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THERMODYNAMIC CHARACTERISATION OF AQUEOUS NICKEL AMMONIA COMPLEXES AT HYDROTHERMAL CONDITIONS

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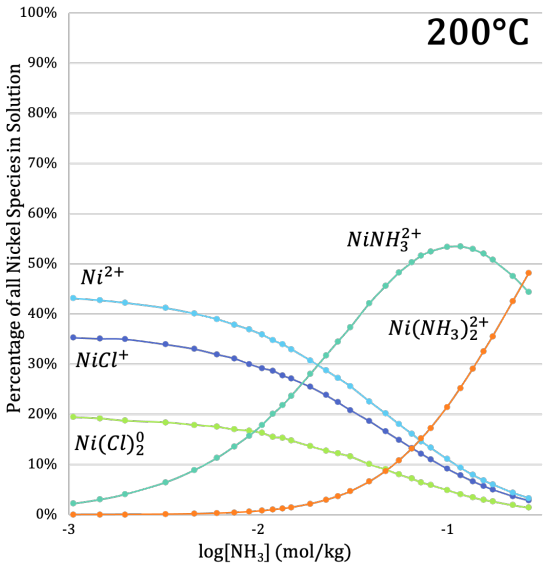
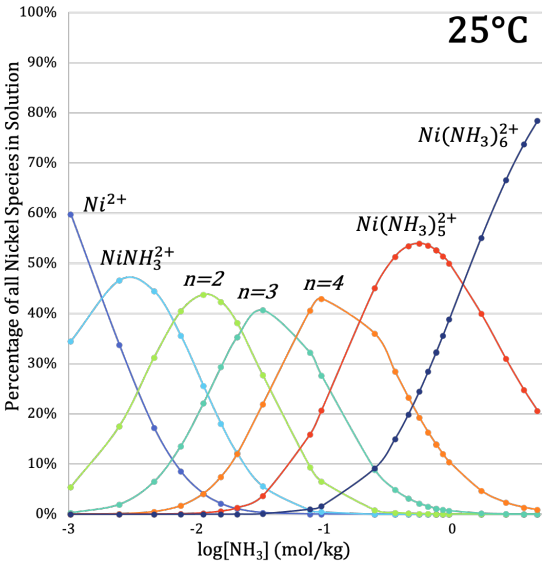
Ammonia forms very stable aqueous complexes with nickel ($Ni(NH_3)_n^{2+}$, n = 1-6) at ambient temperatures, and these can significantly enhance its solubility – a fact that is utilized in hydrometallurgical processing methods for nickel ores.

Ammonia-bearing hydrothermal fluids are known terrestrially ([1] and references therein) and have been inferred on icy planetesimals from the presence of hydrated ammonium-bearing alteration products such as smectites ([2] and references therein). As such is it feasible that such fluids may interact with nickel-bearing minerals under hydrothermal conditions but how important such interactions may be for hydrothermal nickel transport is unclear. Hydrothermal nickel-ammonia interactions are also of interest to nuclear power plant operators. To minimize corrosion in the nickel-alloy secondary coolant/heat transport systems of CANDU and PWR nuclear reactors, additives such as ammonia, morpholine, ethanolamine and cyclohexamine are used for pH and redox chemistry control. Under operating conditions, the latter three thermally decompose to produce ammonia [3].

To be able to accurately model the stability of aqueous nickel-ammonia complexes in these varied environments, accurate thermodynamic properties (e.g. equilibrium constants) are required. However, the thermodynamic equilibrium constants for nickel-ammonia complexes are poorly known at temperatures above 25 ºC making accurate calculations of hydrothermal nickel-ammonia transport difficult.

Here we report equilibrium constants for nickel-ammonia complexes for temperatures up to 200 ºC building upon prior potentiometric results that were limited to 180 ºC and high ionic strength solutions. Measurements were carried out with a UV-visible spectrophotometer equipped with a custom-made titanium-sapphire flow-through cell. We derived equilibrium constants for the $Ni(NH_3)^{2+}$ and $Ni(NH_3)_2^{2+}$ complexes up to 200 ºC, and for the $Ni(NH_3)_3^{2+}$ complex up to 100 ºC. We observed (see below for speciation plots of our solutions), that for a given activity of ammonia, higher temperatures increasingly favoured the lower order complexes especially $Ni(NH_3)^{2+}$ and $Ni(NH_3)_2^{2+}$ over the higher order n=3-6 complexes.





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[2] Tokumon, H., Noji, Y., Fukushi, K., Sekine, Y. (2025) *Geochimica et Cosmochimica Acta* **404**, 188-201.

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