Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.


# Supporting Information

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## Dielectric Fitting Data

**General:** The obtained THz dielectric spectra of a series of binary liquid mixtures were fitted to either a two-component Debye model (D2), a constrained three-component Debye model (D3), or a Debye model containing a single Lorentz vibrational term (D1-V1).

$$\tilde{\varepsilon}_{Debye} = \varepsilon_\infty + \sum_{j=1}^{2} \frac{\varepsilon_j}{1 + i\omega\tau_j} \qquad (D2)$$

$$\tilde{\varepsilon}_{Debye} = \varepsilon_\infty + \sum_{j=1}^{3} \frac{\varepsilon_j}{1 + i\omega\tau_j} \qquad (D3)$$

$$\tilde{\varepsilon}_{Debye} = \varepsilon_\infty + \frac{\varepsilon_1}{1 + i\omega\tau_1} + \frac{A_v}{\omega_v^2 - \omega^2 + i\omega\gamma_v} \qquad (D1-V1)$$

The values $\varepsilon_j$ and $\tau_j$ represent the relative relaxation strength and time of the $j^{th}$ relaxation component, $A_v$ is the amplitude of the vibrational mode, $\omega_v$ is the characteristic frequency of the vibrational mode and $\gamma_v$ is the damping factor. For the D3 model, the values of $\tau_1$ and $\tau_2$ are fixed to that of the pure liquid components obtained from the D2 model.

Static dielectric constants ($\varepsilon_s$) of the mixtures are obtained from previously published results and fixed as

$$\varepsilon_s = \varepsilon_\infty + \sum_j \varepsilon_j$$

The measured THz dielectric spectra were fitted to these models by minimising both weighted least-squares, $S_w$

$$S_w = \sum_\omega \left[\frac{R(\tilde{\varepsilon}_{Debye}) - R(\tilde{\varepsilon}_{THz})}{R(\tilde{\varepsilon}_{THz})}\right]^2 + \sum_\omega \left[\frac{I(\tilde{\varepsilon}_{Debye}) - I(\tilde{\varepsilon}_{THz})}{I(\tilde{\varepsilon}_{THz})}\right]^2$$

where $\tilde{\varepsilon}_{THz}$ is the measured THz dielectric spectrum. Normalised residues were calculated for every fit as

$$\delta_w^2 = \frac{S^{min}}{2m}$$

**Diol-water and diol-methanol mixtures:** The obtained THz dielectric spectra of these mixtures were fitted to both the D2 and the constrained D3 models.

Dielectric relaxation parameters of diol-methanol mixtures evaluated using the D2 model. Values of the static dielectric constant ($\varepsilon_s$) were obtained from Ref. 1.

| *mol%* | $\varepsilon_s$ | $\varepsilon_1$ | $\tau_1$ (ps) | $\varepsilon_2$ | $\tau_2$ (fs) | $\delta_w^2$ ($10^{-4}$) |
|---|---|---|---|---|---|---|
| Ethylene glycol-methanol | | | | | | |
| 0 | 32.88 | 29.3 | 13.5 | 1.47 | 158 | 2.68 |
| 10 | 33.55 | 30.1 | 12.0 | 1.40 | 142 | 4.67 |
| 20 | 34.59 | 31.1 | 14.1 | 1.44 | 126 | 6.19 |
| 30 | 35.47 | 31.9 | 16.5 | 1.49 | 111 | 3.70 |
| 40 | 36.34 | 32.7 | 17.9 | 1.47 | 125 | 4.34 |
| 50 | 37.14 | 32.7 | 16.5 | 1.58 | 115 | 4.74 |
| 60 | 37.84 | 33.4 | 16.2 | 1.56 | 113 | 4.95 |
| 70 | 38.48 | 34.8 | 19.7 | 1.47 | 108 | 4.00 |
| 80 | 39.02 | 35.2 | 20.3 | 1.50 | 111 | 3.04 |
| 90 | 39.78 | 35.7 | 24.9 | 1.60 | 112 | 3.08 |
| 100 | 40.68 | 36.9 | 33.6 | 1.55 | 104 | 2.00 |
| 1,2-propanediol-methanol | | | | | | |
| 0 | 32.88 | 29.4 | 14.5 | 1.48 | 180 | 5.88 |
| 10 | 31.97 | 28.5 | 16.6 | 1.39 | 152 | 7.16 |
| 20 | 31.53 | 28.2 | 20.8 | 1.35 | 140 | 8.60 |
| 30 | 31.07 | 27.8 | 21.9 | 1.20 | 117 | 3.87 |
| 50 | 30.15 | 26.9 | 24.2 | 1.12 | 104 | 1.63 |
| 60 | 29.74 | 26.4 | 24.9 | 1.13 | 103 | 1.92 |
| 70 | 29.38 | 26.1 | 34.9 | 1.12 | 104 | 2.07 |
| 100 | 28.30 | 25.1 | 40.8 | 1.01 | 96.4 | 2.32 |

Dielectric relaxation parameters of diol-methanol mixtures evaluated using the D3 model. Values of the static dielectric constant ($\varepsilon_s$) were obtained from Ref. 1.

| *mol%* | $\varepsilon_s$ | $\varepsilon_1$ | $\varepsilon_2$ | $\varepsilon_3$ | $\tau_3$ (fs) | $\delta_w^2$ ($10^{-4}$) |
|---|---|---|---|---|---|---|
| Ethylene glycol-methanol | | | | | | |
| 0 | 32.88 | 29.3 | 0.06 | 1.47 | 158 | 2.68 |
| 10 | 33.55 | 23.7 | 6.35 | 1.40 | 142 | 4.60 |
| 20 | 34.59 | 19.0 | 12.1 | 1.43 | 126 | 6.11 |
| 30 | 35.47 | 14.6 | 17.4 | 1.49 | 111 | 3.68 |
| 40 | 36.34 | 12.7 | 20.1 | 1.47 | 125 | 4.31 |
| 50 | 37.14 | 14.8 | 17.9 | 1.58 | 114 | 4.73 |
| 60 | 37.84 | 15.8 | 17.5 | 1.56 | 113 | 4.94 |
| 70 | 38.48 | 10.8 | 24.1 | 1.46 | 108 | 3.98 |
| 80 | 39.02 | 10.2 | 25.0 | 1.50 | 110 | 3.05 |
| 90 | 39.78 | 5.53 | 30.2 | 1.60 | 112 | 3.08 |
| 100 | 40.68 | 0.00 | 36.9 | 1.55 | 104 | 2.00 |
| 1,2-propanediol-methanol | | | | | | |
| 0 | 32.88 | 29.9 | 0.00 | 1.49 | 168 | 5.99 |
| 10 | 31.97 | 25.2 | 3.31 | 1.39 | 152 | 7.14 |
| 20 | 31.53 | 15.8 | 12.4 | 1.34 | 140 | 8.56 |
| 30 | 31.07 | 13.7 | 14.1 | 1.20 | 117 | 3.86 |
| 50 | 30.15 | 10.1 | 16.8 | 1.12 | 104 | 1.62 |
| 60 | 29.74 | 9.03 | 17.4 | 1.13 | 103 | 1.91 |

| | | | | | | |
|---|---|---|---|---|---|---|
| 70 | 29.38 | 3.12 | 22.9 | 1.12 | 104 | 2.07 |
| 100 | 28.30 | 0.00 | 25.1 | 1.03 | 99 | 2.21 |

Dielectric relaxation parameters of diol-water mixtures evaluated using the D2 model. Values of the static dielectric constant ($\varepsilon_s$) were obtained from Ref. 2 and 3.

| *mol%* | $\varepsilon_s$ | $\varepsilon_1$ | $\tau_1$ (ps) | $\varepsilon_2$ | $\tau_2$ (fs) | $\delta_w^2$ ($10^{-4}$) |
|---|---|---|---|---|---|---|
| Ethylene glycol-water | | | | | | |
| 0 | 80.40 | 75.3 | 7.45 | 2.02 | 102 | 4.29 |
| 10 | 70.62 | 65.7 | 11.3 | 2.01 | 122 | 4.41 |
| 20 | 64.97 | 60.2 | 14.8 | 1.88 | 129 | 3.41 |
| 30 | 60.46 | 56.1 | 21.0 | 1.83 | 112 | 6.54 |
| 40 | 56.60 | 52.3 | 25.3 | 1.81 | 110 | 8.73 |
| 50 | 53.18 | 49.0 | 23.9 | 1.73 | 100 | 2.33 |
| 60 | 50.09 | 46.0 | 27.7 | 1.69 | 96 | 2.80 |
| 70 | 47.29 | 43.3 | 31.0 | 1.60 | 106 | 4.47 |
| 80 | 44.80 | 40.9 | 31.2 | 1.56 | 100 | 4.59 |
| 90 | 42.62 | 38.8 | 38.6 | 1.59 | 101 | 13.7 |
| 100 | 40.82 | 36.9 | 35.2 | 1.58 | 101 | 4.32 |
| 1,2-propanediol-water | | | | | | |
| 0 | 80.40 | 75.3 | 7.45 | 2.02 | 102 | 4.29 |
| 10 | 64.81 | 59.9 | 12.5 | 1.93 | 110 | 7.80 |
| 20 | 56.44 | 52.0 | 18.5 | 1.65 | 105 | 4.92 |
| 30 | 50.42 | 46.4 | 25.7 | 1.53 | 94 | 5.73 |
| 40 | 45.74 | 42.0 | 21.7 | 1.33 | 78 | 2.42 |
| 50 | 41.91 | 38.1 | 30.6 | 1.32 | 93 | 1.92 |
| 60 | 38.35 | 34.8 | 23.2 | 1.16 | 90 | 6.20 |
| 70 | 35.36 | 32.0 | 24.9 | 1.08 | 85 | 1.03 |
| 80 | 32.67 | 29.4 | 24.4 | 1.06 | 80 | 1.01 |
| 90 | 30.34 | 27.1 | 38.1 | 1.01 | 101 | 0.82 |
| 100 | 28.36 | 25.1 | 29.7 | 0.98 | 89 | 1.86 |
| 1,3-propanediol-water | | | | | | |
| 0 | 80.40 | 75.3 | 7.45 | 2.02 | 102 | 4.29 |
| 10 | 66.21 | 61.5 | 14.2 | 1.78 | 116 | 5.36 |
| 20 | 58.49 | 54.2 | 19.8 | 1.48 | 105 | 7.78 |
| 30 | 53.07 | 49.1 | 33.1 | 1.35 | 115 | 13.7 |
| 40 | 48.97 | 45.1 | 40.1 | 1.26 | 105 | 9.07 |
| 50 | 45.67 | 42.1 | 38.7 | 1.10 | 95 | 5.44 |
| 60 | 42.84 | 39.4 | 41.2 | 1.03 | 96 | 8.32 |
| 70 | 40.28 | 37.1 | 37.0 | 0.96 | 90 | 13.9 |
| 80 | 37.96 | 34.6 | 44.7 | 0.95 | 98 | 11.4 |
| 90 | 35.91 | 32.3 | 59.9 | 0.94 | 103 | 8.29 |
| 100 | 34.30 | 31.1 | 68.3 | 0.90 | 105 | 8.72 |
| 1,2-butanediol-water | | | | | | |
| 0 | 80.40 | 75.3 | 7.45 | 2.02 | 102 | 4.29 |
| 10 | 60.57 | 55.7 | 15.1 | 1.85 | 122 | 13.5 |
| 20 | 50.40 | 45.8 | 20.4 | 1.65 | 119 | 13.0 |
| 30 | 43.65 | 39.9 | 23.3 | 1.13 | 109 | 16.3 |
| 40 | 38.72 | 35.2 | 27.9 | 1.06 | 102 | 4.71 |
| 50 | 34.88 | 31.5 | 39.5 | 0.98 | 109 | 5.63 |
| 60 | 31.73 | 28.5 | 37.7 | 0.89 | 108 | 1.78 |

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|---|---|---|---|---|---|---|
| 70 | 28.99 | 25.8 | 44.1 | 0.85 | 98 | 2.32 |
| 80 | 26.53 | 23/4 | 42.5 | 0.78 | 105 | 4.15 |
| 90 | 24.33 | 21.3 | 68.9 | 0.79 | 118 | 3.43 |
| 100 | 22.57 | 19.5 | 61.4 | 0.76 | 112 | 3.83 |
| 1,4-butanediol-water | | | | | | |
| 0 | 80.40 | 75.3 | 7.45 | 2.02 | 102 | 4.29 |
| 10 | 63.71 | 59.3 | 15.8 | 1.59 | 107 | 3.87 |
| 20 | 55.26 | 50.9 | 25.4 | 1.48 | 155 | 8.79 |
| 30 | 49.61 | 45.7 | 34.4 | 1.19 | 118 | 5.76 |
| 40 | 45.48 | 41.8 | 38.7 | 1.16 | 77 | 3.98 |
| 50 | 42.25 | 38.7 | 53.8 | 0.96 | 113 | 9.88 |
| 60 | 39.59 | 36.2 | 59.2 | 0.92 | 95 | 4.22 |
| 70 | 37.27 | 34.1 | 80.3 | 0.80 | 111 | 3.88 |
| 80 | 35.18 | 32.1 | 59.1 | 0.76 | 107 | 6.43 |
| 90 | 33.32 | 30.2 | 68.7 | 0.73 | 111 | 6.99 |
| 100 | 31.82 | 28.8 | 96.7 | 0.78 | 68 | 9.23 |

Dielectric relaxation parameters of diol-water mixtures evaluated using the constrained D3 model. Values of the static dielectric constant ($\varepsilon_s$) were obtained from Ref. 2 and 3.

| *mol%* | $\varepsilon_s$ | $\varepsilon_1$ | $\varepsilon_2$ | $\varepsilon_3$ | $\tau_3$ (fs) | $\delta_w^2$ ($10^{-4}$) |
|---|---|---|---|---|---|---|
| Ethylene glycol-water | | | | | | |
| 0 | 80.40 | 75.3 | 0.00 | 2.02 | 102 | 5.83 |
| 10 | 70.62 | 38.1 | 27.7 | 1.99 | 119 | 4.34 |
| 20 | 64.97 | 22.5 | 37.8 | 1.87 | 127 | 3.64 |
| 30 | 60.46 | 10.1 | 46.0 | 1.82 | 111 | 6.35 |
| 40 | 56.60 | 5.29 | 47.0 | 1.81 | 109 | 8.62 |
| 50 | 53.18 | 6.05 | 42.9 | 1.73 | 99 | 2.38 |
| 60 | 50.09 | 3.14 | 42.9 | 1.69 | 96 | 2.81 |
| 70 | 47.29 | 1.35 | 42.0 | 1.60 | 106 | 4.45 |
| 80 | 44.80 | 1.17 | 39.7 | 1.56 | 100 | 4.58 |
| 90 | 42.62 | 0.00 | 38.8 | 1.58 | 94 | 17.0 |
| 100 | 40.82 | 0.00 | 37.0 | 1.58 | 101 | 6.34 |
| 1,2-propanediol-water | | | | | | |
| 0 | 80.40 | 75.3 | 0.00 | 2.02 | 102 | 5.83 |
| 10 | 64.81 | 30.1 | 29.8 | 1.92 | 108 | 7.41 |
| 20 | 56.44 | 13.3 | 38.7 | 1.65 | 104 | 4.83 |
| 30 | 50.42 | 5.10 | 41.3 | 1.53 | 94 | 5.64 |
| 40 | 45.74 | 7.44 | 34.6 | 1.33 | 78 | 2.41 |
| 50 | 41.91 | 1.88 | 36.3 | 1.32 | 93 | 1.93 |
| 60 | 38.35 | 5.17 | 29.6 | 1.16 | 90 | 0.62 |
| 70 | 35.36 | 3.84 | 28.1 | 1.08 | 85 | 1.03 |
| 80 | 32.67 | 3.78 | 25.6 | 1.06 | 80 | 1.01 |
| 90 | 30.34 | 0.00 | 27.1 | 1.00 | 99 | 0.94 |
| 100 | 28.36 | 0.00 | 25.1 | 0.98 | 90 | 2.12 |
| 1,3-propanediol-water | | | | | | |
| 0 | 80.40 | 75.3 | 0.00 | 2.02 | 102 | 5.83 |
| 10 | 66.21 | 29.3 | 32.3 | 1.77 | 114 | 4.90 |
| 20 | 58.49 | 16.6 | 37.6 | 1.47 | 103 | 7.78 |
| 30 | 53.07 | 6.56 | 42.5 | 1.35 | 114 | 13.4 |
| 40 | 48.97 | 3.96 | 41.1 | 1.26 | 105 | 9.01 |

| | | | | | | |
|---|---|---|---|---|---|---|
| 50 | 45.67 | 3.99 | 38.1 | 1.10 | 94 | 5.45 |
| 60 | 42.84 | 3.23 | 36.2 | 1.03 | 96 | 8.29 |
| 70 | 40.28 | 3.90 | 33.2 | 0.96 | 90 | 13.9 |
| 80 | 37.96 | 2.27 | 32.3 | 0.95 | 98 | 11.4 |
| 90 | 35.91 | 0.57 | 32.0 | 0.94 | 103 | 8.29 |
| 100 | 34.30 | 0.00 | 31.1 | 0.90 | 105 | 8.71 |
| 1,2-butanediol-water | | | | | | |
| 0 | 80.40 | 75.3 | 0.00 | 2.02 | 102 | 5.83 |
| 10 | 60.57 | 24.2 | 31.5 | 1.83 | 119 | 13.3 |
| 20 | 50.40 | 13.0 | 32.8 | 1.64 | 117 | 12.5 |
| 30 | 43.65 | 9.19 | 30.8 | 1.12 | 108 | 16.0 |
| 40 | 38.72 | 5.92 | 29.2 | 1.06 | 102 | 4.70 |
| 50 | 34.88 | 2.44 | 29.0 | 0.98 | 109 | 5.64 |
| 60 | 31.73 | 2.50 | 26.0 | 0.89 | 108 | 1.78 |
| 70 | 28.99 | 1.42 | 24.4 | 0.85 | 98 | 2.32 |
| 80 | 26.53 | 1.46 | 22.0 | 0.78 | 105 | 4.14 |
| 90 | 24.33 | 0.00 | 21.3 | 0.78 | 115 | 3.72 |
| 100 | 22.57 | 0.00 | 19.5 | 0.76 | 112 | 3.64 |
| 1,4-butanediol-water | | | | | | |
| 0 | 80.40 | 75.3 | 0.00 | 2.02 | 102 | 5.83 |
| 10 | 63.71 | 25.1 | 34.3 | 1.59 | 100 | 3.32 |
| 20 | 55.26 | 12.0 | 38.9 | 1.45 | 148 | 9.74 |
| 30 | 49.61 | 6.98 | 38.8 | 1.18 | 113 | 4.38 |
| 40 | 45.48 | 5.20 | 36.7 | 1.17 | 74 | 3.55 |
| 50 | 42.25 | 2.61 | 36.1 | 0.96 | 111 | 8.72 |
| 60 | 39.59 | 1.95 | 34.2 | 0.92 | 94 | 4.22 |
| 70 | 37.27 | 0.61 | 33.4 | 0.80 | 110 | 3.98 |
| 80 | 35.18 | 1.74 | 30.4 | 0.75 | 105 | 6.41 |
| 90 | 33.32 | 1.04 | 29.2 | 0.73 | 109 | 6.49 |
| 100 | 31.82 | 0.00 | 28.7 | 0.78 | 68 | 8.74 |

**Aprotic liquid-water mixtures:** The measured THz dielectric spectra were fitted to the D2, D1-V1 and constrained D3 models.

Dielectric relaxation parameters of aprotic liquid-water mixtures evaluated using the D2 model. Values of the static dielectric constant ($\varepsilon_s$) were obtained from Ref. 4, 5 and 6.

| *mol%* | $\varepsilon_s$ | $\varepsilon_1$ | $\tau_1$ (ps) | $\varepsilon_2$ | $\tau_2$ (fs) | $\delta_w^2$ ($10^{-4}$) |
|---|---|---|---|---|---|---|
| Acetonitrile-water | | | | | | |
| 0 | 80.40 | 75.3 | 7.45 | 2.02 | 102 | 4.29 |
| 10 | 72.35 | 66.9 | 6.64 | 2.47 | 112 | 5.61 |
| 20 | 63.90 | 58.4 | 6.74 | 2.85 | 132 | 4.88 |
| 30 | 57.04 | 52.0 | 5.83 | 2.73 | 137 | 9.03 |
| 40 | 51.97 | 47.1 | 4.95 | 2.71 | 151 | 8.72 |
| 50 | 48.11 | 43.6 | 4.94 | 2.76 | 144 | 9.47 |
| 60 | 44.93 | 40.7 | 4.33 | 2.66 | 146 | 10.4 |
| 70 | 42.27 | 38.3 | 3.61 | 2.55 | 135 | 19.1 |
| 80 | 40.22 | 36.3 | 3.15 | 2.44 | 150 | 21.0 |
| 90 | 38.46 | 35.3 | 2.63 | 1.89 | 108 | 15.3 |
| 100 | 36.74 | 33.5 | 3.03 | 2.41 | 141 | 14.7 |
| Acetone-water | | | | | | |

| | | | | | | |
|---|---|---|---|---|---|---|
| 0 | 80.40 | 75.3 | 7.45 | 2.02 | 102 | 4.29 |
| 10 | 61.89 | 57.4 | 10.4 | 1.81 | 113 | 5.80 |
| 20 | 51.36 | 47.2 | 10.6 | 1.79 | 119 | 6.04 |
| 30 | 42.87 | 39.0 | 9.19 | 1.67 | 148 | 3.52 |
| 40 | 37.65 | 34.0 | 8.45 | 1.65 | 152 | 5.22 |
| 50 | 30.85 | 27.4 | 5.96 | 1.58 | 166 | 3.08 |
| 60 | 27.75 | 24.4 | 4.83 | 1.51 | 189 | 2.93 |
| 70 | 25.57 | 22.4 | 4.80 | 1.47 | 185 | 3.57 |
| 80 | 23.64 | 20.4 | 3.85 | 1.53 | 254 | 3.51 |
| 90 | 22.00 | 19.2 | 3.26 | 1.24 | 216 | 2.27 |
| 100 | 20.80 | 18.1 | 3.01 | 1.15 | 222 | 3.19 |
| Tetrahydrofuran-water | | | | | | |
| 0 | 80.40 | 75.3 | 7.45 | 2.02 | 102 | 4.29 |
| 10 | 57.76 | 53.1 | 12.0 | 1.72 | 115 | 7.96 |
| 20 | 41.20 | 37.2 | 10.1 | 1.34 | 115 | 6.78 |
| 30 | 30.57 | 26.8 | 10.0 | 1.23 | 137 | 7.97 |
| 40 | 23.55 | 20.1 | 8.26 | 1.07 | 152 | 6.44 |
| 50 | 18.75 | 15.5 | 5.94 | 0.95 | 159 | 5.54 |
| 60 | 15.32 | 12.6 | 5.17 | 0.71 | 149 | 1.94 |
| 70 | 12.72 | 9.75 | 3.31 | 0.77 | 158 | 3.01 |
| 80 | 10.60 | 7.75 | 3.41 | 0.81 | 236 | 1.92 |
| 90 | 8.82 | 6.31 | 2.23 | 0.54 | 204 | 2.60 |
| 100 | 7.58 | 5.22 | 2.26 | 0.47 | 231 | 6.18 |

Dielectric relaxation parameters of aprotic liquid-water mixtures evaluated using the D1-V1 model.

| *mol%* | $\varepsilon_s$ | $\varepsilon_1$ | $\tau_1$ (ps) | $\frac{A_v}{4\pi^2}$ (THz$^2$) | $\frac{\gamma_v}{2\pi}$ (THz) | $\delta_w^2$ ($10^{-4}$) |
|---|---|---|---|---|---|---|
| Acetonitrile-water ($\omega_v$ = 2.79 THz) | | | | | | |
| 0 | 80.40 | 75.3 | 7.42 | 4.48 | 2.70 | 15.2 |
| 10 | 72.35 | 66.9 | 6.54 | 15.4 | 6.12 | 8.10 |
| 20 | 63.90 | 58.4 | 6.70 | 19.0 | 7.25 | 9.42 |
| 30 | 57.04 | 52.1 | 5.78 | 18.3 | 7.43 | 11.0 |
| 40 | 51.97 | 47.1 | 4.93 | 18.6 | 8.17 | 9.63 |
| 50 | 48.11 | 43.6 | 4.38 | 17.4 | 6.57 | 17.9 |
| 60 | 44.93 | 40.8 | 4.29 | 18.0 | 7.78 | 11.6 |
| 70 | 42.27 | 38.4 | 3.58 | 16.9 | 7.34 | 21.8 |
| 80 | 40.22 | 36.3 | 3.13 | 16.6 | 8.09 | 23.1 |
| 90 | 38.46 | 35.4 | 2.60 | 11.5 | 6.02 | 17.2 |
| 100 | 36.74 | 33.6 | 2.97 | 15.7 | 7.34 | 14.1 |
| Acetone-water ($\omega_v$ = 1.83 THz) | | | | | | |
| 0 | 80.40 | 75.3 | 7.42 | 4.48 | 2.70 | 15.2 |
| 10 | 61.89 | 57.0 | 9.77 | 3.91 | 2.65 | 28.3 |
| 20 | 51.36 | 46.9 | 10.0 | 4.01 | 2.80 | 26.2 |
| 30 | 42.87 | 38.5 | 8.56 | 4.05 | 3.26 | 15.9 |
| 40 | 37.65 | 33.0 | 7.79 | 4.06 | 3.37 | 15.2 |
| 50 | 30.85 | 27.0 | 5.60 | 3.95 | 3.58 | 10.0 |
| 60 | 27.75 | 24.5 | 4.65 | 3.92 | 4.00 | 7.26 |
| 70 | 25.57 | 22.2 | 4.54 | 3.73 | 3.88 | 5.86 |
| 80 | 23.64 | 20.7 | 3.72 | 4.05 | 4.98 | 2.25 |
| 90 | 22.00 | 19.5 | 3.28 | 3.51 | 4.86 | 2.49 |

| | | | | | | |
|---|---|---|---|---|---|---|
| 100 | 20.80 | 18.2 | 2.97 | 3.19 | 4.81 | 3.18 |
| Tetrahydrofuran-water ($\omega_v$ = 1.78 THz) | | | | | | |
| 0 | 80.40 | 75.3 | 7.42 | 4.48 | 2.70 | 15.2 |
| 10 | 57.76 | 53.2 | 11.5 | 3.59 | 2.62 | 31.2 |
| 20 | 41.20 | 37.2 | 9.61 | 2.78 | 2.57 | 32.1 |
| 30 | 30.57 | 26.8 | 9.59 | 2.78 | 2.98 | 25.9 |
| 40 | 23.55 | 20.2 | 7.80 | 2.45 | 3.14 | 21.1 |
| 50 | 18.75 | 15.6 | 5.60 | 2.16 | 3.19 | 16.5 |
| 60 | 15.32 | 12.6 | 4.89 | 1.55 | 2.93 | 8.58 |
| 70 | 12.72 | 9.83 | 3.15 | 1.69 | 3.06 | 8.58 |
| 80 | 10.60 | 7.94 | 3.01 | 1.68 | 3.96 | 3.22 |
| 90 | 8.82 | 6.43 | 2.10 | 1.10 | 3.45 | 1.50 |
| 100 | 7.58 | 5.31 | 2.15 | 1.02 | 3.96 | 3.48 |

Dielectric relaxation parameters of aprotic liquid-water mixtures evaluated using the constrained D3 model. Values of the static dielectric constant ($\varepsilon_s$) were obtained from Ref. 4, 5 and 6.

| *mol%* | $\varepsilon_s$ | $\varepsilon_1$ | $\varepsilon_2$ | $\varepsilon_3$ | $\tau_3$ (fs) | $\delta_w^2$ ($10^{-4}$) |
|---|---|---|---|---|---|---|
| Acetonitrile-water | | | | | | |
| 10 | 72.35 | 61.3 | 5.71 | 2.44 | 105 | 5.52 |
| 20 | 63.90 | 53.8 | 4.69 | 2.81 | 119 | 9.13 |
| 30 | 57.04 | 42.9 | 9.25 | 2.67 | 130 | 10.2 |
| 40 | 51.97 | 33.4 | 13.8 | 2.66 | 143 | 10.2 |
| 50 | 48.11 | 29.5 | 14.3 | 2.66 | 129 | 7.56 |
| 60 | 44.93 | 22.5 | 18.4 | 2.55 | 133 | 7.71 |
| 70 | 42.27 | 13.8 | 24.7 | 2.47 | 123 | 19.1 |
| 80 | 40.22 | 6.79 | 29.6 | 2.36 | 139 | 20.6 |
| 90 | 38.46 | 0 | 35.1 | 2.02 | 121 | 16.7 |
| Acetone-water | | | | | | |
| 10 | 61.89 | 57.4 | 0.00 | 1.47 | 62 | 70.3 |
| 20 | 51.36 | 47.3 | 0.00 | 1.44 | 69 | 90.9 |
| 30 | 42.87 | 38.8 | 0.00 | 1.39 | 115 | 17.6 |
| 40 | 37.65 | 33.1 | 0.00 | 1.52 | 137 | 8.10 |
| 50 | 30.85 | 21.2 | 5.70 | 1.54 | 161 | 2.47 |
| 60 | 27.75 | 14.5 | 9.96 | 1.45 | 181 | 2.43 |
| 70 | 25.57 | 12.7 | 9.48 | 1.41 | 177 | 2.55 |
| 80 | 23.64 | 6.77 | 13.9 | 1.45 | 245 | 3.48 |
| 90 | 22.00 | 2.29 | 17.2 | 1.21 | 212 | 2.25 |
| Tetrahydrofuran-water | | | | | | |
| 10 | 57.76 | 53.7 | 0.00 | 1.77 | 27 | 341 |
| 20 | 41.20 | 37.6 | 0.00 | 1.14 | 49 | 112 |
| 30 | 30.57 | 27.1 | 0.00 | 0.94 | 73 | 93.4 |
| 40 | 23.55 | 20.3 | 0.00 | 0.89 | 119 | 16.6 |
| 50 | 18.75 | 14.2 | 1.36 | 0.93 | 156 | 5.27 |
| 60 | 15.32 | 10.4 | 2.22 | 0.69 | 144 | 1.53 |
| 70 | 12.72 | 4.35 | 5.44 | 0.74 | 151 | 2.40 |
| 80 | 10.60 | 3.69 | 4.10 | 0.78 | 230 | 1.92 |
| 90 | 8.82 | 0.00 | 6.29 | 0.56 | 210 | 2.63 |