IDO-14602
January 1963

PHYSICAL PROPERTIES OF NITRATE SOLUTIONS OF IRON- AND NICKEL-BASE ALLOY NUCLEAR FUELS

D. P. Pearson

PHILLIPS
PETROLEUM
COMPANY

ATOMIC ENERGY DIVISION

NATIONAL REACTOR TESTING STATION
US ATOMIC ENERGY COMMISSION

DISCLAIMER

PRICE $ .50

Available from the
Office of Technical Services
U. S. Department of Commerce
Washington 25, D. C.

Printed in USA

IDO-14602
AEC Research and Development Report
Chemical Separations Processes
for Plutonium and Uranium
TID-4500 (18th Ed.)
Issued: January 31, 1963

PHYSICAL PROPERTIES OF NITRATE SOLUTIONS OF IRON- AND NICKEL-BASE ALLOY NUCLEAR FUELS

D. P. Pearson

PHILLIPS
PETROLEUM
COMPANY

Atomic Energy Division
Contract AT(10-1)-205
Idaho Operations Office
U. S. ATOMIC ENERGY COMMISSION

THIS PAGE

WAS INTENTIONALLY

LEFT BLANK

PHYSICAL PROPERTIES OF NITRATE SOLUTIONS OF IRON- AND NICKEL-BASE ALLOY NUCLEAR FUELS

D. P. Pearson

A B S T R A C T

Specific gravity, viscosity, electrical conductivity and stability data are given for the dissolver product solutions resulting from the electrodissolution in nitric acid of nuclear fuels with stainless steel or Nichrome cladding.

PHYSICAL PROPERTIES OF NITRATE SOLUTIONS OF IRON- AND NICKEL-BASE ALLOY NUCLEAR FUELS

D. P. Pearson

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

PHYSICAL PROPERTIES OF NITRATE SOLUTIONS OF IRON- AND NICKEL-BASE ALLOY NUCLEAR FUELS

D. P. Pearson

I. SUMMARY

Physical property data were determined and compiled for the dissolver product solutions which result from the electrodissolution in nitric acid of stainless steel- or Nichrome-clad nuclear fuels. Specific data given are specific gravity, viscosity, electrical conductivity, and stability for solutions simulating the dissolver product from an SM-1 (APPR) fuel element; also the density and electrical conductivity of a solution simulating the dissolver products from 80-20 Nichrome and the stability data for the dissolver solution resulting from the dissolution of HTRE type Nichrome-clad fuel. Equations are given for the relationship of specific gravity to composition in the two nitrate systems.

II. INTRODUCTION

There has been extensive development work completed on the electrodissolution in nitric acid of nuclear fuels clad in iron- or nickel base alloys[1-2]. The most prominent of these at this time are the Nichrome clad fuels from the HTRE program[3] and the stainless steel clad cermet fuels from the Army Package Power Reactor Program[1].

It is the purpose of this report to compile all of the data on solution density, viscosity, electrical conductivity, and solubility of the dissolver solutions of these fuels which have been obtained at the Idaho Chemical Processing Plant. Data have been reported earlier on the electrochemical properties of Nichrome[4] and stainless steel[5] in nitric acid. The formation of precipitates when molybdenum, silicon, or other difficultly soluble elements are present in the alloys has also been studied briefly[1].

These physical property data are required for preparation of the chemical material balances for the process, for calculation of the heat produced during electrodissolution, and for evaluation of the performance of solvent extraction contactors which will use the dissolver product solution as aqueous feed. The conductivity and density data have already been applied to the simultaneous, in-line analysis of the dissolver product solution for nitric acid and metals[6].

III. EXPERIMENTAL

The solutions used in the measurements and for the stability studies were prepared by weighing reagent or analytical reagent grade salts and reagent grade nitric acid and diluting with distilled water. Selected compositions were checked by colorimetric analysis of the metal constituents and titration of the acid. All saturated solutions (whose composition had changed since make-up) were analyzed. Correlation of compositions based on the weights of reagents and analytical data on the samples indicates that the compositions were within 1 per cent of the values stated.

Specific gravities were determined by weighing a known volume of solution and comparing to weight of distilled water. Viscosities were determined with Ostwald viscosimeters calibrated with distilled water. All samples were incubated at and measurements made at the temperatures stated with the data, which usually was 25°C.

Conductivities were measured in the usual manner in conventional glass cells in a 25°C oil bath.

IV. PHYSICAL PROPERTY DATA

A. Solutions of Iron, Chromium, Nickel, and Uranium

The specific gravities, viscosities, and electrical conductivities of solutions of nitric acid and iron, chromium, nickel, and uranium nitrates are given in Table 1. The metal ions are present in proportions which represent the dissolver product from a fuel which contains 92 per cent stainless steel and 8 per cent uranium. The stainless steel was assumed to be 19 per cent chromium, 10 per cent nickel, and 71 per cent iron. The stability diagram for this system at 25°C is shown in Figure 1. In Figure 1, the solutions whose compositions are indicated by crosses were unstable or metastable (they crystallized immediately upon seeding). Solutions of compositions indicated by the open circles were observed and found to be stable for longer than two months at 25°C.

Figure 2 shows the solubility curve for various mixtures of iron, chromium and nickel nitrates in nitric acid. The composition and densities of these saturated solutions are given in Appendix I. In Figure 2, the crosses are an arbitrary grouping of the compositions of solutions at equilibrium with at least one salt.

B. Solutions of Nickel, Chromium and Uranium

The specific gravity and electrical conductivity of solutions of nickel and chromium nitrate (in the proportions of 80-20 Nichrome) and nitric acid are shown in Table 2. No uranium was present.

Figure 3 illustrates the stability of Nichrome fuel dissolver product solutions. The metal nitrate concentration of the solutions varied from 50 to 400 grams of total metals per liter (metal ratio: Ni:Cr:U=66.3:16.6:17.0) and the nitric acid concentration varied from 0 to 13M. The solutions were allowed to stand in glass bottles for five months at 23°C. Crystallization of the chromium nitrate occured in the unstable solutions in less than two weeks. The upper curve of Figure 3 represents the lower limit of metal concentration in solutions from which crystals separated. The lower curve represents the upper composition limit for solutions in which crystallization did not occur.

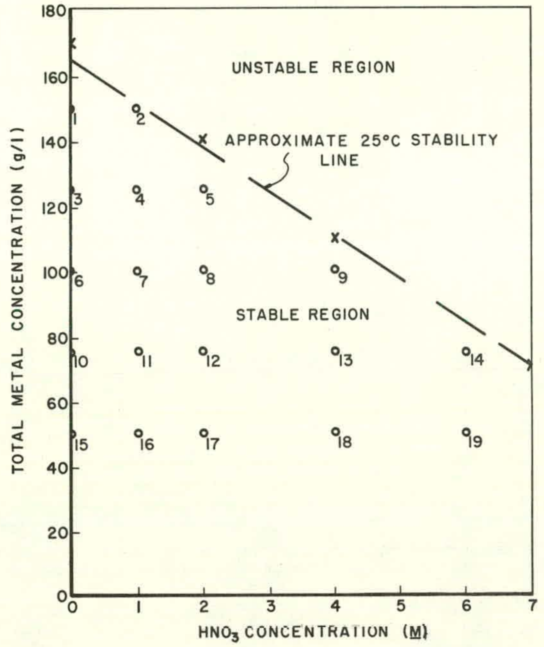

Fig. 1 Stability of APPR Fuel Dissolver Solution at 25°C.

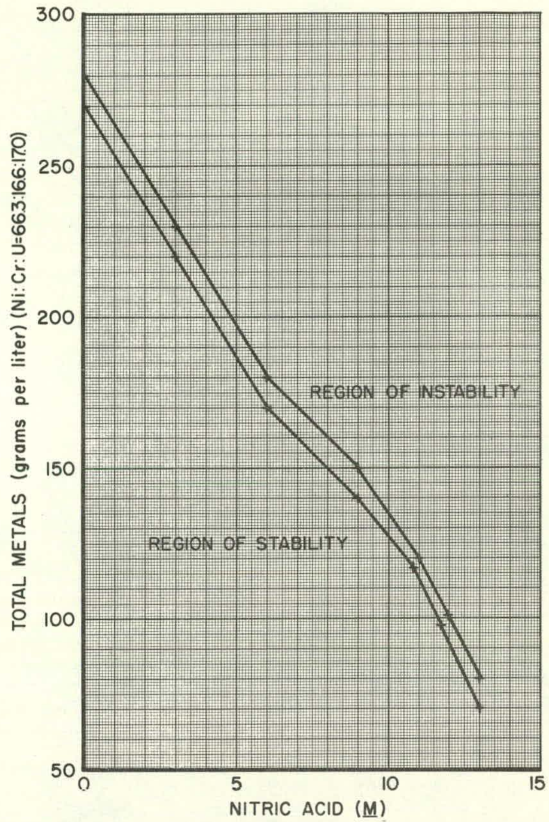

Fig. 3 Stability of Nichrome Fuel Dissolver Solution at 23°C.

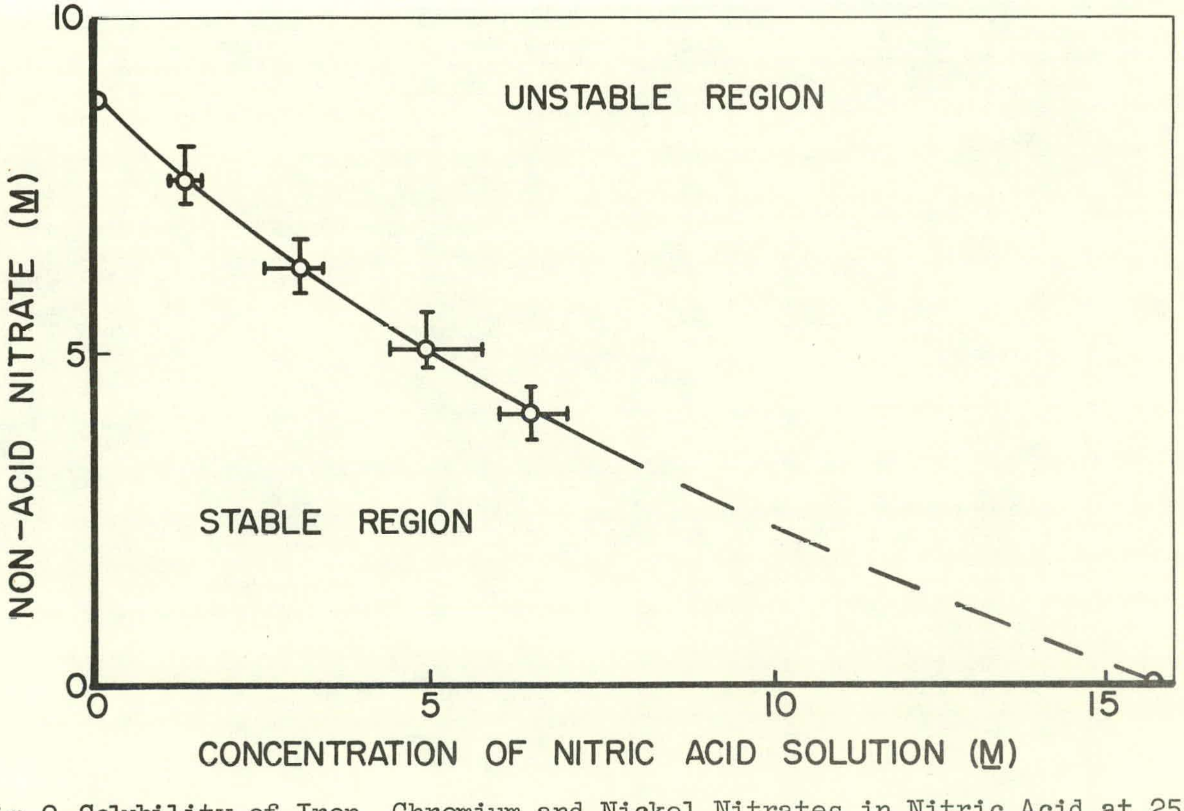

Fig. 2 Solubility of Iron, Chromium and Nickel Nitrates in Nitric Acid at 25°C.

Table 1

Summary of Physical Property Data for Solutions of Stainless Steel and Uranium in Nitric Acid

Total Metal g/l		HNO_3, M 0	1	2	4	6
50	(a)	1.1504	1.1823	1.2079	1.2735	1.3291
	(b)	1.5577	1.6229	1.6693	1.9860	2.2961
	(c)	122	264	376	470	478
	(d)	280	477	665	836	873
75	(a)	1.2284	1.2546	1.2855	1.3440	1.3952
	(b)	2.2483	2.2962	2.4368	2.8238	3.2571
	(c)	132	222	290	349	350
	(d)	306	433	555	671	690
100	(a)	1.2982	1.3256	1.3547	1.4097	
	(b)	2.9092	3.4165	3.6433	4.2782	
	(c)	125	178	220	250	
	(d)	303	382	459	571	
125	(a)	1.3689		1.4262		
	(b)	4.4481		4.9858		
	(c)	108	137			
	(d)	280	325			
150	(a)	1.4381	1.4634			
	(b)	6.8321	7.4115			
	(c)	87	102			
	(d)	248	271			

(a) $\rho^{25°/25}$

(b) $\eta^{25°}$, cp

(c) $10^3 L_{25°C}$

(d) $10^3 L_{80°C}$

Metal consists of 92 per cent stainless steel, 8 per cent uranium by weight.

Stainless steel consists of 19 per cent Cr. 10 per cent Ni, 71 per cent Fe by weight.

Eq. wt. of metal = 20.43

Eq. fraction U in metal 0.0138

Table 2

Summary of Physical Property Data for Solution of Nichrome in Nitric Acid.

"NiCr-NO_3" Eq/l \ HNO_3, Eq/l	0	1	2	3	4	5	6	7	8
0	1.000 0	1.0337 329	1.0669 562	1.0996 714	1.1322 (a) 805 (b) 1.23 (c)	1.1634 849	1.1925 856	1.2232 844	1.2507 816
1	1.0724 67	1.1055 312	1.1368 488	1.1683 601 1.29	1.1995 662	1.2303 689	1.2574 691	668	
2	103	1.1736 286	1.2044 422	1.2345 498	1.2656 540	1.2951 552 1.45	538		
3		1.2414 257	1.2731 347 1.52	407	1.3328 426 1.54				
4	1.2769 131	1.3081 225	1.3393 286	1.3673 320 1.68	1.3946 333 1.73				
5		1.3729 188	1.4040 225	1.4319 245 1.88					
6	113	1.4391 149 2.10	1.4650 175						
7		1.5016 116							
8	1.5392 74								

(a) $\rho^{25°/25}$

(b) $10^3 L_{25°}$

(c) $\frac{d \ln L}{dt} \times 100$

"NiCr" consists of 80 per cent Ni, 20 per cent **Cr**, by weight. Eq. wt. = 26.95

(c) The change in conductance with temperature is applicable between 25° and 45°C.

C. Equations Describing the Physical Property Data

In three cases where the measurements were extensive enough, analytical expressions were derived to represent the data.

For the specific gravity of solutions of nitric acid and the nitrate salts of iron, chromium, and nickel (in 18-8 stainless steel proportions) and uranium, the following expression represents the data of Table 1:

$$\rho^{25/25^\circ} = 1.0000 + 0.0029x + 0.0310y \ (\pm\ 0.005) \text{ where } y \leq 6\underline{M} \text{ and } x \leq 150 \text{ g/l}$$

An expression of greater precision for the same data is:

$$\rho^{25/25^\circ} = 1.0000 + 0.00309x + 0.0344y - 0.00000115x^2 - 0.0000493xy - 0.000381y^2 \ (\pm\ 0.0012)$$

The viscosities in this system are represented by:

$$\eta^{25^\circ} = 0.89 - 0.0103x + 0.101y + 0.000323x^2 + 0.00144xy \ (\pm\ 0.14) \text{ where } x \leq 125 \text{ g/l and } 1 < y < 6$$

In all of the above equations x represents the concentration of total metals in grams per liter and y represents the concentration of nitric acid in moles per liter.

For nitrate dissolver solutions of Nichrome the following expressions may be used for the density:

$$\rho^{25/25^\circ} = 1.0000 + 0.0678z + 0.0322y \ (\pm\ 0.004)$$

$$\rho^{25/25^\circ} = 1.0000 + 0.0717z + 0.0345y - 0.00055z^2 - 0.00090yz - 0.00040y^2 \ (\pm\ 0.0009)$$

Here z is the concentration of nickel and chromium (in 80-20 Nichrome proportions) in equivalents per liter and y is the nitric acid concentration in moles per liter.

V LITERATURE CITED

1. Slansky, C. M., K. L. Rohde, H. T. Hahn, Review of Research and Development at the Idaho Chemical Processing Plant on the Electrolytic Dissolution of Nuclear Fuel, IDO-14535, February (1961).

2. Slansky, C. M., M. W. Roberts, K. L. Rohde, Nuclear Science and Engineering: 12, 33-38 (1962), Electrolytic Dissolution of Reactor Fuels.

3. Roberts, M. W., Laboratory Studies for HTRE Fuel Reprocessing, IDO-14523, June (1961).

4. Aylward, J. R., and E. M. Whitener, Electrolytic Dissolution of Nuclear Fuels, Part II, Nichrome in Nitrate Solutions, IDO-14575, December (1961).

5. Aylward, J. R., and E. M. Whitener, Electrolytic Dissolution of Nuclear Fuels, Part III, Stainless Steel (304) in Nitrate Solutions, IDO-14584.

6. Pearson, D. P., The Inline Analysis of Electrolytic Dissolver Solutions for Nitric Acid and Salt Concentrations, (In Press)

VI. ACKNOWLEDGEMENT

The data reported here were derived from measurements made and compiled by C. E. May, D. P. Pearson, and M. W. Roberts.

Appendix I

Density and Composition of Various Saturated Solutions of Iron, Chromium, Nickel and Uranium Nitrates in Nitric Acid.

Sample	$D_{25°}$	HNO_3 (M)	Fe (M)	Cr (M)	Ni (M)	U (M)	Total NO_3 (M)	Non-acid NO_3 (M)
I - 101	-	0	0.600	0.56	2.51	0.279	9.06	9.06
102	1.655	1.8	0.508	0.44	2.11	0.455	9.77	7.97
103	1.649	3.6	0.385	0.33	1.84	0.533	10.5	6.89
104	1.710	5.3	0.296	0.24	1.46	0.906	11.6	6.33
105	1.684	6.8	0.253	0.20	1.23	0.856	12.3	5.52
II - 101	1.521	0	0.689	0.548	2.54	0	8.79	8.79
102	1.578	2.08	0.551	0.486	2.46		10.1	8.03
103	1.561	4.16	0.456	0.405	2.13		11.0	6.84
104	1.540	6.16	0.379	0.333	1.77		11.8	5.68
105	1.523	7.86	0.332	0.281	1.54		12.8	4.91
111	1.490	1.16	1.79	0.147	0.688		8.35	7.19
115	1.512	1.48	1.31	0.235	1.36		8.83	7.35
112	1.586	1.10	0.152	0.132	3.63		9.21	8.11
116	1.567	1.45	0.299	0.275	3.04		9.25	7.80
113	1.474	1.42	0.150	1.80	0.700		8.67	7.25
117	1.510	1.65	0.288	1.26	1.38		9.03	7.38
121	1.454	2.80	1.34	0.152	0.696		8.67	5.87
125	1.490	3.29	0.964	0.241	1.27		9.45	6.16
122	1.547	2.49	0.133	0.125	2.97		9.20	6.71
126	1.535	3.21	0.276	0.252	2.44		9.67	6.46
123	1.460	2.88	0.148	1.45	0.654		8.98	6.10
127	1.491	3.42	0.277	0.896	1.25		9.44	6.02
131	1.451	4.77	1.13	0.114	0.608		9.72	4.95
135	1.477	5.45	0.711	0.161	1.16		10.4	4.94
132	1.518	4.36	0.133	0.116	2.43		9.97	5.61
136	1.511	5.43	0.246	0.229	1.97		10.8	5.36
133	1.447	5.23	0.114	1.06	0.628		10.0	4.78
137	1.474	5.76	0.224	0.602	1.18		10.6	4.84
141	1.433	6.66	0.812	0.117	0.559		10.6	3.91
145	1.469	6.41	0.579	0.103	1.04		10.5	4.12
142	1.495	6.08	0.131	0.105	1.90		10.6	4.51
146	1.493	6.45	0.206	0.186	1.50		10.6	4.18
143	1.430	6.27	0.233	0.716	0.610		10.4	4.07
147	1.461	7.01	0.144	0.440	0.993		10.7	3.74

PHILLIPS
PETROLEUM
COMPANY

ATOMIC ENERGY DIVISION