1172. *The Study of the Equilibrium Constant of Complex Formation between Phenol and Pyridine in Solution by a Dielectric Constant Method.*

By R. J. Bishop and L. E. Sutton.

Few and Smith's dielectric-constant method is used to measure the equilibrium constant complex formation, and dipole moment of the complex, between phenol and pyridine in benzene, carbon tetrachloride, and n-heptane solutions in order to check and extend results obtained by spectroscopic methods. Solvent effects are detected. Excess moments are derived and discussed.

One of the fundamental steps in studying complexes in solution is to determine the equilibrium constant of the reaction $x\text{A} + y\text{B} \rightleftharpoons \text{A}_x\text{B}_y$ in the presence of a third substance S as solvent. It is necessary to ascertain x/y, which is usually done by some form of Job's method,[1] and then measure the actual concentration of one species, *viz.*, A, B, or A_xB_y, the initial concentrations of two of A, B, or A_xB_y being known.

The available methods for measuring concentrations are all of limited usefulness. For the particular case of complex formation through hydrogen bonding[2] the most commonly used ones for such homogeneous three-component systems are infrared or ultraviolet spectroscopy. The first of these methods cannot normally be used for solutions in hydrocarbons but only in solvents such as carbon tetrachloride, whereas for the second the reverse applies. Cross-checking of methods is therefore limited. If, as actually happens, there is discordance between the results, it is not certain whether this shows genuine solvent effect or is merely due to systematic error.

Few and Smith[3] developed a method based on dielectric-constant measurements from a suggestion made by Hammick, Norris, and Sutton,[4] and Cleverdon, Collins, and Smith[5] applied it. This method is not as quick and possibly not as accurate as the spectroscopic methods but it can be applied to solutions in any non-polar solvent. In addition, because the electric dipole moment of the complex can be obtained, this method may give information about the stereochemistry of the complex and the electronic perturbations therein.[6] The method has now been applied to determine the equilibrium between pyridine and phenol in benzene, carbon tetrachloride, and n-heptane solutions in order to extend and clarify the earlier spectroscopic investigations and also in order to establish it further.

[1] P. Job, *Ann. Chim. (France)*, 1928, **9**, 113.
[2] G. C. Pimental and A. L. McClellan, "The Hydrogen Bond," Freeman, San Francisco, 1960.
[3] A. V. Few and J. W. Smith, *J.*, 1949, 2781.
[4] D. Ll. Hammick, A. Norris, and L. E. Sutton, *J.*, 1938, 1755.
[5] D. Cleverdon, G. B. Collins, and J. W. Smith, *J.*, 1956, 4499.
[6] J. R. Hulett, J. A. Pegg, and L. E. Sutton, *J.*, 1955, 3901.

EXPERIMENTAL

Purification of Compounds.—Benzene (AnalaR) was fractionally frozen three times, an unfrozen fifth part being discarded each time, leaving about one-half of the original amount. This was kept over phosphorus pentoxide, distilled from fresh phosphorus pentoxide in a stream of dry air on the day of use, and kept under a pressure of dry air. Carbon tetrachloride (AnalaR and n-heptane (I.P. specification) were kept over phosphorus pentoxide, distilled from fresh phosphorus pentoxide on the day of use, and kept under a pressure of dry air. Phenol (AnalaR) was distilled at atmospheric pressure (b. p. 182°); the middle cut was redistilled into breakable glass phials which were sealed until used. Pyridine (AnalaR) was fractionally distilled through a 1 m. × 1 cm. column packed with glass helices; the middle cut (b. p. 115°) was redistilled and pipetted into phials similar to those used for phenol.

Physical Measurements.—Measurements of dielectric constant (ε) and specific volume (v) were made on dilute solutions of phenol (A) in solvent (S) and in various constant mixtures of pyridine (B) and solvent. In the pure solvent both the dielectric constant and the specific volume were linear functions of the weight fraction of phenol (w_A), and the slopes of these functions were used in the Halverstadt and Kumler[7] expression to calculate the extrapolated total molar polarisation of phenol in the solvent $(P_{A\infty})_S$. In the mixed solvents the specific volume was a linear function of w_A within the limits of experimental error, but the dielectric constant was not. Within the limits of experimental error the variation of dielectric constant was adequately fitted to a cubic function of w_A over the range of concentrations used. The limiting value of $d\varepsilon/dw_A$ at infinite dilution was taken as the intercept of $(\varepsilon_{ABS} - \varepsilon_{BS})/w_A$ as a function of w_A, which was fitted by computer[8] to a cubic polynomial in w_A. The extrapolated total molar polarisation of phenol in the mixed solvent $(P_{A\infty})_{BS}$ is calculated using the slope dv/dw_A and the limiting value of $d\varepsilon/dw_A$ at infinite dilution in the Halverstadt and Kumler expression. w_B' is the weight fraction of pyridine in the solvent mixture, M_B is the molecular weight of pyridine, ΔP is the change in total molar polarisation on making 1 mole of complex from phenol and pyridine in solution. ΔP is used to calculate the dipole moment of the complex, ΔP and the equilibrium constant K l. mole^{-1} being obtained from the Few and Smith[3] type of expression:

$$\frac{w_B'}{v_{BS}[(P_{A\infty})_{BS} - (P_{A\infty})_S]} = \frac{w_B'}{v_{BS}\Delta P} + \frac{M_B}{K\Delta P}$$

The derivation of this type of expression involves the assumption of 1 : 1 complex formation, and this has been shown to occur between phenol and pyridine in 0·1 molar solution by Chrétien and Laurent[9] who studied the dielectric constant of various mixtures and applied Job's method.

The measurement of physical properties has involved the assumption of the following values for pure solvents (n is the refractive index for sodium light):

TABLE 1.

Solvent properties.

Solvent	Temp.	ε	v	n
Benzene	25°	2·2741[10]	1·1446[11]	1·4980[11]
	40	2·2440[12]	1·1665[11]	—
Carbon tetrachloride	25	2·2280[12]	0·6312[11]	1·4576[11]
	45	2·1880[12]	0·6467[11]	—
n-Heptane	25	1·9139[12]	1·4717[11]	1·3851[11]
	45	1·8890[12]	1·5103[11]	

[7] I. F. Halverstadt and W. D. Kumler, *J. Amer. Chem. Soc.*, 1942, **64**, 2988.
[8] J. H. Cadwell, Ferranti Mercury Computer Library Service, Autocode Programme 1000, Issue 1, 1959.
[9] A. Chrétien and P. Laurent, *Compt. rend.*, 1932, **195**, 792.
[10] L. Hartshorn, J. V. L. Parry, and L. Essen, *Proc. Phys. Soc.*, 1955, **68B**, 422.
[11] J. Timmermans, "Physico Chemical Constants of Pure Organic Compounds," Elsevier, Amsterdam, 1950.
[12] A. A. Maryott and E. R. Smith, "Tables of Dielectric Constants of Pure Liquids," N.B.S. Circular 514, 1951.

TABLE 2.

Results for solutions.

Phenol in benzene + pyridine at 25°.

$10^6 w_B'$	$10^6 w_A$	ε	v	n	$10^6 w_B'$	$10^6 w_A$	ε	v	n
0	0	2·2741	1·1446	1·49800	3565	0	2·2982	1·1442	1·49800
	600	2·2758	1·1445	—		1455	2·3104	—	1·49804
	1560	2·2782	1·1444	—		4642	2·3322	1·1433	1·49816
	2445	—	1·1441	1·49817		7708	2·3493	1·1425	1·49841
	2510	2·2808	1·1441	—		12,992	2·3739	—	1·49871
	3300	2·2828	1·1439	—		16,543	2·3882	1·1404	1·49890
	3454	—	1·1438	1·49821	3869	0	2·2997	1·1440	1·49803
	3790	2·2840	1·1438	—		770	2·3067	1·1439	1·49805
	7217	—	1·1430	1·49836		1623	2·3137	1·1437	1·49807
	13,238	—	1·1415	1·49874		3089	2·3246	1·1434	1·49818
647	0	2·2786	1·1445	1·49800		5654	2·3414	1·1429	1·49834
	754	2·2820	—	1·49804	8552	0	2·3311	1·1433	1·49807
	1948	2·2866	1·1442	1·49804		1651	2·3496	1·1430	1·49813
	3877	2·2934	1·1438	1·49815		2953	2·3630	1·1426	1·49830
	5604	2·2993	1·1433	1·49822		4732	2·3810	1·1421	1·49841
	8001	2·3068	1·1428	1·49830		8156	2·4118	—	1·49860
					5333	0	2·3095	1·1440	1·49800
					16,268	0	2·3835	1·1425	—

Phenol in benzene + pyridine at 40°.

$10^6 w_B'$	$10^6 w_A$	ε	v	$10^6 w_B'$	$10^6 w_A$	ε	v
0	0	2·2440	1·1665	2905	0	2·2619	1·1660
	2025	2·2490	1·1660		956	2·2678	1·1659
	3068	2·2516	1·1658		2134	2·2743	1·1656
	4797	2·2561	—		5287	2·2901	—
	7577	2·2630	1·1649		9449	2·3077	1·1638
	10,060	2·2690	1·1642		13,386	2·3227	1·1629
552	0	2·2476	1·1665	7611	0	2·2909	1·1654
	1154	2·2516	1·1661		1506	2·3042	1·1650
	2660	2·2565	—		3531	2·3211	1·1646
	4438	2·2619	—		7221	2·3476	1·1636
	6816	2·2689	1·1648		13,075	2·3835	—
	9074	2·2753	1·1643		17,785	2·4075	1·1609
				1536	0	2·2537	1·1664

Phenol in carbon tetrachloride + pyridine at 25°.

$10^6 w_B'$	$10^6 w_A$	ε	v	n	$10^6 w_B'$	$10^6 w_A$	ε	v	n
0	0	2·2280	0·6312	1·45760	1202	0	2·2442	0·6317	1·45775
	1287	2·2336	0·6317	1·45799		982	2·2574	0·6319	
	3607	2·2442	0·6324	1·45824		3199	2·2786	0·6326	
	6461	2·2577	0·6333	1·45860		5384	2·2951	0·6333	
	9430	2·2718	—	1·45903		7593	2·3091	0·6340	
	12,280	2·2851	0·6360	1·45942	2307	0	2·2589	0·6320	
327	0	2·2323	0·6314			1510	2·2834	0·6325	
	481	2·2362	0·6315			3830	2·3126	0·6332	
	1468	2·2429	0·6318			5950	2·3331	0·6338	
	2583	2·2495	0·6321			9712	2·3617	0·6350	
	3535	2·2547	—		4565	0	2·2894	0·6330	1·45816
	4353	2·2591	0·6327			1670	2·3222	0·6334	1·45860
						4430	2·3685	0·6342	1·45895
						7553	2·4088	0·6351	1·45935
						10,075	2·4352	0·6358	1·45990

Phenol in carbon tetrachloride + pyridine at 45°.

$10^6 w_B'$	$10^6 w_A$	ε	v	$10^6 w_B'$	$10^6 w_A$	ε	v
0	0	2·1880	0·6467	664	0	2·1960	0·6470
	1640	2·1944	—		749	2·2016	0·6472
	2871	2·1996	0·6476		1817	2·2089	—
	4373	2·2056	—		2800	2·2147	0·6477
	6439	2·2143	0·6487		4147	2·2223	—
285	0	2·1915	0·6468		6088	2·2327	0·6489
	331	2·1934	—	1351	0	2·2041	0·6471
	978	2·1969	0·6471		395	2·2081	—
	2066	2·2025	0·6471		1619	2·2195	0·6477
	3182	2·2081	—		3490	2·2342	0·6483
	4608	2·2148	0·6482		5205	2·2457	—
					7048	2·2572	0·6493

TABLE 2. (*continued*).

Phenol in n-heptane + pyridine at 25°.

$10^6 w_B'$	$10^6 w_A$	ε	v	n	$10^6 w_B'$	$10^6 w_A$	ε	v	n
0	0	1·9139	1·4717	1·38510	2144	0	1·9238	1·4708	1·38526
	5784	1·9243	—	1·38560		1503	1·9333	—	1·38548
	11,525	1·9347	1·4660	1·38610		3437	1·9431	1·4688	1·38573
	17,523	1·9452	1·4630	1·38669		5671	1·9517	1·4677	1·38598
	25,395	1·9595	—	1·38758		7760	1·9582	1·4665	1·38619
0	0	1·9139	1·4717	1·38510		12,147	1·9698	1·4642	1·38657
	1663	1·9169	1·4712	1·38519	4713	0	1·9358	1·4697	1·38556
	4086	1·9210	—	1·38540		1837	1·9505	1·4686	1·38577
	5781	1·9242	—	1·38561		2949	1·9584	1·4679	1·38585
	7579	1·9276	—	1·38589		4434	1·9681	1·4671	1·38602
	9940	1·9315	1·4668	1·38595		6728	1·9806	1·4660	1·38627
1197	0	1·9194	1·4711	1·38522		12,440	2·0042	—	1·38686
	1545	1·9271	1·4703	1·38539		15,035	2·0124	1·4612	1·38720
	2864	1·9320	—	1·38547	8361	0	1·9525	1·4682	1·38577
	4022	1·9358	—	1·38556		1972	1·9702	—	1·38598
	5399	1·9396	—	1·38577		4214	1·9885	1·4657	1·38623
	6599	1·9428	1·4678	1·38590		6437	2·0048	—	1·38652
						10,138	2·0284	1·4625	1·38690
						16,709	2·0600	1·4588	1·38762

Phenol in n-heptane + pyridine at 45°.

$10^6 w_B'$	$10^6 w_A$	ε	v	$10^6 w_B'$	$10^6 w_A$	ε	v
0	0	1·8890	1·5103	1663	0	1·8918	1·5100
	3177	1·8940	—		1936	1·9035	—
	6102	1·8989	1·5070		3683	1·9090	1·5078
	10,211	1·9059	1·5049		5702	1·9149	1·5069
	13,190	1·9109	1·5034		8722	1·9226	—
696	0	1·8918	1·5100		12,471	1·9310	1·5029
	965	1·8946	—	5888	0	1·9136	1·5078
	1682	1·8965	1·5091		2154	1·9274	—
	3014	1·8996	1·5085		4019	1·9381	—
	4490	1·9029	1·5076		4693	1·9416	1·5053
	6621	1·9074	1·5066		8653	1·9604	1·5031
					13,502	1·9791	—
					18,598	1·9953	1·4975

Values of functions for phenol

Temp.	$10^6 w_B'$	$d\varepsilon/dw_A$	dv/dw_A	dn/dw_A	$(_T P_A^\infty)_{BS}$	$_E P_A$ Calc.: 27·7
Benzene						
25°	0	2·61 ± 0·03	−0·22 ± 0·01	0·06	72·1	27·3
	647	4·86	−0·21 ± 0·01	0·04	112·0	28·2
	3565	8·99	−0·22 ± 0·01	0·05	183·5	28·2
	3869	9·63	−0·19 ± 0·01	0·06	195·4	29·5
	8552	11·4	−0·24 ± 0·02	0·07	222·3	28·8
40°	0	2·50 ± 0·01	−0·23 ± 0·02		71·5	
	552	3·59	−0·24 ± 0·01		91·2	
	2905	6·41	−0·26 ± 0·02		142·2	
	7611	9·13	−0·25 ± 0·01		192·2	
Carbon tetrachloride						
25°	0	4·55 ± 0·01	0·36 ± 0·03	0·15	72·5	29·7
	327	8·69⁻	0·27 ± 0·03		112·2	
	1202	15·33	0·29 ± 0·02		177·2	
	2307	17·99	0·31 ± 0·01		203·5	
	4565	20·73	0·27 ± 0·02	0·17	226·9	29·1
45°	0	4·05 ± 0·03	0·31 ± 0·01		67·7	
	285	5·89	0·32 ± 0·02		87·1	
	664	7·79	0·32 ± 0·02		106·7	
	1351	10·33	0·32 ± 0·02		132·9	

TABLE 2—*continued*

n-Heptane

25°	0	1·795 ± 0·005	−0·496 ± 0·002	0·10	70·1	28·7
	1197	5·94	−0·50 ± 0·01	0·10	182·0	28·9
	2144	6·96	−0·55 ± 0·01	0·13	208·1	29·9
	4713	8·57	−0·57 ± 0·01	0·11	249·7	27·7
	8361	9·38	−0·56 ± 0·01	0·11	269·5	28·3
45°	0	1·64 ± 0·02	−0·53 ± 0·01		67·3	
	696	3·12	−0·51 ± 0·02		109·4	
	1663	4·46	−0·54 ± 0·02		146·2	
	5888	6·82	−0·55 ± 0·01		210·7	

Values of functions for pyridine

Temp.	$d\varepsilon/dw_B$	dv/dw_B	dn/dw_B	$(_T P_{B\infty})_S$	$_E P_B$ Calc.: 24·5
Benzene					
25°	6·68 ± 0·05	−0·14 ± 0·02	0	123·0	25·3
40°	6·16 ± 0·05	−0·14 ± 0·02	—	118·5	—
Carbon tetrachloride					
25°	13·45 ± 0·02	0·40 ± 0·02	0·12	136·4	25·4
45°	12·0 ± 0·1	0·30 ± 0·02	—	126·2	—
n-Heptane					
25°	4·62 ± 0·01	−0·42 ± 0·02	0·08	124·7	24·7
45°	4·18 ± 0·02	−0·42 ± 0·01	—	118·8	—

DISCUSSION

The variation of the apparent total polarisation of phenol ($P_{A\infty}$) with mole fractions of pyridine (f_B') in the solvent mixture at 25° is shown in the Figure.
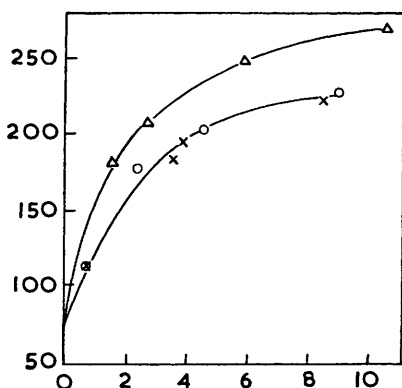
Graph of apparent total polarisation of phenol at 25° ($P_{A\infty}$) in c.c. (as ordinate) against mole fraction of pyridine (f_B') (as abscissa).

Solvents: × = benzene,
            ○ = carbon tetrachloride,
            △ = n-heptane.

From this it appears that the complex is more stable in n-heptane than in either benzene or carbon tetrachloride and this is borne out by the equilibrium constants (see Table 3). It is clear also that equilibrium-constant values from dielectric-constant observations agree well with those obtained by infrared measurements on carbon tetrachloride solutions and by ultraviolet measurements on n-heptane solutions. Furthermore, although the error limits are greater in the values from dielectric measurements than in the best of the spectroscopic ones, we confirm the view of Few and Smith that the dielectric method is very useful. Curve fitting (see Experimental section) by computer gives a great increase in consistency.

There is a definite solvent effect which is especially marked in benzene solutions. This is probably due to complexing of one or more components with the solvent; even if the equilibrium constant for this were much smaller than that for the A–B reaction, the relatively great solvent concentration might make it important. Interaction between phenol and

TABLE 3.

Equilibrium results for phenol and pyridine.

Solvent	Temp.	Ref.	Method	K (l. mole^{-1})	$-\Delta H$ (kcal. mole^{-1})	$-\Delta S$ e.u.
Benzene	25°	a	ε	36 ± 2 ⎫	9 ± 2	24 ± 3
	40	a	ε	17 ± 1 ⎭		
Carbon tetrachloride	25	a	ε	45 ± 5 ⎫	7 ± 2	17 ± 3
	45	a	ε	21 ± 1 ⎭		
	18—20	13	i.r.	55 ± 10		
	25	14	i.r.	42		
	20	15	i.r.	52·1 ± 1·5 ⎫	6·5 ± 0·4	14·4 ± 1·3
	55	15	i.r.	15·5 ± 0·6 ⎭		
	30—60	16	i.r.	5		
n-Heptane	25	a	ε	80 ± 5 ⎫	6 ± 2	11 ± 3
	45	a	ε	43 ± 2 ⎭		
	22	17	u.v.	88 ⎫	6	11·4
	40	17	u.v.	49 ⎭		

a = present work.

benzene or halogenocarbons has already been reported from infrared studies[18,19] as has been interaction between pyridine and carbon tetrachloride from dielectric studies.[20] The small variations of electric dipole moment in the several solvents (see Table 4) are consistent with these interactions and suggest further that there is no specific interaction between pyridine and benzene.

TABLE 4.

Measured dipole moments (D).

Solvent	Temp.	Phenol	Pyridine	Complex	$\Delta\mu$
Benzene	25°	1·47 ± 0·02	2·20 ± 0·02	4·06 ± 0·05	0·6
	40	1·51 ± 0·02	2·20 ± 0·02	4·20 ± 0·05	0·7
Carbon tetrachloride	25	1·47 ± 0·01	2·34 ± 0·01	4·15 ± 0·05	0·6
	45	1·45 ± 0·01	2·30 ± 0·02	4·11 ± 0·05	0·6
n-Heptane	25	1·44 ± 0·01	2·21 ± 0·01	4·30 ± 0·03	0·9
	45	1·44 ± 0·01	2·22 ± 0·01	4·25 ± 0·03	0·9

It is relatively easy to show that the true equilibrium constant, K_{AB}, is related to the apparent one, K'_{AB}, reported in Table 3, by the expression

$$K_{AB} = K'_{AB}(1 + sK_{AS})(1 + sK_{BS})$$

where K_{AS} and K_{BS} are the association constants for reactants with solvent (all evaluated in concentration units) and s is the concentration of the solvent. From the equilibrium constant for the phenol–benzene interaction[21] which is 0·25 l. mole^{-1} at 25° and the concentration of benzene (11·1 moles l.$^{-1}$), assuming that activity equals concentration and also that there is no pyridine–benzene interaction (see above), a value of $K_{AB} = 136$ l. mole^{-1} is obtained. This is considerably greater than the value of 80 ± 5 at 25° found for K'_{AB} in n-heptane which we believe approximates to K_{AB}, the true value, because there is no evidence for specific interaction of either component with this solvent. The agreement is

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about as good as can be expected and it certainly shows the importance of such specific solvent interactions. The values of K'_{AB} in carbon tetrachloride are nearer to those in benzene than to those in n-heptane so the arguments against regarding benzene as an inert solvent in hydrogen-bonding studies may on occasion apply also to carbon tetrachloride. Because values for K_{AS} and K_{BS} are lacking, no correction can yet be made to the apparent equilibrium constant in this solvent. Solvent effects on the infrared spectra of pure compounds in solution have been widely discussed. Several attempts have been made to correlate band shifts in solution with dielectric effects and with specific interactions with the solvent.[18, 22, 23]

Values of $-\Delta H$ calculated from the variation of K'_{AB} with temperature are given in Table 3. These are likely to be affected by solvent, so the value of 6 kcal. mole^{-1} found from both spectroscopic and dielectric measurements on n-heptane solutions has most significance.

From the electric dipole moment found for the complex (see Table 4) the excess moment $\Delta\mu$, arising from the electron and nuclear perturbations caused by hydrogen bonding, can be obtained if the moment of the unperturbed system can be calculated. For this to be done, however, the relative orientations of the moments of phenol and of pyridine must be known, *i.e.*, the structure must be known.

(I)

(II)
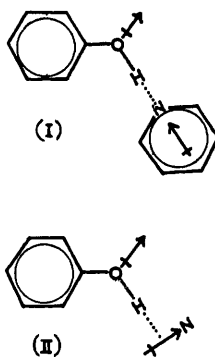
Structure (I) is the more probable but, because of the complexing found between phenol and benzene[2] and because of the structure found for the bromine–benzene compound,[24] the possibility of a structure such as (II) cannot be ignored. Assuming the moment of phenol to act towards the oxygen lone pairs at an angle of 66·25 °with the Ph–O direction and at 138·25° with the O–H bond,[25] a value of 3·44 D is calculated for the moment of (I) and of 2·65 D for (II). The excess moments calculated for structure (I) are given in Table 4; they average about 0·7 D, but the value of 0·9 D in n-heptane is probably the most significant. The excess moment for structure (II) would be at least 3 D (the pyridine molecule could rotate about the OH axis), which is so improbably large that this structure is rejected. The excess moment calculated for structure (I) is comparable with those obtained by Hulett, Pegg, and Sutton[6] for complexes between trimethylamine and various phenols, by a different method but again assuming a structure of type (I).

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