. High quality data, most easily obtained at low pressures, are few in number because of the high volatility of many anionic components.

The influence of F on the properties of silicate melts was relatively extensively investigated (see reviews by Dingwell et al. 1985; Dingwell 1988; Lange 1994). In contrast, the influence of Cl on melt viscosity is poorly constrained and there are no high precision, low pressure studies on chemically well-constrained samples in these relatively volatile melt systems, which are compositionally unstable at high temperatures and 1 bar. A recent investigation of the geochemistry of ‘‘paralavas’’ (melt created naturally by a fusion of sedimentary rock, Cosca et al. 1989) from White Island demonstrated very high chlorine levels (up to 1.7 wt%) in Fe-rich melts co-erupted with peralkaline lavas (Wood and Browne 1996). The assumption that the presence of Fe could enhance the stability of Cl in high temperature (based on the compositions reported by Wood and Browne 1996), led us to design the present study to compare directly the influences of F and Cl on the viscosity of a silicate melt.

EXPERIMENTAL METHODS

The compositions are based on the addition of Fe_2O_3, $FeCl_3$, and FeF_3 to $Na_2Si_2O_5$. $Na_2Si_2O_5$ was initially synthesized by fusion of a mixture of Na_2CO_3 and SiO_2 powders at 1200 °C in a thin-walled platinum crucible in air for several hours. During fusion the melt was stirred at 10 rpm using a $Pt_{80}Rh_{20}$ spindle. The glassy product of this fusion was ground in an agate mortar. Next, this glass powder was mixed with powders of Fe_2O_3, $FeCl_3(6H_2O)$, and FeF_3, and then re-fused in viscometry crucibles (see below) at the temperatures corresponding to the highest temperature viscosity data presented here. The samples

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were stirred using the viscometry spindle (described below) during the second fusion.

The low range viscosities were obtained using the concentric cylinder system and methods described by Dingwell (1989a and 1989b; 1991). Viscosity is determined in steps of decreasing temperature with a final reoccupation of the highest temperature data point as a check against instrumental or sample drift. The system is calibrated against DGG-1, a viscosity standard glass. The viscosities are accurate to ±5%. The viscometer head used was the Brookfield DVIII+, which has a full-scale torque of 7.2 $\times 10^{-2}$ N·m. The spindle used was the $Pt_{80}Rh_{20}$ low viscosity spindle described by Dingwell and Virgo (1988). After concentric cylinder viscometry, the samples were permitted to cool in the crucibles. From the glassy products, with the exception of sample NS2FeF3 (see below), which could not be quenched to a glass, cores were drilled with a diamond coring tool and the cores were cut into disks and polished in preparation for micropenetration viscometry.

The high range viscosities were measured using a micropenetration technique (Hess et al. 1995; Dingwell et al. 1996), wherein the rate at which an Ir-indenter under a fixed load moves into the melt surface is determined. These measurements were performed in a BÄHR DIL 802 V vertical pushrod dilatometer. The sample is placed in a silica rod sample holder under an argon gas flow. The measurements are performed over indentation distances of less than 100 μm. The technique yields an absolute determination of viscosity up to 1100 °C in the range of 10^8 to $10^{11.5}$ Pa·s. Viscosities determined on the DGG-1 standard glass using 3 mm samples were reproduced within an error of ±0.06 log units. All samples were heated up to the measuring temperature, held at this temperature for 1 h to allow thermal equilibration and complete structural relaxation of the sample, and then the viscosity measurement was performed. After the measurement, each sample was cooled at approximately 20 K per minute.

Chips of the quenched glassy products of the concentric cylinder viscometry were analyzed using electron microprobe methods for composition and homogeneity (Table 1). Great care was taken to avoid beam-induced volatilization of Na, and special attention was paid to the resulting stoichiometries of the glasses as discussed below. A continuous raster of a defocused beam was used. The O contents were derived (not by difference) but stoichiometrically from the analyzed elements as equaling $2Si + 0.5Na + 1.5Fe - 0.5Cl - 0.5F$. These stoichiometrically derived O contents yield complete analytical totals that lie within 99 to 101 wt%. For subsequent stoichiometric comparisons of the data several elemental ratios are included in Table 1. The atomic fractions of the elements, derived from the weight fractions, were used to calculate values for the anionic atomic fraction of F and Cl as F/(F+O) and Cl(Cl+O), respectively. Also, the cationic atomic fraction of Fe as Fe/(Na+Fe+Si) is presented. This value is used to normalize the comparisons of oxide, chloride, and fluoride additions on melt viscosity below. The fraction of added F and Cl lost via volatilization is also included in Table 1. Fluorine loss ranges from 15 to 25% of the amounts added whereas Cl loss ranges from 30 to 55%. The Na/(Na+Si) ratio is constant at 0.472 ± 0.05 except for the most Cl-rich sample for which significant Na was lost, presumably as NaCl, during fusion. Despite volatility, up to 3.2 wt% Cl was stabilized in these melts at 1 atm in air. From our previous attempts to synthesize Cl-bearing silicate melts at 1 atm pressure, this stability is remarkable. We surmise that the presence of Fe^{3+} stabilizes Cl in these melts. The subsequent discussion of the stoichiometric influences of Cl and F on melt viscosity is based on the actual analyzed compositions of the glasses presented in Table 1. For the one melt, which could not be quenched to a glass, sample NS2FeF3, only high temperature viscosity data were obtained and no analysis is available. The viscosity data are presented for completeness but are not used in the quantitative discussion of the trends.

The oxidation state of Fe in these melts was investigated using ^{57}Fe Mössbauer spectroscopy on glasses quenched from the highest temperature at which viscosity measurements were performed by dipping an alumina rod into the melt and quenching the extracted melt bead in water. Those spectra indicate that the Fe is virtually completely oxidized (Dingwell and McCammon, unpublished data). Under these viscosities, no significant oxidation during the quench is expected. Thus, all stoichiometric calculations above and conclusions drawn below are based on fully oxidized melts. This important observation simplifies the discussion of the compositional trends of viscosity below.

RESULTS AND DISCUSSION

The concentric cylinder and micropenetration viscometric determinations (Table 2) are illustrated as a function of the reciprocal absolute temperature (Fig. 1). The concentric cylinder results cluster closely, except the two most F-rich melts. The activation energies are also constant in this temperature range. The addition of each of Fe_2O_3, $FeCl_3$, and FeF_3 results in reduced viscosity. The reduction for F addition is strong, up to over a log unit at these concentration levels, whereas the reductions for the chloride and oxide additions are small and roughly similar in magnitude. At the lower temperatures and higher viscosities of the micropenetration viscometry (Fig. 1, Bottom), the trends are slightly different. The addition of a small amount of Fe oxide increases melt viscosity whereas larger amounts decrease the melt viscosity. This effect may influence the halide results in that the initial addition of Cl results in a very slight melt viscosity decrease. The effects of the F additions are again much larger than those of the oxides or chlorides although the activation energies are similar. The combined data sets for the complete temperature-dependent viscosities of the eight compositions (Fig. 2) show significant non-Arrhenian dependence of the viscosity on temperature. The sim-

TABLE 1. Chemical compositions of quenched glasses determined by electron microprobe

	NS2	NS2FeF2	NS2FeF1	NS2FeO2	NS2FeO1	NS2FeCl2	NS2FeCl1
F (wt%)	0	4.14	1.54	0	0	0	0
Fe (wt%)	0	5.16	2.46	7.17	2.25	5.11	2.31
Cl (wt%)	0	0	0	0	0	3.16	2.51
Na (wt%)	23.64	21.59	22.65	21.65	22.91	19.9	22.11
Si (wt%)	31.73	29	30.61	28.78	30.77	30.06	30.58
O (wt%)	44.36	41.01	43.15	43.39	43.98	42.64	42.95
Total	99.73	100.90	100.41	100.99	99.91	100.87	100.46
F (at)	0	0.2178	0.0810	0	0	0	0
Fe (at)	0	0.0923	0.0440	0.1283	0.0402	0.0914	0.0413
Cl (at)	0	0	0	0	0	0.0891	0.0708
Na (at)	1.0282	0.9391	0.9852	0.9417	0.9965	0.8655	0.9617
Si (at)	1.129	1.032	1.090	1.024	1.095	1.070	1.089
O (at)	2.772949	2.563	2.697	2.712	2.749	2.665	2.684
F (at%)	0	0.04496	0.01654	0	0	0	0
Fe (at%)	0	0.01906	0.00899	0.02670	0.00825	0.01913	0.0853
Cl (at%)	0	0	0	0	0	0.01864	0.01460
Na (at%)	0.2085	0.1937	0.2011	0.1958	0.2041	0.1810	0.1984
Si (at%)	0.2291	0.2131	0.2225	0.2132	0.2244	0.2238	0.2246
O (at%)	0.5623	0.5290	0.5507	0.5641	0.5631	0.5573	0.5537
F/(F + Cl + O)	0	0.0783	0.0291	0	0	0	0
Cl/(F + Cl + O)	0	0	0	0	0	0.03236	0.02569
Fe/(Fe + Na + Si)	0	0.04475	0.02078	0.06127	0.01889	0.04512	0.01976
Na/(Na + Si)	0.4764	0.4762	0.4747	0.4788	0.4762	0.4470	0.4689
lost F		0.01223	0.01043				
lost Cl						0.03875	0.01099
fraction Fe as FeF3		0.8649	0.7366				
fraction Fe as FeCl3						0.4550	0.7004

Note: Electron microprobe analyses performed with a defocused raster. Standards are albite (Na, Si), hematite (Fe), vanadinite (Cl), and Apatite (F). Typical standard deviations on the analyses in wt% are (Na, 0.36; Fe, 0.06; Si, 0.47, and O, 0.45; for Cl, 0.04 at 2.5 wt% and for F, 0.27 at 1.8 wt%).

TABLE 2. Viscosity data determined by concentric cylinder viscometry and micropenetration

Sample	*T* °C	log η Pa·s	Sample	*T* °C	log η Pa·s	Sample	*T* °C	log η Pa·s
no. NS2	1000.0	2.09	no. NS2FeO2	900.0	2.33	no. NS2FeF1	1100.0	1.15
	950.0	2.37		850.0	2.69		1075.0	1.27
	900.0	2.69		800.0	3.10		1050.0	1.39
	850.0	3.05		495.4	9.20		1025.0	1.51
	800.0	3.48		474.1	10.20		1000.0	1.63
	750.0	3.97		463.9	10.76		975.0	1.76
	700.0	4.57	no. NS2FeCl1	1000.0	1.98		950.0	1.90
	508.4	8.89		950.0	2.26		925.0	2.05
	490.6	9.58		900.0	2.57		900.0	2.21
	470.9	10.60		850.0	2.94		875.0	2.38
no. NS2FeO1	920.0	2.37		800.0	3.36		850.0	2.57
	900.0	2.49		750.0	3.86		825.0	2.76
	880.0	2.63		700.0	4.47		800.0	2.97
	860.0	2.77		498.8	9.22		474.4	8.98
	840.0	2.92		485.1	9.74		455.0	9.78
	800.0	3.26		469.5	10.58		436.0	10.91
	780.0	3.44		457.7	11.19	no. NS2FeF2	975.0	1.28
	760.0	3.64	no. NS2FeCl2	1000.0	1.92		950.0	1.42
	740.0	3.86		975.0	2.01		925.0	1.56
	720.0	4.10		950.0	2.14		900.0	1.71
	512.5	8.86		925.0	2.29		875.0	1.87
	492.5	9.95		900.0	2.46		850.0	2.04
	470.0	10.74		875.0	2.63		459.6	8.89
	469.9	10.90		850.0	2.83		436.5	9.81
no. NS2FeO2	1200.0	0.89		825.0	3.04		415.1	11.20
	1150.0	1.07		800.0	3.25	no. NS2FeF3	1000.0	0.71
	1100.0	1.27		493.5	9.05		980.0	0.81
	1050.0	1.49		471.0	10.05		960.0	0.92
	1000.0	1.73		468.0	10.20			
	950.0	2.01		454.3	10.95			

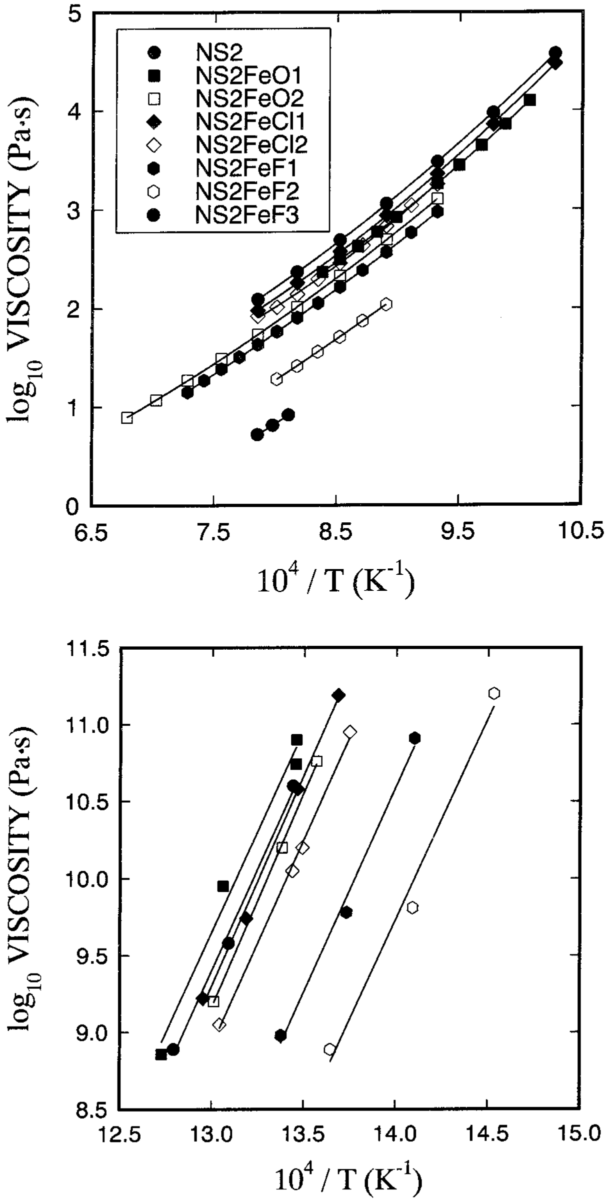

FIGURE 1. Viscosity-temperature relationships of F and Cl-bearing melts. (top) High temperature viscosity data obtained using concentric cylinder viscometry. (bottom) Low temperature viscosity data obtained using the micropenetration method. Symbols are given in the inset.

ilar activation energies from Figure 1 leads to the observation that the non-Arrhenian behavior of all investigated compositions is very similar. That is, the curves in Figure 2 are close to being parallel over the entire temperature range, and thus, the molar influences of F and Cl on melt viscosities do not vary greatly with respect to one another as a function of the absolute viscosity or temperature. These data were fit to the Tammann-Vogel-Fulcher (TVF) equation (Table 3). Viscosities derived from the fits of all investigated F and Cl-bearing melts, with the exception of NS2FeF3 as noted above (Fig. 3) show that the weight fraction influence of F on the viscosity of such depolymerized alkalic melts is much larger than that of Cl at all temperatures.

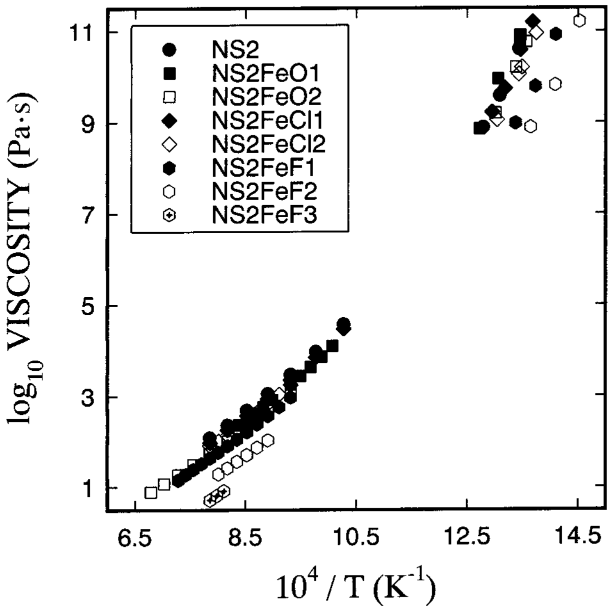

FIGURE 2. Viscosity-temperature relationships from the combined concentric cylinder and micropenetration viscosity data set. TVF-equation fits to these curves serve as the basis for further comparisons of the data based on their normalized compositions.

Further comparisons of the data require stoichiometric recalculations of the melt compositions. To compare the molar influences of the exchanges of Cl and F for O in these melts, we recast the melt compositions in Table 1 in terms of two essential compositional variables used for normalization, the anionic fraction of the halide [i.e., X/(X+O) where X = F, Cl] and the cationic fraction of Fe [i.e., Fe/(Na+Fe+Si)]. The first variable permits a molar comparison of the actual analyzed values for the anionic fraction of halides in the melts. The second variable accounts for the variable Fe contents of the melts as a mole or atomic fraction of the cations. Thus we derive a

TABLE 3. Fit parameters for the Tamman-Vogel-Fulcher equation

Sample	a $\log_{10}$ Pa·s	b K	c K	FitStdErr*
NS2	−1.745	2971	502.52	0.03
NS2FeO1	−2.053	2981	512.76	0.08
NS2FeO2	−2.241	3063	501.14	0.01
NS2FeCl1	−1.919	3020	500.44	0.02
NS2FeCl2	−1.937	2985	495.37	0.02
NS2FeF1	−2.354	3228	463.96	0.04
NS2FeF2	−2.537	2981	470.43	0.06

Note: TVF equation is $\log_{10}\eta = a + b/(T - c)$.

* FitStdErr denotes the actual least squares error of fit and is the root of mean square error.

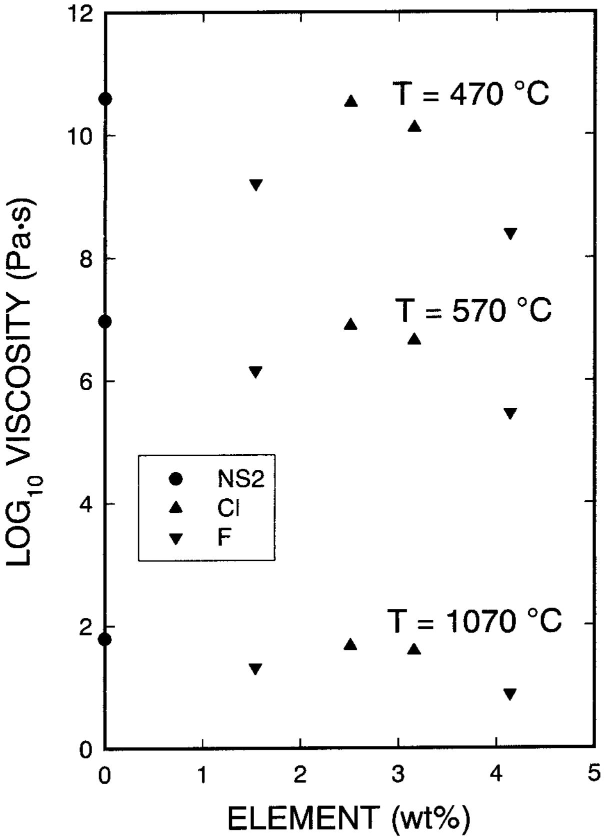

FIGURE 3. Isothermal comparison of the influence of weight percent additions (analyzed values) of F and Cl on the viscosity of the Na-Fe-Si-O melts at 470, 570, and 1070 °C. The data are interpolated from the TVF fits to the combined data set of Figure 2. These data are not corrected for variable Fe content but the relative influences of F and Cl are already apparent.

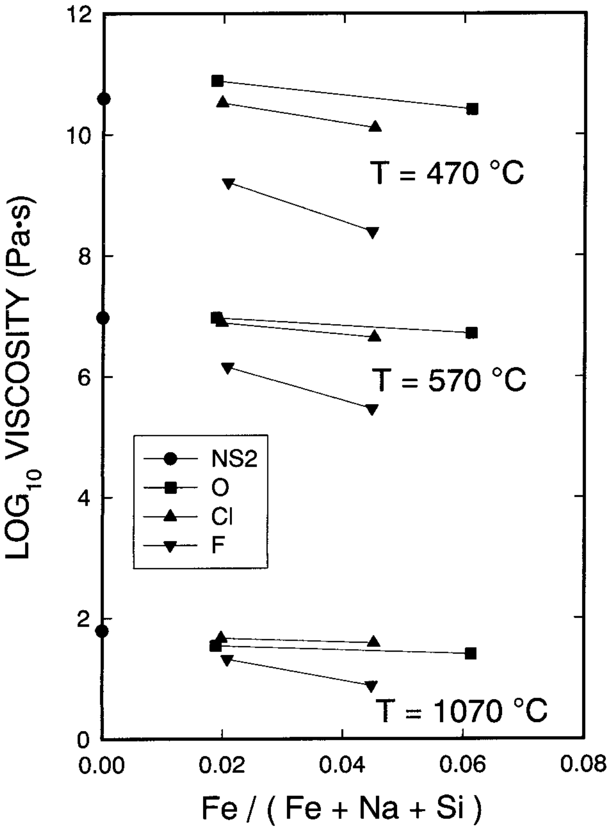

FIGURE 4. Isothermal comparison of the viscosities of melts with added Fe oxide, Fe chloride, and Fe fluoride expressed as a function of the Fe content [cationic fraction as Fe/(Fe+Na+Si)]. This diagram is used as a normalization basis for Figure 5.

direct comparison of the molar influences of F and Cl by normalizing to a common Fe content of the melt.

Before proceeding with the normalized comparison, two important points must be made. First, the Na/(Na+Si) content of the melts does not vary significantly except for one melt composition, NS2FeCl2. As noted above, this is presumably due to NaCl volatility. Even with the slight shift in the Na/Si ratio caused by this effect, the viscosity of the base Na-Si-O melt composition, carefully investigated in detail recently by Knoche et al. (1994) should not shift significantly for the purposes of comparison in the present study. This independence was a reason for choosing the Na-Si-O system for this study. Second, the magnitude of the influence of Fe oxidation state on viscosity (Dingwell and Virgo 1987; Dingwell 1991) combined with the relatively low concentrations of Fe and the invariance of the oxidation state rule out Fe oxidation state as a variable controlling the relative viscosities here. These observations allow normalization of the viscosity effects of F and Cl, as follows.

In Figure 4, the viscosities from the fits of Table 3 are plotted for the temperatures 470, 570, and 1070 °C vs. the Fe/(Fe+Na+Si) content of the melts. This figure is used as a normalization basis for Figure 5 wherein the difference between the influence of the Fe oxide on the viscosity and that of the Fe chloride and Fe fluoride (corrected for volatilization) is plotted for the same three temperatures as a $\Delta\log_{10}$ viscosity. The curves of Figure 5 now provide the relative molar influences of the exchange operators F_2O_{-1} and Cl_2O_{-1} on the viscosity of a single fully analyzed silicate melt series.

The effect of F in reducing the viscosity of the relatively silica-poor melts of this investigation might be surprising considering the depolymerized state of the base melt compositions. It is known that the polymerization of the silicate melt can strongly affect the magnitude and even the sign of the influence of many components on melt viscosity (e.g., Al_2O_3, P_2O_5, Toplis and Dingwell 1996; Toplis et al. 1997a and 1997b). Consideration of the structural role of F raised speculation that nominally depolymerizing elements like F might in fact lead to an increase in the viscosity of depolymerized melts such as $CaMgSi_2O_6$ (Luth 1988). Dingwell (1989) demonstrated that this is not the case for diopside melt with added F and the present results are consistent with those in sign

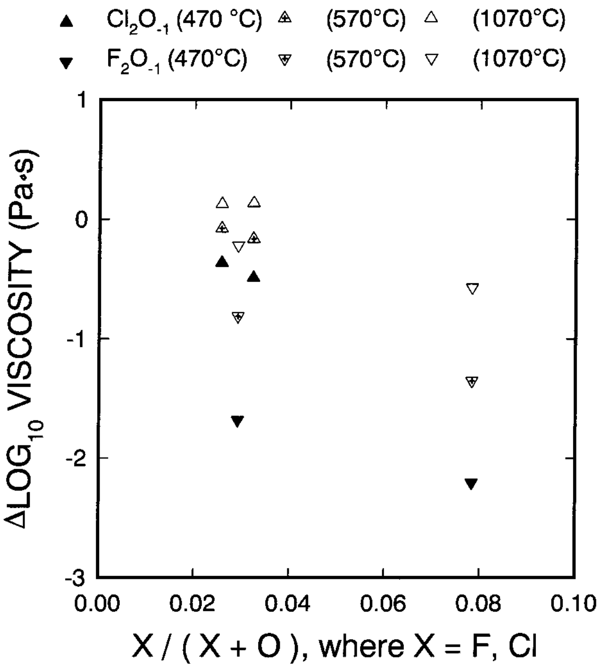

FIGURE 5. The relative influences of Cl_2O_{-1} and F_2O_{-1} on the viscosities of Na-Fe-Si-O melts determined by comparison of the influences (corrected for volatilization) of the iron oxide, fluoride, and chloride components presented in Figure 3 on viscosity. Also included for comparison are limited data on the influence of water on sodium disilicate melt viscosity.

and magnitude. No melt has yet been reported where the addition of F in substitution for O or as a fluoride component has led to an increase in viscosity. In contrast then we observe here the very small influence of Cl on the viscosity of Na-Fe-Si-O melts. In contrast to the case for F, interpretation of early work on the influence of Cl or chloride on the viscosity of low silica slags was clouded by the fact that compositions were poorly documented. This raised the possibility of Cl volatility distorting the conclusions. Nevertheless, these early works indicated a very slight influence and a possible increase in viscosity with chloride addition (Bills 1963; Hirayama and Camp 1969). Very recently, Baker and Vaillancourt (1995) have stated that Cl increases the viscosity of albite melt at high pressures. That possibility appears to be confirmed in principle, at the highest temperatures of the present investigation.

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