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Effect of Additional Hydrofluoric Acid on the Conductivity of Concentrated Fluorosilicic Acid Aqueous Solutions

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The specific conductivities (κ) of the $H_2SiF_6 + HF + H_2O$ ternary system have been measured in a wide range of compositions, $8 < 100w_{H_2SiF_6} < 45$ and $0 < 100w_{HF} < 12$, at 25 and 35 °C by an automated system for conductimetric titration based on the "electrodeless conductivity" technique. An empirical model of the type $\kappa = f(100w_{H_2SiF_6}, 100w_{HF})$, which correctly fits the results at the two temperatures studied, is also presented.

Introduction

Although many articles can be found in the literature dealing with the electrical conductivity of dilute systems, not so much information is available with respect to concentrated electrolyte solutions (Lobo, 1989). Full sets of (κ, c, t) data from concentrated systems is usually restricted to solutions of a unique electrolyte in water at 25 °C (Barthel, 1985), and frequently, large differences are observed when results given by different authors (Lobo, 1986) are compared. The main reason for that poor agreement probably arises from the difficulty of completely avoiding the polarization effects on the surface of the electrodes when measuring high conductivities (Lobo, 1985).

In 1947, a new technique based on the measurement of low electrical intensities induced in the sample solution by an alternating current of not very high frequency (~20 kHz) and in which the use of classical electrodes is absolutely avoided, the so-called electrodeless conductivity or induced conductivity, was proposed by Relis (1947, 1951). No polarization process interferes with the conductivity measurement since there is no direct contact between the electrodes and the sample solution. Consequently, the technique becomes a very useful tool to measure high conductivities. Electrodeless conductivity has been extensively used in industrial applications (Foxboro, 1977; Shaw and Light, 1982) but, due to the large dimensions of the measuring probe, it was supposed not to be a very appropriate technique in laboratory work conditions. In the last years a reduction in the dimensions of the probe has been achieved and, therefore, a laboratory-scale automated system for conductimetric titration based on the electrodeless conductivity technique was first presented by some of the authors of this paper (de Diego et al., 1994). Recently, very precise $(\kappa, 100w)_t$ data corresponding to HBF_4, H_2SiF_6, and H_2TiF_6 aqueous electrolytic systems have been measured in a wide range of temperatures by means of that automated system (de Diego et al., 1996a).

The actual tendency in the chemical industry goes toward highly automated plants with continuous manufacturing processes. Control of such processes requires "in-line" analysis procedures, e.g., measurement of any changing parameter in the occurring chemical reaction. In those processes involving the presence of fluoride in acidic medium, the measurement of the pH is, at least, embarrassing and, therefore, alternatives must be proposed (Derivados, 1995). Electrical conductivity may be a potential candidate to substitute pH glass sensors. H_2SiF_6 aqueous solutions with relatively high amounts of dissolved HF are a very commonly obtained byproduct in the fluorine derivatives manufacturing industry. Not only environmental but also economical reasons make advisable the separation, recovery, and recycling of both H_2SiF_6 and HF (Derivados, 1995). In order to check the validity of conductivity as the control parameter in a possible automated continuous process treating such kinds of byproducts, the conductivity of the $H_2SiF_6 + HF + H_2O$ ternary system has been measured at two temperatures, 25 and 35 °C, in a wide range of compositions ($8 < 100w_{H_2SiF_6} < 45$ and $0 < 100w_{HF} < 12$), making use of an automated system for conductimetric titration based on the electrodeless conductivity technique.

Automatic computing of the composition of a chemical system at real time needs a functional relation among the measured property, the concentration of each component of the solution, and the temperature. In a previous paper, an empirical model of the type $\kappa = f(100w, t)$ to fit (κ, $100w$, t) data referring to a unique electrolyte in water solution was proposed by the authors (de Diego et al., 1996b). Now, an empirical equation ($\kappa = f(100w_{H_2SiF_6}, 100w_{HF})_t$) is presented, which correctly fits the experimental data at the two temperatures studied.

Experimental Details

Materials and Reagents. The concentrations throughout the text are in mass percent ($100w$) unless otherwise stated. Three stock solutions of different compositions were prepared by dissolving the appropriate mass of SiO_2 (s) (Merck, p.a.) in an excess of an HF aqueous solution ($100w_{HF} = (48.7 \pm 0.1)$) delivered by the enterprise Derivados del Fluor, S.A. This HF solution was obtained by purifying a more concentrated one by a method resembling that of Mattison (1972), the resulting HF solution being of very high quality, the so-called electronic grade solution.

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Table 1. Compositions and Densities of the H_2SiF_6 (1) + HF (2) Aqueous Stock Solutions

stock solution	$100w_1$	$100w_2$	ρ/g·cm^{-3}
I	45.2 ± 0.1	1.4 ± 0.1	1.445 ± 0.002
II	41.3 ± 0.1	6.2 ± 0.1	1.421 ± 0.002
III	35.8 ± 0.1	11.9 ± 0.1	1.388 ± 0.002

Very low contents of typical impurities in such solutions are referenced in the product sheet (Derivados, 1994):

$$100w_{H_2SiF_6} < 0.0005 \qquad 100w_{Fe} < 0.0001$$

$$100w_{Cl^-} < 0.0005 \qquad 100w_{phosphates} < 0.0001$$

$$100w_{sulfates} < 0.0001 \qquad 100w_{Cu} < 0.0001$$

$$100w_{sulfides} < 0.0002 \qquad 100w_{As} < 0.000\,005$$

$$100w_{heavy\ metals\ (as\ Pb)} < 0.000\,05$$

The analysis (Ryss, 1956) of the stock solutions prepared in this way yielded the compositions appearing in Table 1. The density of each solution, also collected in Table 1, was measured too by weighing a known volume of solution (5 cm^3 Justor DG 5000 micropipet, accurate to ±0.001 cm^3) by a Mettler AJ150 balance, accurate to ±0.0001 g at constant temperature. MilliQ quality water (Millipore water purification system) with a specific conductivity lower than 0.05 μS·cm^{-1} was used throughout. The pipets, micropipets, and autoburets were calibrated before use by weighing the mass of the delivered water at constant temperature by a Mettler AJ150 balance.

The three stock solutions, and several dilutions of them, were conductimetrically titrated, making use of an automated system based on the electrodeless conductivity technique described previously (de Diego et al., 1994, 1996a). In the electrodeless conductivity technique, a coil, acting as a primary transformer, is powered with a constant ac voltage; the latter induces an alternating current within the surrounding sample solution, the magnitude of which is a function of the solution conductivity. That current is detected by a second coil acting as a secondary transformer. The automated system was calibrated with HCl aqueous solutions (Fluka, p.a.) following the procedure described elsewhere (de Diego and Madariaga, 1996). HCl was purified before use by a method similar to that of Mattison (1972) and it was analyzed (Jeffery et al., 1989) by volumetric titration with Na_2CO_3 (Merck, p.a.) and methyl orange (Merck, p.a.) as indicator.

Fitting of the experimental data to the empirical model was carried out by NLREG (Sherrod, 1995), a nonlinear regression program which minimizes the sum of squares of the absolute errors between calculated and experimental values and which is based on an algorithm published by Dennis et al. (1981).

Experimental Procedure. In order to cover a wider range of composition, and taking into account the geometrical limitations due to the dimensions of the measuring probe and the titration vessel (minimum volume needed to make a conductivity/temperature measurement and the maximum volume of solution allowed inside the vessel), three different titrations were carried out per each stock solution. In all of them milliQ quality water was used as titrant. The solution checked in the first titration was one of those characterized in Table 1. The resulting solutions after first and second titrations finished were respectively starting points for the second and third ones. This procedure was repeated for each stock solution at two different temperatures, close to 25 and 35 °C.

Before each titration, the measuring probe was placed inside the empty vessel and thermostated at the working temperature, and zeroing of the system, as well as calibration of the temperature measurement by means of a precalibrated mercury thermometer, was attempted.

The procedure in each titration was as follows: a volume of (300.00 ± 0.25) cm^3 of the sample solution was placed into the thermostated vessel and time to reach thermal equilibrium was allowed to elapse. Twelve additions of water of about (20.00 ± 0.01) cm^3 each were made. After each addition a time, typically $2^1/_2$ h, elapsed in order to get chemical and thermal equilibrium inside the vessel. The conductivity (±0.2% of the measurement) and temperature (±0.1 deg) of the solution were measured. All the process was automatically controlled by CONDUCTO, a computer program for conductivity titration developed in our group. More details about the program, criteria for signal stability, waiting times and correction factors can be found elsewhere (de Diego et al., 1994; Cazallas et al., 1995; de Diego, 1996). The sample was magnetically stirred for all the experiments, and the temperature of the oil bath was kept constant at the working temperature within ±0.3 deg. Taking into account the whole experimental procedure, an overall precision better than ±0.5% of the measurement has been estimated for the experimental measurement of the specific conductivity. The composition of the solution inside the vessel was calculated by taking into account the composition and density of the starting solution in each titration and the total volume of water added at each point of the titration.

Results and Data Treatment

Experimentally measured (κ, $100w_1$, $100w_2$, t) data from the H_2SiF_6 + HF + H_2O ternary electrolytic system have been collected in Table 2.

Polynomials in the concentration (eq 1) have frequently been attempted to fit conductimetric data from electrolytic systems at constant temperature (Fedotov and Maksimova, 1971; Maksimova and Sergeev, 1974):

$$\kappa_t = \sum_{i=1}^{i} a_i (100w)^i \tag{1}$$

where a_i are adjustable parameters with no physical meaning.

As has already been shown elsewhere (de Diego et al, 1996a), a second-order polynomial is enough to explain the random (κ, $100w_{H_2SiF_6}$) experimental data at constant temperature. Moreover, the κ vs $100w_{HF}$ plot for the HF aqueous system in the $0 < 100w_{HF} < 48$ range can be conveniently considered as a straight line passing through the origin (Broderick, 1962). If we take into account all the possible interactions, the next general polynomial results for the concentration dependence of the electrical conductivity of the ternary system H_2SiF_6 (1) + HF (2) + H_2O (3) at constant temperature:

$$\kappa_t = a100w_1 + b100w_2 + c100w_1100w_2 + d(100w_1)^2 + e(100w_1)^2 100w_2 \tag{2}$$

where a, b, c, d, and e are adjustable parameters to be determined by data analysis. The best value for b can be previously calculated if the round conditions of the problem are taken into account. When $100w_1 = 0$, e.g., for the binary system HF + H_2O, eq 2 reduces to

$$\kappa_t = b100w_2 \tag{3}$$

Table 2. Measured Conductivities κ at Temperatures t for Different Mass Percents $100w$ of H_2SiF_6 (1) and HF (2), Obtained by Conductimetric Titration Assays

		$t = (25.3 \pm 0.2)$ °C		$t = (35.4 \pm 0.1)$ °C				$t = (25.3 \pm 0.2)$ °C		$t = (35.4 \pm 0.1)$ °C	
$100w_1$	$100w_2$	t/°C	κ/mS·cm^{-1}	t/°C	κ/mS·cm^{-1}	$100w_1$	$100w_2$	t/°C	κ/mS·cm^{-1}	t/°C	κ/mS·cm^{-1}
I Stock Solution											
45.2	1.4	25.7	542.3	35.2	621.3	21.3	0.7	25.4	703.0	35.5	786.3
43.2	1.3	25.6	585.1	35.2	666.7	20.5	0.6	25.3	691.3	35.5	772.2
41.4	1.3	25.8	621.1	35.3	705.6	19.8	0.6	25.2	679.6	35.4	756.3
39.7	1.2	25.7	651.5	35.4	738.6	19.1	0.6	25.3	667.5	35.5	741.1
38.2	1.2	25.7	676.3	35.4	767.5	18.5	0.6	25.2	656.0	35.3	725.6
36.7	1.1	25.6	697.1	35.4	790.3	17.8	0.6	25.2	643.4	35.3	711.1
35.4	1.1	25.5	713.0	35.4	807.9	17.8	0.6	25.1	638.5	35.6	716.1
34.2	1.1	25.5	726.0	35.3	821.8	16.8	0.5	25.3	617.9	35.7	692.0
33.0	1.0	25.5	735.7	35.2	830.9	15.9	0.5	25.3	596.9	35.7	669.2
31.9	1.0	25.4	741.6	35.2	837.2	15.1	0.5	25.3	576.7	35.7	646.3
30.9	1.0	25.3	745.3	35.1	840.2	14.4	0.5	25.3	557.5	35.7	624.2
30.0	0.9	25.2	748.3	35.2	843.3	13.8	0.4	25.2	539.0	35.7	603.2
29.1	0.9	25.2	750.2	35.2	845.8	13.2	0.4	25.2	521.5	35.7	583.2
29.1	0.9	25.2	754.3	35.2	846.6	12.6	0.4	25.1	504.8	35.8	564.8
27.7	0.9	25.4	754.2	35.3	846.1	12.1	0.4	25.1	489.2	35.7	547.1
26.4	0.8	25.4	749.2	35.3	841.9	11.7	0.4	25.1	474.0	35.7	530.2
25.2	0.8	25.5	741.7	35.3	835.6	11.2	0.3	25.0	459.9	35.7	514.4
24.1	0.7	25.5	733.0	35.5	856.0	10.8	0.3	25.0	445.8	35.6	499.4
23.1	0.7	25.4	724.2	35.5	814.9	10.5	0.3	24.9	433.4	35.5	485.1
22.2	0.7	25.4	713.3	35.5	800.6						
II Stock Solution											
41.3	6.2	25.6	597.4	35.4	680.6	19.2	2.9	25.3	672.6	35.5	749.0
39.4	5.9	25.6	631.7	35.4	718.7	18.5	2.8	25.2	660.7	35.4	735.8
37.7	5.6	25.6	660.8	35.4	749.3	17.8	2.7	25.1	647.3	35.4	721.2
36.2	5.4	25.5	683.7	35.5	775.8	17.2	2.6	25.0	634.7	35.5	707.6
34.7	5.2	25.5	702.5	35.5	796.5	16.6	2.5	25.1	622.6	35.4	693.2
33.4	5.0	25.4	717.3	35.4	811.5	16.1	2.4	25.0	610.4	35.3	680.2
32.2	4.8	25.4	728.2	35.5	824.3	16.1	2.4	25.3	606.3	35.3	690.1
31.0	4.6	25.3	735.6	35.5	832.3	15.3	2.3	25.3	586.1	35.3	666.3
30.0	4.5	25.3	742.1	35.6	838.0	14.5	2.2	25.3	565.1	35.4	643.2
29.0	4.3	25.4	745.3	35.5	840.8	13.8	2.1	25.3	545.2	35.4	620.9
28.1	4.2	25.3	746.8	35.5	840.7	13.1	2.0	25.3	526.4	35.4	599.8
27.2	4.1	25.3	746.5	35.4	840.4	12.5	1.9	25.2	508.9	35.4	579.7
26.4	4.0	25.3	746.1	35.4	840.8	12.0	1.8	25.2	492.6	35.5	560.0
26.4	4.0	25.3	741.6	35.4	838.3	11.5	1.7	25.2	476.9	35.5	542.5
25.0	3.7	25.3	735.0	35.2	832.3	11.0	1.6	25.1	461.7	35.5	525.4
23.8	3.6	25.3	727.0	35.4	822.0	10.6	1.6	25.1	448.1	35.6	509.3
22.7	3.4	25.3	717.9	35.4	809.5	10.2	1.5	25.0	430.8	35.5	493.9
21.7	3.2	25.3	707.7	35.5	795.4	9.9	1.5	24.9	419.1	35.4	478.7
20.8	3.1	25.3	696.1	35.6	780.0	9.5	1.4	24.9	409.3	35.3	465.1
20.0	3.0	25.3	685.0	35.5	764.7						
III Stock Solution											
35.8	11.9	25.4	648.7	35.2	735.0	16.4	5.4	25.3	628.0	35.6	699.4
34.2	11.3	25.4	672.0	35.1	760.5	15.8	5.2	25.3	614.5	35.7	684.6
32.7	10.8	25.5	689.3	35.3	779.1	15.2	5.0	25.2	601.1	35.5	668.3
31.3	10.4	25.5	702.8	35.3	792.7	14.7	4.9	25.2	588.0	35.5	655.6
30.0	10.0	25.4	712.4	35.4	802.8	14.2	4.7	25.1	575.1	35.5	642.9
28.9	9.6	25.2	719.8	35.3	810.7	13.7	4.5	25.1	562.9	35.4	629.3
27.8	9.2	25.3	724.0	35.3	815.2	13.7	4.6	25.3	564.4	35.4	628.3
26.8	8.9	25.2	726.4	35.2	817.1	13.0	4.3	25.4	544.1	35.5	605.0
25.9	8.6	25.2	727.7	35.3	816.6	12.4	4.1	25.5	524.6	35.6	582.4
25.0	8.3	25.1	726.5	35.2	815.5	11.7	3.9	25.4	505.8	35.6	560.7
24.2	8.0	25.1	723.8	35.3	812.8	11.2	3.7	25.4	487.3	35.6	540.9
23.4	7.8	25.0	720.2	35.2	808.1	10.7	3.5	25.4	470.8	35.6	521.5
22.7	7.5	25.0	716.4	35.2	803.4	10.2	3.4	25.4	455.0	35.6	503.8
22.7	7.5	25.4	713.6	35.4	802.1	9.8	3.3	25.4	440.2	35.6	487.0
21.5	7.1	25.3	704.6	35.3	791.0	9.4	3.1	25.3	425.9	35.6	471.6
20.5	6.8	25.3	693.7	35.4	778.0	9.0	3.0	25.3	412.8	35.7	457.0
19.5	6.5	25.4	681.7	35.5	762.7	8.7	2.9	25.3	396.9	35.6	442.6
18.6	6.2	25.4	669.0	35.4	746.7	8.4	2.8	25.2	387.4	35.5	429.1
17.8	5.9	25.3	655.4	35.4	730.0	8.1	2.7	25.3	376.9	35.4	416.6
17.1	5.7	25.3	642.2	35.6	714.3						

(κ, $100w_{HF}$) data from the literature (Pick, 1912; Fredenhagen and Cadenbach, 1930; Broderick, 1962) were fitted to eq 3. Data at different temperatures were simultaneously considered since the temperature coefficient of HF aqueous solutions in the $0 < 100w_{HF} < 50$ range is not statistically different from zero (Cairns, 1967). The best value encountered for the b parameter is $b = (12.788 \pm 0.071)$ mS·cm^{-1}. The b parameter was calculated in this way since the conductivity of the $H_2SiF_6 + HF + H_2O$ system was not experimentally measured in the domain of $100w_1 = 0$. This fact did not allow a precise calculation of the b parameter by direct fit of experimental data to eq 2.

Conductimetric data appearing in Table 2 at each temperature in the study were fitted to eq 2 by means of NLREG. In all the calculations the b parameter was fixed constant at 12.788. The contribution due to the interaction between the lineal term in the HF mass percent and the

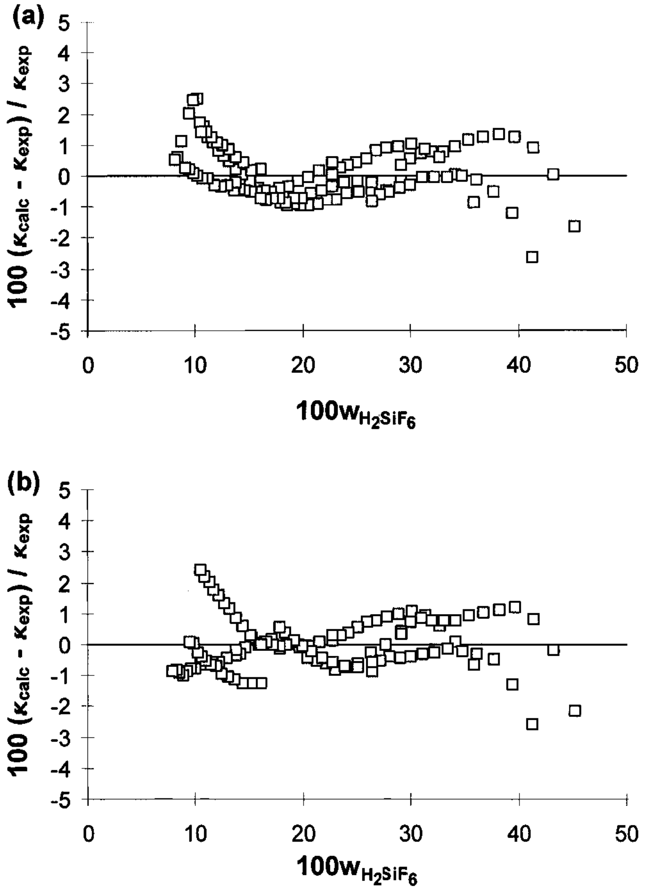

Figure 1. Relative errors ($100(\kappa_{calc} - \kappa_{exp})/\kappa_{exp}$) between experimental data and data calculated by eq 4 using the best set of adjustable parameters (see Table 3) at (a) (25.3 ± 0.2) °C and (b) (35.4 ± 0.1) °C.

Table 3. Calculated Values for the Parameters of Eq 4 Which Best Fit Experimental Data at the Two Temperatures Studied

	t = (25.3 ± 0.2) °C	t = (35.4 ± 0.1) °C
a/mS·cm^{-1}	50.887 ± 0.068	57.208 ± 0.068
b/mS·cm^{-1}	12.788 ± 0.071	12.788 ± 0.071
c/mS·cm^{-1}	−0.5468 ± 0.0059	−0.5644 ± 0.0064
d/mS·cm^{-1}	−0.8559 ± 0.0021	−0.9589 ± 0.0023

second-order term in the H_2SiF_6 mass percent was found not to be statistically significant to explain the randomness of the data ($e = 0$). The values for the adjustable parameters of the finally selected model (eq 4) which best fit experimental data at each temperature studied are collected in Table 3.

$$\kappa_t = a100w_1 + b100w_2 + c100w_1100w_2 + d(100w_1)^2 \quad (4)$$

Relative errors ($100(\kappa_{calc} - \kappa_{exp})/\kappa_{exp}$) between calculated and experimental data are plotted in Figure 1.

Discussion

The values for the a and d parameters given in this work agree with those given for the coefficients of the second-order polynomial proposed to describe the conductivity of the binary system $H_2SiF_6 + H_2O$ (de Diego et al., 1996a).

The proposed model has been used to estimate the specific conductivities of the ternary system H_2SiF_6 + HF + H_2O at the two temperatures studied in the $0 < 100w_{H_2SiF_6} < 50$ range at round HF mass percents. Figure 2 illustrates the result of this calculation. It can be observed that, in the analyzed composition range, the maximum conductivity of the curves corresponding to each HF mass percent shifts toward lower H_2SiF_6 mass percents and lower values of conductivity with increasing HF content. Again, increasing HF mass percent results in a higher conductivity in the $0 < 100w_{H_2SiF_6} < 23$ domain and the opposite behavior is observed for compositions with higher H_2SiF_6 content.

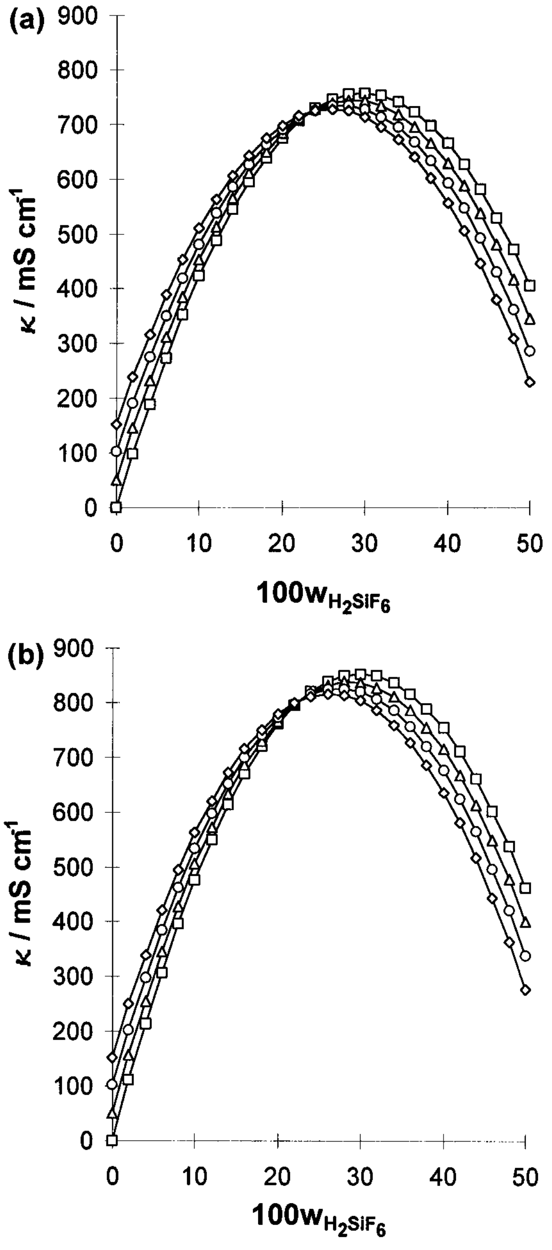

Figure 2. Conductivities of the H_2SiF_6 (1) + HF (2) + H_2O (3) ternary system estimated by the proposed model at round HF mass percents at (a) (25.3 ± 0.2) °C and (b) (35.4 ± 0.1) °C): (□) $100w_2 = 0$; (△) $100w_2 = 4$; (○) $100w_2 = 8$; (◇) $100w_2 = 12$.

The proposed model allows the interpolation of conductivity in the $25\ °C < t < 35\ °C$ temperature range, after estimation of the corresponding coefficients of eq 4 by considering the linear relation between the values of the coefficients and the temperature. Previous work (de Diego, 1996; de Diego et al., 1996a,b) provides a good base to believe that, in fact, a linear relationship does exit.

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