methylamine and dimethyl ether containing two molecules of complexing agent per mole of salt. Using J. T. Baker C. P. aluminum chloride which was sublimed several times approximately 1.9 moles of ether or amine react. However, using salt prepared from very pure aluminum as previously described the reaction with two moles of complexing agent was quantitative.

The dietherates and diamine complexes of the aluminum halides decompose with pumping at room temperature to the respective 1:1 addition compounds. These complexes are all solids; hence the dissociation pressures of Table I may be treated as equilibrium constants K_p for the general reaction $AB_2(s) \longleftrightarrow AB(s) + B(g)$ where $AB_2(s)$ is the 2:1 addition compound, $AB(s)$ is the 1:1 complex, and $B(g)$ is the complexing molecule. The dissociation pressure of aluminum chloride dietherate is given by the equation $\log_{10} P_{mm.} = -2462/T + 9.75$. The heat of reaction as calculated from the equation is 11.27 kcal. The dissociation pressure of the 2:1 complex of trimethylamine with aluminum chloride is given by the equation $\log_{10} P_{mm.} = -2280/T + 9.70$. The heat of reaction as computed from the vapor pressure equation is 10.43 kcal. The dissociation pressure of aluminum bromide dietherate is given by the equation $\log_{10} P_{mm.} = -3111/T + 12.25$. The heat of reaction as calculated from the equation is 14.24 kcal. The dissociation pressure of the 2:1 complex of trimethylamine with aluminum bromide is given by the equation $\log_{10} P_{mm.} = -3318/T + 13.05$. The heat of reaction as calculated from the equation is 15.18 kcal. The average deviation of the experimental data in Table I from the values computed from the empirical equations is 1.0% for the aluminum chloride complexes, 1.3% for aluminum bromide dietherate and 0.95% in the case of the diamine complex of aluminum bromide.

A comparison of the heats of reaction is of considerable interest. The value of ΔH for the reaction of $AlBr_3 \cdot (CH_3)_2O$ with ether to produce the dietherate is approximately 3.0 kcal./mole greater than the value for the corresponding reaction of $AlCl_3 \cdot (CH_3)_2O$. A similar comparison in the case of amine complexes shows that the formation of $AlBr_3 \cdot 2(CH_3)_3N$ involves 4.75 kcal./mole in excess of heat of reaction of the chloride complex with amine. On the other hand, the ΔH values for the formation of $AlCl_3 \cdot 2(CH_3)_2O$ and $AlCl_3 \cdot 2(CH_3)_3N$ from the 1:1 complexes differ by only 0.84 kcal./mole. A similar comparison in the case of the bromide complexes shows a difference of 0.94 kcal./mole. These results suggest that the 1:1 aluminum bromide complexes are stronger acids than the corresponding chloride addition compounds. However, a complete discussion of these results must await further investigation, particularly, the determination of the ΔH values for the addition of the first mole of complexing agent to the aluminum halides.

V. Summary

1. Dimethyl ether addition compounds of the gallium halides $GaBr_3 \cdot (CH_3)_2O$, and $GaCl_3 \cdot (CH_3)_2O$ have been prepared and their vapor pressure curves determined.

2. Trimethylamine addition compounds of the gallium halides containing 2 moles of amine per mole of salt are reported.

3. 2:1 complexes of aluminum chloride and bromide with trimethylamine and dimethyl ether are reported.

4. Heats of reaction for the addition of the second mole of complexing agent to the 1:1 aluminum halide complexes have been determined from the vapor pressure curves.

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[CONTRIBUTION FROM THE LABORATORIES OF THE BENZIMA FACTORIES, NATIONAL ENTERPRISE]

The Electrolysis of Aqueous Solutions of Ammonium Iodide

BY FRANK JIRSA

The electrolysis of aqueous solutions of ammonium iodide is characterized by the following primary reactions

$$NH_4I \rightleftarrows NH_4^+ + I^- \qquad (1)$$

$$2NH_4^+ + 2e^- \longrightarrow 2NH_3 + H_2 \qquad (2)$$

$$NH_3 + H_2O \longrightarrow NH_4OH$$

$$2I^- + 2\oplus \longrightarrow 2I \longrightarrow I_2 \qquad (3)$$

At the cathode, hydrogen gas is liberated together with the formation of aqueous ammonia, while iodine is formed at the anode. In a separate compartment secondary processes can take place, reactions which normally do not occur because iodine itself is a very weak iodinating agent, but which are possible during the electrolysis when iodine is present in the nascent state, *i. e.*, in a more active form than is its molecular one

$$NH_4I + 2I \longrightarrow NH_2I + 2HI \qquad (4)$$

$$NH_2I + 2I \longrightarrow NHI_2 + HI \qquad (5)$$

$$NHI_2 + 2I \longrightarrow NI_3 + HI \qquad (6)$$

When solid iodine is caused to react with a concentrated solution of ammonium iodide, none of the above amines will be obtained. An addition compound, ammonium triiodide NH_4I_3, will result, forming nice crystals which are easily hydrolyzed with the separation of iodine.[1,2,3]

It is true that by the interaction of elementary iodine with ammonia under other conditions some compounds have been prepared containing iodine and nitrogen in ratios of I:N = 1, 2, 3, but they were not true amines. Their composition was not uniform and it changed with reaction conditions so that in no case were pure substances isolated. A number of authors attempted to prepare NHI_2[4]; but all products described in the cited papers were nothing else but the addition compound $NI_3 \cdot NH_3$.[5,6] On the other hand it is possible that at the same time an intermediate is formed containing two atoms of iodine.[4i,7]

The product NH_3I_2, prepared by Guthrie[8] and Seamon,[9] is soluble in absolute alcohol, glycerol, less so in benzene and carbon disulfide. It is decomposed according to the equation

$$2NH_3I_2 \longrightarrow NHI_2 + NH_4I + HI \qquad (7)$$

The compound NI_3 known as nitrogen triiodide exists as $NH_3 \cdot NI_3$.[10,5,7b] As already found by Dumas[11] and Silberrad[12] the ratio I:N = 3 is certain. It should be stated that all substances discussed so far, and resulting from the action of iodine on ammonia, are not amines, but amides, since they are derived from hypoiodous acid HIO.

sesquiiodoamide	$NH_3 \cdot NI_3$
diiodylamide	NHI_2
triiodylamide	NI_3

Ammonia reacts first with iodine according to the equation

$$NH_3 + H_2O + I_2 \longrightarrow NH_4I + HIO \qquad (8)$$

then the following reactions take place

$$2NH_3 + 3HIO \longrightarrow NH_3 \cdot NI_3 + 3H_2O \qquad (9)$$

$$NH_3 + 2HIO \longrightarrow NHI_2 + 2H_2O \qquad (10)$$

$$NH_3 + 3HIO \longrightarrow NI_3 + 3H_2O \qquad (11)$$

According to Seliwanow[13] the reaction (11) is reversible so that NI_3 with water yields hypoiodous acid. Iodylamides react with an alkaline base in a similar way as proved by Serullas,[4a] Chattaway

$$NI_3 \cdot NH_3 + 3KOH \longrightarrow 2NH_3 + 3KOI \longrightarrow 2NH_3 + 2KI + KIO_3 \qquad (12)$$

and Orton[5] found that, besides the hydrolysis as the main reaction, a decomposition to nitrogen and hydriodic acid takes place.

In a separated cathode compartment, no opportunity would arise for iodine to react with ammonia to yield HIO. However, if the iodine liberated at the anode reacted with the ammonium iodide according to reactions 4, 5 and 6, there still exists the possibility of the iodyl amides decomposing according to the reverse of reactions 9, 10 and 11. It is not possible to determine whether this decomposition takes place, since any HIO formed by hydrolysis would react with the hydriodic acid produced in reactions 4, 5 and 6, producing free iodine. Inasmuch as the amides were formed from free iodine, the net effect of formation and decomposition *via* these paths is zero and therefore hydrolysis of the amides cannot be detected. Once formed, however, decomposition may also occur like that of chloroamines and bromoamines.[14]

The stability of haloamines decreases with increasing atomic weight of the respective halogen, which makes it doubtful whether iodoamines could be isolated as stable products. However, their transient existence could be detected from their decomposition products

$$4NH_2I \longrightarrow 2NH_4I + N_2 + I_2 \qquad (13)$$

$$3NH_2I \longrightarrow NH_4I + N_2 + 2HI \qquad (14)$$

Thus, if unstable monoiodoamine is formed by the electrolysis of an aqueous solution of ammonium iodide, nitrogen is bound to appear at the anode and at the same time the anolyte should turn acid due to the hydriodic acid according to reactions 4 and 14.

Experimental

Aqueous solutions of ammonium iodide were electrolyzed using a graphite cathode and a platinum anode, separated by a diaphragm. The gas liberated at the anode was collected in a gas buret and analyzed after completion of the electrolysis. The iodine formed was determined by titration with 0.1 N solution of thiosulfate. The acid was titrated by a 0.1 N solution of potassium hydroxide using brom phenol blue as an indicator.

On the assumption that monoiodoamine is decomposed according to reaction 13, 1 mg. of nitrogen is equivalent to 20.42 mg. of NH_2I, 1 cc. of 0.1 N KOH to 7.15 mg. of NH_2I and one ampere-hour to 2666.4 g. of NH_2I.

The results in Table I indicate that both the current efficiencies for NH_2I, calculated from the amount of nitrogen and from the titration, fully agree, and that the sum of efficiencies for iodine, NH_2I and oxygen is 100% within the limit of experimental errors.

This is a proof that monoiodoamine is actually formed

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TABLE I

TEMPERATURE 16°. VOLUME OF ANOLYTE, 250 CC.

No.	NH_4I, g./l.	Amp. per sq. dm.	Amp., hr.	Gas total, cc.	O_2 %	N_2 %	N_2, mg./amp. hr.	O_2 mg./amp./hr.	O_2 efficiency, %	$Na_2S_2O_3$ 0.1 *N* cc.	I g./amp. hr.	I efficiency, %	NH_2I efficiency, %	Total efficiency, %
1	10	0.833	0.0627	4.10	6.83	93.17	5.58	87.06	29.17	17.00	3.400	71.78	4.20	105.15
2	10	0.833	.0800	2.68	19.40	80.60	8.120	38.58	12.93	23.80	3.748	79.15	6.22	98.30
3	10	1.250	.0600	4.20	8.81	91.19	7.700	91.22	30.57	15.30	3.213	67.83	5.90	104.30
4	10	1.250	.0900	2.40	23.33	76.67	7.770	29.22	10.13	27.30	3.850	81.27	5.95	97.35
5	10	1.250	.0600	2.45	26.86	73.14	18.750	58.35	19.55	...	...	...	..	....
6	10	1.670	.0580	..	...	...	....	....	...	19.60	4.258	89.23	..	....
7	10	2.080	.0690	..	...	...	....	....	...	24.00	4.380	92.50	..	....
8	20	1.670	.0788	3.85	9.36	90.64	5.710	63.29	21.21	22.30	3.566	75.27	4.37	100.85
9	30	1.660	.1200	4.30	23.26	76.74	10.410	39.30	13.16	33.80	3.549	74.92	8.00	96.00
10	30	2.000	.1210	6.51	13.98	86.02	9.400	66.24	22.19	33.30	3.473	73.30	7.20	102.70
11	30	2.500	.0980	4.60	12.60	87.40	7.400	58.62	19.64	24.20	3.111	65.48	5.70	90.80
12	50	1.250	.1500	0.00	...	...	....	...	...	56.00	4.700	99.43	..	99.50
13	50	1.670	.2000	2.10	14.52	85.48	1.220	12.82	4.30	71.30	4.492	94.83	0.94	100.00
14	50	1.870	.1800	3.20	14.37	85.63	3.190	21.75	7.29	62.80	4.396	92.80	2.44	102.50
15	50	2.080	.1500	3.40	25.29	74.71	7.170	24.20	8.11	48.70	4.091	86.36	6.97	101.40
16	50	2.700	.1350	3.70	7.56	92.44	2.590	36.20	12.13	24.20	4.041	85.30	2.50	99.90
													Average	99.92

TABLE II

TEMPERATURE 16°, VOLUME OF ANOLYTE, 250 CC.

No.	NH_4I, g./l.	Amp. per sq. dm.	Amp., hr.	$Na_2S_2O_3$, 0.1 *N*, cc.	I g./amp. hr.	I efficiency, %	N_2 mg./amp. hr.	O_2 mg./amp. hr.	O_2 efficiency, %	NaOH 0.1 *N*, cc.	NH_2I efficiency from N_2	NH_2I efficiency from titrat.	Efficiency total, % I	Efficiency total, % II
1	10	1.041	0.0750	26.8	4.502	95.04	2.166	9.53	3.19	0.45	1.66	1.60	99.89	99.83
2	10	1.670	.0580	19.6	4.258	89.23	...	...	...	.50	..	2.34	....	....
3	10	2.080	.0690	24.0	4.380	92.50	...	...	...	.60	..	2.33	....	....
4	20	1.670	.0788	22.3	3.566	75.27	5.710	63.29	21.21	1.30	4.37	4.42	100.85	100.90
5	20	0.833	.0400	15.0	4.725	99.75	0.000	0.00	0.00	0.00	0.00	0.00	....	99.75
6	40	1.670	.0400	15.0	4.725	99.75	0.000	0.00	0.00	0.00	0.00	0.00	....	99.75
7	40	2.400	.0800	23.0	3.622	76.47	6.000	58.00	19.10	1.37	4.59	4.59	100.16	100.16
8	40	2.000	.0900	24.0	3.790	80.00	3.667	53.14	17.50	0.75	2.50	2.50	100.00	100.00

TABLE III

VOLUME OF ANOLYTE, 300 CC.

No.	Temp., °C.	NH_4I, g./l.	Amp. per sq. dm.	Amp., hr.	$Na_2S_2O_3$ 0.1 *N*, cc.	I g./amp. hr.	I efficiency, %	N_2 mg./amp. hr.	O_2 mg./amp. hr.	O_2 efficiency, %	NaOH 0.1 *N*, cc.	Efficiency, % NH_2I	Efficiency, % Total	Gas % N_2	Gas % O_2
1	50	20	1.670	0.0800	30.00	4.725	99.79	0.000	0.00	0.00	0.00	0.00	99.79	0.00	0.00
2	50	20	1.670	.1600	60.20	4.731	99.91	0.000	0.00	0.00	0.00	0.00	99.91	0.00	0.00
3	50	20	2.708	.1137	38.60	4.277	90.30	1.978	24.13	8.08	..	1.52	99.90	8.58	91.42
4	1	20	1.670	.0800	23.20	3.654	77.13	5.781	53.23	17.84	1.35	4.43	99.40	11.04	88.96
5	0	20	2.000	.1000	29.32	3.695	78.00	7.400	58.62	19.64	0.72	2.37	100.01	12.60	87.40

at the anode. Oxygen is liberated by the discharge of hydroxyl ions

$$H_2O \rightleftharpoons H^+ + OH^- \qquad (15)$$

$$2OH^- + 2\oplus \longrightarrow H_2O + O$$

The current efficiencies of iodine, NH_2I and oxygen depend on the concentration of the solution, on the current density, temperature and duration of electrolysis; however, it is impossible to interpret exactly this relation by experiment. The layer of iodine deposited changes the character of the anode in an uncontrollable way so that, under otherwise identical conditions, different results are obtained depending on whether the iodine adheres to the surface of platinum or not, due to some external causes. In some cases (Table I, no. 12; Table II, nos. 5, 6; Table III, nos. 1, 2) neither oxygen nor nitrogen was liberated, the anolyte remained neutral; therefore no NH_2I resulted, so that iodine separated quantitatively, which equally proves that the relation between the amount of nitrogen and the amount of temporarily formed NH_2I corresponds to reality.

Summary

The electrolysis of aqueous solutions of ammonium iodide is represented by fundamental equations

$$2NH_4^+ + 2\ominus \longrightarrow 2NH_3 + H_2$$

$$2I^- + 2\oplus \longrightarrow 2I \longrightarrow I_2$$

The iodine formed reacts further with NH_4I to give HI and NH_2I

$$NH_4I + 2I \longrightarrow NH_2I + 2HI$$

The monoiodoamine is not stable and decomposes according to the equation

$$4NH_2I \longrightarrow 2NH_4I + N_2 + I_2$$

A side-reaction, *i. e.*, the discharge of hydroxyl ions, leads to liberation of oxygen

$$2OH^- + 2\oplus \longrightarrow H_2O + O$$

The electrolysis results in aqueous ammonia being formed at the cathode, iodine with nitrogen and oxygen gas at the anode.

ROUDNICE, CZECHOSLOVAKIA

RECEIVED JANUARY 14, 1949

[CONTRIBUTION FROM OAK RIDGE NATIONAL LABORATORY]

The Absorption Spectrum of Element 61, Promethium[1,2]

BY G. W. PARKER AND P. M. LANTZ

Introduction

Since the isolation of milligrams[3] of promethium in chemically pure state is now almost routinely performed in this Laboratory, advantage has been taken of the opportunity to investigate some of the properties of the synthetic element. Among the investigations conducted was a study of the absorption of light in the visible spectrum as ordinarily done with the rare earth elements by means of the quartz spectrophotometer.

In the fission products of uranium resulting in the chain reacting pile the 4 year isotope 147 of promethium has been shown to occur with a yield of about 2.5%.[4] Sources of mixed fission products sufficient to yield milligrams of promethium have been isolated and the rare earth elements individually separated by ion-exchange methods to produce the first recognizable quantities of the substance of atomic number 61. While the 4 year half-life of the isotope is sufficient to permit freedom in time for handling and experimentation, in order to make investigations involving milligrams of mass it was necessary to work with curies of radioactivity.

J. A. Marinsky and L. E. Glendenin reported in 1945 the isolation and radiochemical identification of the synthetic rare earth promethium in the uranium fission products. Later Winsberg[5] estimated its half-life through observed decay to be 3.7 years, which would indicate its specific activity to be approximately 0.66 curie per milligram.

Improvements in separation techniques having made available milligram quantities of this element, determination of its specific activity (approximately 0.6 curie per milligram) by weighing and absolute beta counting was performed during the course of this absorption study.

Experimental Details

Material.—Prior to this absorption study, the same sample in the form of $PmCl_3$ was used by L. E. Burkhart, W. F. Peed and E. J. Spitzer[6] (Oak Ridge Electromagnetic Plant) to record the K and L emission lines of the X-ray spectrum of promethium. About 3 mg. of the sample of promethium was recovered from the X-ray tube and was tested for purity[7] before spectrophotometric study was made, by comparing copper spark spectrograms with the known lines of the other rare earths and those characterized at this laboratory for promethium.[8] Part of this sample was used by C. Feldman[9] (this Laboratory) to look for "Illinium" lines reported by Harris, Hopkins and Yntema.[10] The results of the spectrographic purity test are given in the table below:

TABLE I

IMPURITIES FOUND IN PROMETHIUM (20–30 MICROGRAMS)

Analyzed by the copper spark method on a Jarrell-Ash grating spectrograph

		Elements not found
Al	Trace	Ag, As, Au, B, Be, Bi, Cd,
Ca	Strong	Ce, Co, Dy, Er, Eu, Ga, Gd,
Cr	Moderate	Ho, K, La, Li, Lu, Mn, Mo,
Fe	Faint trace	Nd, Ni, Pr, Rb, Sb, Si, Sm,
Mg	Moderate	Sn, Sr, Ta, Ti, Tm, V, Zn
Na	Trace	
Pb	Very weak	
Pt	Presence questionable	

In preparation for absorption study, the portion of the sample remaining after applying the spectrographic test above was analyzed for gross beta activity by absolute beta counting techniques and was found to be 822 mc. It was then converted to the $PmCl_3$ and was transferred in 0.2 ml. of 0.1 *N* HCl solution to a calibrated semi-micro quartz cell 2.5 mm. × 10 mm. (10 mm. length of light-path).

After the study was completed the promethium was converted to the oxalate and weighed. Then the oxalate was ignited to the oxide and reweighed.

The promethium (calculated as 0.926 mg. of the element) obtained as the oxide was then found to be 551 mc. On the basis of this weight-activity relationship the mo-

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[1] This document is based on work performed under Contract Number W-7405 eng 26 for the Atomic Energy Project at Oak Ridge National Laboratory.

[2] The name promethium is used in view of the action of the International Union of Chemistry at the XVth Conference at Amsterdam, September, 1949, reported in the *Chemical and Engineering News*, **27**, 2996 (1949). The declassified manuscript on which this paper was based was one of the important ones used in establishing the validity of the promethium claim.

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