

Part I

PHASE EQUILIBRIA MOLECULAR TRANSPORT THERMODYNAMICS

Ammonium Fluoride-Uranyl Fluoride-Water System

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DATA ON THE binary systems ammonium fluoride-water and uranyl fluoride-water have been reported (1, 2), but none were available on the ternary system of these components. This ternary system was examined briefly.

MATERIALS

The ammonium fluoride used was of analytical reagent grade. High purity uranyl fluoride was prepared by reaction of hydrogen fluoride with reactor grade uranium trioxide. Distilled water was used in preparing the solutions.

EXPERIMENTAL

An excess of solid uranyl fluoride was added to solutions which initially contained ammonium fluoride in concentrations of 0.5 to 6*M*. The solutions were maintained at constant temperature, with periodic shaking, for several days to ensure equilibrium. Then samples of the liquid phase were removed for density measurement and chemical analysis. The solid phase was identified by x-ray diffraction analysis. Temperature was constant to within 0.1° C.

RESULTS

Four isotherms were obtained: 25°, 40°, 59.5°, and 80° C. (Table I and Figure 1). The solid phase at equilibrium was found to be $(\text{NH}_4)_3\text{UO}_2\text{F}_5$ when uranyl fluoride and ammonium fluoride were both present in solution. At points where the data could be compared to the results of other workers—for example, the solubility of ammonium fluoride in water at 25° C.—good agreement was obtained.

Figure 1. Isotherms of ammonium fluoride-uranyl fluoride-water system

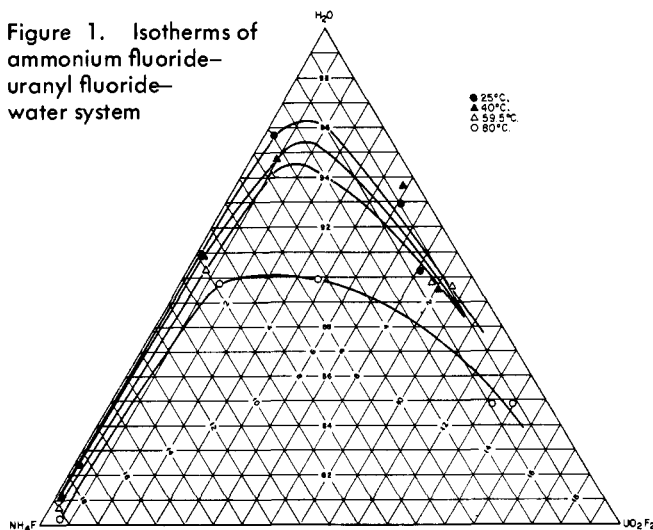


Table I. The System $\text{NH}_4\text{F}-\text{UO}_2\text{F}_2-\text{H}_2\text{O}$ at 25°, 40°, 59.5°, and 80° C.

Temp., ° C.	Solution		Density, g./ml.	Solid Phase in Equilibrium ^a
	% NH_4F	% UO_2F_2		
25	0	64.9	2.292	...
	0	65.6 (1)	2.224 (1)	$\text{UO}_2\text{F}_2 \cdot \text{H}_2\text{O}$ (1)
	0	67.3 (1)	2.405 (1)	...
	0.964	6.16	1.076	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	1.63	8.13	1.089	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	3.92	0.393	1.031	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	8.88	0.168	1.039	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	17.5	0.068	1.084	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	45.8	0	1.119	...
	45.9 (2)	0	...	NH_4F (2)
40	0	68.4 (1)
	0.713	5.81	1.061	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	1.46	9.06	1.083	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	4.30	0.945	1.016	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	8.86	0.292	1.028	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	18.8	0.143	1.068	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	49.2 (2)	0	...	NH_4F (2)

59.5	0	71.0 (1)
	0.946	9.44	1.088	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	1.60	8.65	1.102	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	4.81	1.12	1.016	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	9.12	0.640	1.023	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	19.0	0.264	1.059	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	52.6 (2)	0	...	NH_4F (2)

80	0	72.8 (1)
	1.22	13.8	1.123	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	1.83	13.2	1.136	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	5.35	4.72	1.038	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	8.81	1.43	1.026	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	19.3	0.55	1.056	$(\text{NH}_4)_3\text{UO}_2\text{F}_5$
	54.0 (2)	0	...	NH_4F (2)

^a Solid phase identified by x-ray analysis.

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