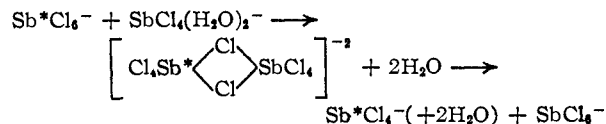shorter than the hydrolysis half-time, as it is in most of the 6 f experiments.

These observations show that neither the interaction absorption complex nor the activated complex for catalyzed hydrolysis is symmetrical. If they were symmetrical, exchange would accompany these reactions.

If the c form of Sb(V) has six chlorides attached to it, the activated complex for exchange has only a limited number of possible structures. Since hydrolysis does not accompany exchange, the Sb atom which was originally in the +3 state must have six chlorides attached to it in the activated complex, as indicated in the example.

If the Sb(III) atom still had H_2O attached while it was in the activated complex, the Sb(V) result-

$$Sb^*Cl_6^- + SbCl_4(H_2O)_2^- \longrightarrow$$

$$\left[Cl_4Sb^* \underset{Cl}{\overset{Cl}{<}} SbCl_4 \right]^{-2} + 2H_2O \longrightarrow$$

$$Sb^*Cl_4^-(+2H_2O) + SbCl_6^-$$

ing from the exchange would have the H_2O attached to it and would thus be hydrolyzed. It is not necessary that the exchange activated complex be symmetrical, but it seems reasonable that it should be.

Acknowledgments.—We gratefully acknowledge the assistance of Mrs. Georgia M. Stephas who performed most of the antimony and acid analyses. We also wish to thank Jack Matthews, Thomas N. Haratani and Karl O. Malmquist for the numerous machine computations they made for us.

Sodium Imidodiphosphate. Synthesis, Identification and Hydrolytic Degradation

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Sodium imidodiphosphate was obtained in 95% yield from saponification of diphenyl imidodiphosphoric acid at 135°. Its identification and assay for purity were made by X-ray diffraction and nuclear magnetic resonance methods. Hydrolytic stability was determined in solutions of 20% concentration and the thermodynamic constants calculated.

Sodium imidodiphosphate, $Na_4P_2O_6NH$, has been prepared by the condensation of disodium phosphoramidate, $Na_2PO_3NH_2$, at 210° by Klement and Biberacher.[1] The product is ordinarily contaminated with impurities of orthophosphate and pyrophosphate, as well as unreacted phosphoramidate, so that a tedious fractional crystallization must be employed to effect a separation. Furthermore, the condensation is quite slow, so that a reaction time of about a week has been recommended. It has now been found that the presence of a fluxing material greatly accelerates the reaction and reduces the formation of undesirable by-products. For this purpose, either potassium formate or potassium phenoxide has proved suitable.

A simple and direct synthesis of sodium imidodiphosphate consists in the saponification of diphenyl imidodiphosphoric acid, the latter compound having been described by Kirsanov and

$$(C_6H_5O)_2P(O)NHP(O)(OH)_2 + 6 \ NaOH \longrightarrow$$
$$Na_4P_2O_6NH + 2 \ C_6H_5ONa + 4 \ H_2O$$
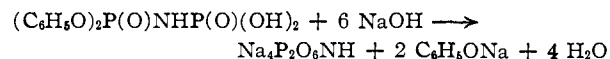
Zhmurova.[2] Unexpectedly, under conditions which readily yield the sodium phosphoramidate or phosphorodiamidate from their phenyl esters,[1,3] there was obtained the monophenyl derivative, $(C_6H_5O)(NaO)P(O)NHP(O)(ONa)_2$. However, when the reaction temperature was raised to 135–150°, either by the addition of sodium phenoxide or by maintaining about 30 lb./sq.in. pressure, the reaction proceeded quickly to yield the tetrasodium

salt. The same process, with potassium hydroxide substituted for sodium hydroxide, has yielded the tetrapotassium salt. While the tetraphenyl ester[2] can also be saponified under similar conditions, its preparation is not as simple as that of the diphenyl imidodiphosphoric acid.

The identification of sodium imidodiphosphate, as well as its assay in mixtures, has been done by paper chromatography,[1,4] but the application of X-ray diffraction and nuclear magnetic resonance has resulted in two new simple, rapid methods. The X-ray diffraction (x.r.d.) technique requires the preparation of crystalline silver salts of imidodiphosphate and possible impurities, in this case phosphoramidate, ortho- and pyrophosphates. Very finely divided amorphous precipitates, typically formed with silver nitrate, are avoided by starting the precipitation slowly with solid silver acetate. Identification of the various phosphorus compounds by nuclear magnetic resonance (n.m.r.), based on the appearance of their characteristic resonance peaks,[5] depends on the assumption of non-interference of peaks due to other phosphorus compounds or ionic species.

The hydrolytic degradation of imidodiphosphate has been reported[4] for 0.02 M solutions at 60°. In the present work 20% solutions at 60 and 98° were used, and the activation energy for the process is recorded.

Experimental

Preparation of $Na_4P_2O_6NH·10H_2O$. A. Condensation of Sodium Phosphoramidate in the Presence of a Flux.—

(1) R. Klement and G. Biberacher, Z. anorg. allgem. Chem., 283 246 (1956).

(2) A. V. Kirsanov and I. N. Zhmurova, Zhur. Obshchei Khim., 28, 2487 (1958); C. A., 53, 3118i (1959).

(3) H. N. Stokes, Am. Chem. J., 15, 198 (1893); 16, 123 (1894).

(4) O. T. Quimby, A. Narath and F. H. Lohman, THIS JOURNAL, 82, 1099 (1960).

(5) J. R. Van Wazer, C. F. Callis, J. N. Shoolery and R. C. Jones, ibid., 78, 5715 (1956).

Disodium phosphoramidate hexahydrate (40 g., 0.16 mole) was dehydrated under oil pump vacuum, with phosphorus pentoxide in a side-bulb, by the schedule: 65°, 4 hr.; 90°, 2 hr.; 110°, 16 hr.; 125°, 2 hr. It was then cooled, crushed to a fine powder in a dry-box and mixed intimately with 50 g. of finely crushed potassium phenoxide. The mixture was heated under oil pump vacuum, with KOH in a side-bulb, by the schedule: 160°, 1 hr.; 200°, 3 hr.; 230°, 24 hr. On cooling, the solids, which had sintered together, were broken up and dissolved in 250 ml. of water. The solution was filtered to clarify it, and the solids were precipitated with denatured ethanol. On reprecipitating from water by slowly adding five volumes of ethanol, 32 g. of needles was obtained. Purity, by n.m.r., was about 95% with small amounts of phosphoramidate, orthophosphate and pyrophosphate present, so that the yield of imidodiphosphate was 86%.

Replacement of potassium phenoxide by potassium formate (adjusted to a pH of 11 by fusing with KOH) gave similar but less favorable results. Recrystallized solids weighed 16 g. and were of 80% purity by n.m.r., representing a yield of 36%. The lower yield may be due to decomposition of the potassium formate. Any water present during the reaction would result in the formation of ortho- and pyrophosphates.

B. Saponification of Diphenyl Imidodiphosphoric Acid at 135°.—A solution of sodium hydroxide (17.6 g., 0.44 mole), phenol (18.8 g., 0.2 mole) and diphenyl imidodiphosphoric acid[2] (6.6 g., 0.02 mole) in 50 ml. of water was heated to boiling in a flask, allowing the water to distil out until the pot temperature reached 135°, whereupon a reflux condenser was connected. After heating at 135° for 15 min., the solution was cooled and the solids recovered as above; yield 8.5 g. (95%) as $Na_4P_2O_6NH \cdot 10H_2O$. Purity (by n.m.r.) $\geqslant 98\%$.

Anal. Calcd. for $Na_4P_2O_6NH \cdot 10 H_2O$: N, 3.15. Found: N, 3.06; C, 0.40.

In another experiment, diphenyl imidodiphosphoric acid and sodium hydroxide, in aqueous solution, were heated in an autoclave to 135° (about 30 lb./sq. in.) for 1 hr. The product, precipitated by ethanol and recrystallized from water with ethanol, represented a 67% yield of Na_4-$P_2O_6NH \cdot 10 H_2O$ of 98% purity (by n.m.r.); carbon content 1.08%. (We believe that the carbon content is entirely due to carbonate, as the sample evolved CO_2 when acidified and, in addition, gave no test for phenol esters by the micro test of Feigl and Jungreis.[6])

Preparation of Trisodium Monophenyl Imidodiphosphate by Saponification at 110°.—To a hot solution of sodium hydroxide (9.13 g., 0.23 mole) in 30 ml. of water was added diphenyl imidodiphosphoric acid (10 g., 0.03 mole). The solution was boiled for 7 min., then chilled to 10° and precipitated with ethanol; yield 9 g. (78%). The crystals gave a new characteristic x.r.d. pattern. The n.m.r. spectrum for P^{31} showed a single peak at 0.0 p.p.m. (with respect to 85% H_3PO_4). This differs from the two peaks (+2.1, +2.9) for disodium diphenyl imidodiphosphate, prepared by dissolving the acid in cold sodium hydroxide solution. It also differs from the chemical shift for sodium imidodiphosphate (−2.5).

Anal. Calcd. for $C_6H_6NP_2O_6Na_3 \cdot 3.5 H_2O$: C, 18.9; H, 3.4; N, 3.7; P, 16.2. Found: C, 21.5; N, 4.0; P, 16.1.

Identification and Assay of Imidodiphosphate by X.r.d.—X-Ray equipment consisted of the General Electric XRD-3 X-ray Diffraction Unit, with CuKα radiation filtered through nickel foil ($\lambda = 1.5418$ Å.). The instrument settings were: X-ray tube current, 15 mamp.; X-ray tube potential, 50 kv.; defining slit, 1°; Soller slit, wide; detector slit, 0.2°; target-to-beam angle, 4°; scanning rate, 0.2°/min.; recorder range, 2; time constant, B; and recorder chart speed 0.4 in./min.

The crystalline silver salt, $Ag_5P_2O_6N$, containing whatever impurities were present, was prepared in several ways. Ordinarily, the use of silver acetate gave the most satisfactory results. Since others[7] have reported difficulty in obtaining crystalline silver salts, the method will be given in detail.

(6) F. Feigl and E. Jungreis, *Anal. Chem.*, **31**, 2101 (1959).

(7) R. Klement, Inter. Union Pure Applied Chem., Colloquium of Section for Inorg. Chem., Münster, Sept. 1954, Verlag Chemie, Weinheim. Germany, 1954, p. 217.

Silver acetate crystals of a uniformly small size are prepared by adding silver nitrate solution to a hot solution of ammonium acetate and chilling in an ice-bath. They are filtered off and dried in air. About 0.4 g. sample is dissolved in 10 ml. of water in a 50 ml. beaker, about 0.1 g. of silver acetate crystals is added and the mixture is allowed to stand undisturbed for several minutes. When seeds of the yellow salt, $Ag_5P_2O_6N$, are definitely present, the mixture is stirred, preferably with a magnetic bar, and silver acetate crystals added in portions of about 0.2 g. as long as white solids persist (the white silver acetate tends to float on the surface), up to a total of about 0.9 g. Supplementary inspection using a microscope is of considerable help. During the precipitation it is important that the pH does not drop below 6.0. Brom thymol blue is used as a suitable indicator, and 5% NaOH is added as necessary. Excess Ag^+ must be present at the end and can be shown in a separate test of the supernatant liquid with a sodium orthophosphate solution. The crystals are filtered off with suction, washed with water (which removes excess silver acetate), alcohol and ether and air-dried briefly.

Anal. Calcd. for $Ag_5P_2O_6N$: N, 1.97; P, 8.71. Found: N, 1.92, 2.36; P, 8.75, 9.00.

An alternate method, avoiding the use of silver acetate crystals, consists of adding excess silver nitrate solution to the sample, carefully redissolving in ammonium hydroxide, then very slowly acidifying. This is best done by the hydrolysis of acetic anhydride, a few drops added to a quiet layer of benzene on the solution. After seeds of yellow silver salt have formed, the solution is stirred gently and about 20 drops of acetic anhydride added over a 10 min. period. Finally, 2 N HNO_3 can be added dropwise until the colorimetric pH is 6.0–6.5.

The X-ray diffraction pattern was obtained in the conventional manner, grinding the sample to pass 325 mesh screen and packing it into an open-frame sample holder against a ground glass plate to minimize preferred orientation of crystallites. The region of $2\theta = 26$–$36°$ was scanned.

TABLE I

ASSIGNMENT OF X-RAY DIFFRACTION LINES OF SILVER SALTS

2θ(CuKα)	Intensity (counts/second)			
	Ag_3PO_4	$Ag_4P_2O_7$	$Ag_2PO_3NH_2$	$Ag_5P_2O_6N$
27.15		95		
27.30				5
28.80		40		
29.75	110			
30.50		35	235	
31.75				245
32.35		350 ⎫		
32.40		⎬ a	220	
32.55		350 ⎭		
33.25		30		
33.30			110	
33.35	845			
33.65			65	
34.10				235
34.20		40		
34.90				95
35.25			110	
35.65			105	

ᵃ Doublet whose resolution varies with crystallite size—sometimes looks like just one broad line—when resolved detectably the 32.55° 2θ line is slightly less intense than the 32.35° line.

Table I indicates the lines in this region. The limit of detection of the three impurities in imidodiphosphate, under ideal conditions, using synthetic mixtures of well-crystallized samples, is of the order of 4% when phosphoramidate is present; in its absence, orthophosphate and pyrophosphate can be detected at 1% and 2%, respectively (by weight as the silver salts). Similarly, in the absence of pyrophosphate, phosphoramidate can be detected at about 2% concentration.

Other Crystalline Salts for Identification by X.r.d.—In addition to the pentasilver salt described above, the

TABLE II

CRYSTALLINE SALTS OF IMIDODIPHOSPHATE

Compound	Three most intense diffraction lines d, Å. (Relative intensity given in parentheses)		
Diammonium dihydrogen, $(NH_4)_2H_2P_2O_6NH \cdot 2H_2O$	3.700 (100)	7.41 (91)	6.39 (40)
Triammonium monohydrogen, $(NH_4)_3HP_2O_6NH \cdot 1.5H_2O$	5.262 (100)	3.063 (93)	4.332 (78)
Tetraammonium, $(NH_4)_4P_2O_6NH \cdot 3H_2O$	5.735 (100)	5.454 (25)	3.900 (18)
Tetrasodium, $Na_4P_2O_6NH \cdot 10H_2O$	2.660 (100)	4.396 (88)	8.01 (43)
Dipotassium dihydrogen, $K_2H_2P_2O_6NH$	3.678 (100)	3.132 (80)	3.284 (47)
Tetrasilver, $Ag_4P_2O_6NH$	2.614 (100)	2.797 (80)	2.574 (67)
Pentasilver, $Ag_5P_2O_6N$	2.818 (100)	2.629 (95)	2.571 (39)
Tetrathallium, $Tl_4P_2O_6NH$	3.678 (100)	2.718 (52)	3.132 (25)
Benzidinium, $(C_{12}H_{12}N_2)_{1.5}(H_4P_2O_6NH) \cdot 2H_2O$	16.7 (100)	4.439 (45) 4.374	3.867 (44)

following salts were obtained in crystalline form. The strongest lines in their X-ray diffraction patterns are given in Table II. Complete data have been reported elsewhere.[8]

Tetrasilver imidodiphosphate, $Ag_4P_2O_6NH$,[7,9] was obtained under slightly acidic conditions. A solution of 11 g. (0.025 mole) of $Na_4P_2O_6NH \cdot 10H_2O$ in 300 ml. of water, with 3 g. of NH_4NO_3 and sufficient HNO_3 to give a pH of 2.5 (yellow color of 2,4-dinitrophenol as internal indicator), was allowed to stand 10 minutes with about 0.5 g. of silver acetate to form seed crystals. Then, with stirring, 70 ml. of 20% $AgNO_3$ solution was added in small portions, the pH at the same time being maintained at about 2.5 by dropwise additions of NH_4OH. Finally, the pH was lowered to 2.0 by adding HNO_3. The solids recovered by filtration and washing were white; yield 10.7 g. (72%).

Anal. Calcd. for $Ag_4P_2O_6NH$: Ag, 71.39; N, 2.32; P, 10.25. Found: Ag, 70.07; N, 2.41; P, 10.49.

Thallium imidodiphosphate, $Tl_4P_2O_6NH$, was prepared by neutralizing a solution of the acid with TlOH solution to pH 12. The acid was prepared[1] by stirring a solution of $Na_4P_2O_6NH$ with a two-fold excess of the acid form of cation exchange resin IR-120 at 0–5° for 10 minutes and filtering off the resin. Crystals of the desired salt were obtained by partially evaporating under vacuum and then precipitating with ethanol; yield 95%. The infrared spectrum of this salt, as well as the spectra of a number of the other salts described, has been reported elsewhere.[10]

Anal. Calcd. for $Tl_4P_2O_6NH$: N, 1.41; P, 6.25; Tl, 82.5. Found: N, 1.43; P, 6.07; Tl, 81.7.

Dipotassium dihydrogen imidodiphosphate, $K_2H_2P_2O_6NH$, was prepared from the tetrapotassium salt (obtained by metathesis of $Ag_5P_2O_6N$ with a 25% solution of K_2S, separating as an oil with methanol and drying to a brittle solid under vacuum) by acidifying an aqueous solution with acetic acid to methyl red end-point, and then precipitating with methanol.

Anal. Calcd. for $K_2H_2P_2O_6NH$: K, 30.9; N, 5.53; P, 24.47. Found: K, 29.3; N, 5.44; P, 24.23.

Tetraammonium imidodiphosphate, $(NH_4)_4P_2O_6NH \cdot 3H_2O$, was obtained by metathesis of $Ag_5P_2O_6N$ with a 25% solution of $(NH_4)_2S$, removing the Ag_2S and precipitating the crystalline ammonium salt with dimethyl formamide. After rinsing the crystals briefly with ether, they were blown dry with NH_3 and preserved under NH_3.

Anal. Calcd. for $H_{17}N_5O_6P_2 \cdot 3H_2O$: N, 23.41; P, 20.71. Found: N, 25.07; P, 20.99.

Triammonium monohydrogen imidodiphosphate, $(NH_4)_3HP_2O_6NH \cdot 1.5 H_2O$, was obtained when the tetraammonium salt stood several hours in air.

Anal. Calcd. for $H_{14}N_4O_6P_2 \cdot 1.5 H_2O$: N, 21.96; P, 24.28. Found: N, 22.54; P, 24.30.

Diammonium dihydrogen imidodiphosphate, $(NH_4)_2H_2P_2O_6NH \cdot 2H_2O$, was obtained by acidifying a solution of the tetraammonium salt with acetic acid to pH 4.7 and precipitating the crystalline salt with dimethyl formamide.

(8) The X-ray powder data have been submitted to G. W. Brindley, Editor, "A. S. T. M. X-Ray Powder Data File," College of Mineral Industries, Pennsylvania State University, University Park, Pa.

(9) H. N. Stokes, *Am. Chem. J.*, (1896).

(10) J. V. Pustinger, Jr., W. T. Cave and M. L. Nielsen, *Spectrochim. Acta.* 909–925 (1959).

Anal. Calcd. for $H_{11}N_3O_6P_2 \cdot 2H_2O$: N, 17.01; P, 25.07. Found: N, 18.36; P, 25.57.

Benzidinium imidodiphosphate, $(C_{12}H_{12}N_2)_{1.5}(H_4P_2O_6NH) \cdot 2H_2O$, was obtained by mixing solutions of the sodium salt (2.23 g. or 0.005 mole in 700 ml. water) and benzidine (1.84 g. or 0.01 mole in 122 ml. of water and 61 ml. of glacial acetic acid); yield 1.43 g. (59%).

Anal. Calcd. for $C_{18}H_{23}N_4O_6P_2 \cdot H_2O$: C, 44.17; H, 5.56; N, 11.45; P, 12.66. Found: C, 43.83; H, 5.62; N, 11.63; P, 12.43.

Identification and Assay of Imidodiphosphate by N.m.r.

The measurements were made with a Varian Model V-4300B High Resolution NMR Spectrometer at a frequency of 16.192 mc. The samples were contained in 15 mm. test tubes filled to a depth of 3 cm. with solution. Since the position of the peak for orthophosphate is dependent upon pH, all solutions were made strongly alkaline (pH above 12) by addition of KOH. Solutions were made as concentrated as possible for maximum signal-to-noise ratio. Table III summarizes the position of the peaks for the respective compounds.

TABLE III

NUCLEAR MAGNETIC RESONANCE PEAKS

Compound		Chemical shift (p.p.m.) relative to 85% orthophosphoric acid (± 0.5)
Phosphoramidate	$Na_2PO_3NH_2$	−8.9
Orthophosphate	Na_3PO_4	−5.4
Imidodiphosphate	$Na_4P_2O_6NH$	−2.5
Pyrophosphate	$Na_4P_2O_7$	+5.5

The limit of detection of impurities in imidodiphosphate is about 1–2% as found by the analysis of solutions containing impurities added in known amounts.

Quantitative analysis by n.m.r. for any component utilizes the ratio of its peak area to that of the total area under all the (phosphorus) peaks since nuclear resonance signals are directly proportional to the number of nuclei in a given environment.

The analysis of known mixtures by the above methods gave satisfactory results. For example, in a mixture of 75% $Na_4P_2O_6NH \cdot 10 H_2O$ and 25% $Na_2PO_3NH_2 \cdot 6 H_2O$, X.r.d. found, respectively, 75% and 25%, while n.m.r. found 77% and 23%. In another mixture containing 83% $Na_4P_2O_6NH \cdot 10H_2O$, 12% $Na_4P_2O_7 \cdot 10 H_2O$ and 5% $NaH_2PO_4 \cdot H_2O$, X.r.d. found, respectively 82 ± 2%, 12 ± 2% and 6%, while n.m.r. found 82%, 13% and 5%.

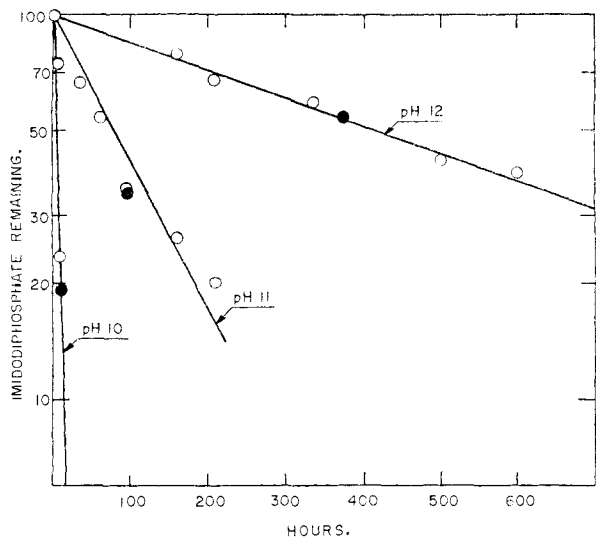**Hydrolytic Degradation.**—The hydrolytic degradation was studied as a function of temperature

Fig. 1.—Hydrolysis of sodium imidodiphosphate in water at 60°: O, initial hydrolysis; ●, repeat hydrolysis.

and pH for comparison with that of pyrophosphate.[11] Solutions of 20% concentration in water were prepared and adjusted to the desired pH (10, 11 and 12) by addition of HCl or NaOH. They were placed in polyethylene bottles and held in a shaker mechanism in a water bath at the desired temperature (automatically thermostated, ±0.25°). For the runs at 60°, the pH was

TABLE IV

OBSERVED FIRST ORDER RATE CONSTANTS FOR DISAPPEARANCE OF IMIDODIPHOSPHATE

	⌐Half-life (hr.)⌐		⌐Rate constant⌐ (hr.$^{-1}$ × 10^3)	
pH	60°C.	98°C.	60°C.	98°C.
10	8		85	
11	80	1.25	8.5	550
12	410		1.7	

adjusted twice daily, for the runs at 98°, every half hour. Samples were taken for analysis from each bottle, and shaking of the remaining solution continued for the time that was left. Each sample was cooled rapidly to room temperature and the hydrolysis quenched by adding KOH pellets until the pH was greater than 13. Analyses were made

(11) J. R. Van Wazer, E. J. Griffith and J. F. McCullough, THIS JOURNAL, 77, 287 (1955).

by n.m.r., the percentage of phosphorus present as imidodiphosphate being reported. Hydrolysis of imidodiphosphate gave a mixture of orthophosphate and phosphoramidate, a secondary reaction resulting in the hydrolysis of the phosphoramidate to orthophosphate

$$\begin{matrix} O & & O \\ \| & H & \| \\ ^-O{-}P{-}N{-}P{-}O^- & + & OH^- \longrightarrow \\ \| & & \| \\ ^-O & & O^- \end{matrix}$$

$$\begin{matrix} O & & O \\ \| & & \| \\ ^-O{-}P{-}NH_2 & + & ^-O{-}P{-}O^- \\ \| & & \| \\ & & O^- \end{matrix}$$

$$\begin{matrix} O & & O \\ \| & & \| \\ ^-O{-}P{-}NH_2 + OH^- \longrightarrow & ^-O{-}P{-}O^- + NH_3 \\ \| & & \| \\ O^- & & O^- \end{matrix}$$
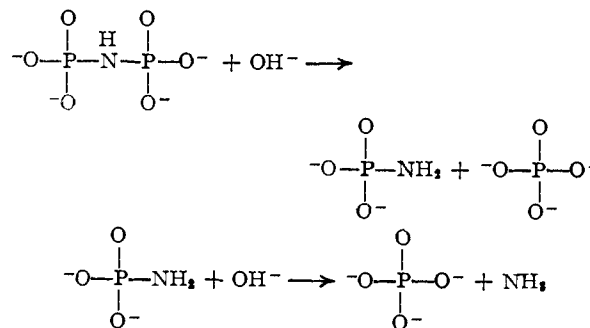
Figure 1 shows the results obtained at 60° expressed on the basis of percentage imidodiphosphate left at a given time. While the points were obtained in a single series of runs, one duplicate point for each pH is included to show the replication obtained. In another run made at 98°, pH = 11, the half-life was found to be 1.25 hr. The rate constants are summarized in Table IV. From these, the thermodynamic data were calculated[12] as: E_A = +28.5, $\Delta F^{\ddagger}{}_{30}°$ = +25.2, and $\Delta S^{\ddagger}$ = +8.9. Compared with pyrophosphate, the imidodiphosphate is less stable in the pH range 10–12.

Solubility and Calcium Sequestration.—The solubility in water was found to be 34.8 g. of Na$_4$-P$_2$O$_6$NH·10H$_2$O/100 ml. of water at 25°. Calcium sequestration at pH 12 by an oxalate titrimetric method[13] was 3.0 g. of calcium/100 g. of Na$_4$P$_2$O$_6$NH·10H$_2$O, comparable to 2.3 g. of calcium/100 g. of Na$_4$P$_2$O$_7$·10 H$_2$O.

(12) A. A. Frost and R. G. Pearson, "Kinetics and Mechanisms," 1st Ed., John Wiley and Sons, Inc., New York, N. Y., 1953, pp. 95–99.

(13) R. R. Irani and C. F. Callis, presented by R. R. Irani before the Division of Inorg. Chem., 137th meeting of the American Chemica Society, Cleveland, Ohio, April, 1960.