

## Thermodynamic properties of SiO<sub>2(aq)</sub>, HSiO<sub>3</sub><sup>-</sup> and SiO<sub>3</sub><sup>2-</sup>

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Equilibrium constants ( $K$ ) describing the formation reactions of SiO<sub>2(aq)</sub>, HSiO<sub>3</sub><sup>-</sup> and SiO<sub>3</sub><sup>2-</sup> can be used to derive their thermodynamic properties. However, Rimstidt [1] found SiO<sub>2(aq)</sub> had been derived from inaccurate quartz solubility data, Sverjensky *et al.* [2] failed to extrapolate HSiO<sub>3</sub><sup>-</sup> data to zero ionic strength and SiO<sub>3</sub><sup>2-</sup> is routinely ignored because of its restricted dominance to very high pH > 13 solutions.

Using quartz and water as well known 'anchor' points,  $K$  values describing the formation reactions of SiO<sub>2(aq)</sub>, HSiO<sub>3</sub><sup>-</sup> and SiO<sub>3</sub><sup>2-</sup> were revised to derive new thermodynamic properties and revised Helgeson-Kirkham-Flowers equation of state (r-H-K-F EoS) parameters (Table 1).

Property	SiO <sub>2(aq)</sub>	HSiO <sub>3</sub> <sup>-</sup>	SiO <sub>3</sub> <sup>2-</sup>
$\Delta_f G^0$ (cal/mol)	-199531	-242836	-224609
$\Delta_f H^0$ (cal/mol)	-212510	-274200	-264000
$S^0$ (cal/mol/K)	9.972	4.000	-23.000
$a_1 \times 10$ (cal/mol/bar)	1.8942	2.8399	1.6651
$a_2 \times 10^{-2}$ (cal/mol)	10.7244	-0.8470	-3.7157
$a_3$ (calK/mol/bar)	-2.0213	5.9002	7.2105
$a_4 \times 10^{-4}$ (calK/mol)	-3.2223	-2.7440	-2.6254
$c_1$ (cal/mol/K)	26.5268	29.3208	-1.7129
$c_2 \times 10^{-4}$ (calK/mol)	-14.6835	-17.8011	-17.2732
$\omega^0 \times 10^{-5}$ (cal/mol)	0.1952	1.5608	3.5913

**Table 1.** Thermodynamic properties and r-H-K-F EoS parameters for SiO<sub>2(aq)</sub>, HSiO<sub>3</sub><sup>-</sup> and SiO<sub>3</sub><sup>2-</sup>

Values shown in Table 1 can be used in the derivation of the thermodynamic properties of other Si bearing complexes and minerals, and in calculating groundwater compositions relevant to the geological disposal of radioactive wastes.

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[1] Rimstidt (1997) *Geochim Cosmochim Acta* **61**, 2553-2558. [2] Sverjensky *et al.* (1997) *Geochim Cosmochim Acta* **61**, 1359-1412.