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PROGRESS IN THE ANALYSIS OF INORGANIC AND ORGANIC COMPOUNDS IN NON-AQUEOUS SOLUTIONS

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I. INTRODUCTION

Until recently, chemists used chiefly chemical and physicochemical methods of analysis in the study of matter, based on reactions in aqueous solutions. As a result, while physical chemists up to the end of the 19th century studied the properties of aqueous solutions, analytical chemists developed methods of analysis in such solutions. At the present time all the theories, definitions and chemical and physicochemical methods of analysis in analytical chemistry are based on principles developed for aqueous solutions.

The rapid increase in the production of monomeric and polymeric organic compounds led to considerable interest in the analytical chemistry of non-aqueous solutions since analytical methods developed for aqueous solutions are inapplicable to such compounds. This is indicated by the ever-increasing number of original and review articles and monographs published in different countries on problems associated with the theory of non-aqueous solutions and their practical application[1-26].

The idea of using non-aqueous solutions for analytical purposes has recently attracted the attention not only of analytical chemists, but also of many physical and organic chemists[27-53].

In non-aqueous solutions it is possible to determine various compounds by alkalimetric and acidimetric titration[54-80]. On the basis of the specific properties shown by salts in non-aqueous solutions, it is possible to titrate non-aqueous salt solutions successfully by the precipitation method, making use of the insolubility, in non-aqueous solvents, of certain salts which are soluble in water[81-83]. Since the solvents influence redox potentials, it is possible to titrate non-aqueous solutions by oxidimetric methods[84-96]. The use of non-aqueous solutions for analytical purposes can also be extended to complexometric, photometric, chromatographic, electrochemical, and other analytical methods[97-114].

In view of the vast amount of published work on all these methods, we shall in the present review consider only acid-base titration methods in non-aqueous solutions, of greatest theoretical and practical interest.

The use of non-aqueous solvents makes it possible to extend considerably the possibilities of acid-base titration. Methods of non-aqueous titration are of great importance in the study of inorganic substances and particularly for the analysis of monomeric and polymeric organic compounds. The application of non-aqueous solutions in analysis is extremely promising, since it shows a number of advantages over aqueous solutions[15,17].

Considerable progress has now been made in the analysis of inorganic and organic compounds, and of organic derivatives of the elements, in non-aqueous solutions. It is sufficient to point out that only by acid–base titration in non-aqueous media is it possible to determine compounds irrespective of their pK, isomers, chemical compounds of the same homologous series, substances which have nothing in common with our usual ideas of acids and bases, multicomponent mixtures of various compounds, etc. This progress is due chiefly to the development of the chemical theory of solutions, whose foundations were laid by the Russian school of D.I.Mendeleev[17,20,26,115-130].

II. FUNDAMENTALS OF THE THEORY OF NON-AQUEOUS SOLUTIONS

Detailed studies of solutions of various substances in liquid inorganic and organic solvents and in liquefied gases have shown that acids and bases exist not only in aqueous solution, in which there is a dynamic equilibrium between hydrogen and hydroxide ions, but also in non-aqueous solutions, in which this equilibrium is not present [7,9,15,17,20,26,130-132].

Study of the solutions shows that the individual character of the solvent strongly influences the state of ions in solution. This effect is indicated, for example, by data on the electrical conductivity of salts which, although showing equal degrees of dissociation in aqueous solutions, behave differently in solvents of different chemical character with similar values of the dielectric constant, for example in methanol, acetonitrile, and nitromethane [117,118,124-126,133].

The solvent also influences the direction of the reaction. For example, the reaction

$$H_2S + I_2 \rightleftarrows 2HI + S,$$

proceeds from left to right in aqueous solution and from right to left in the vapour phase, but does not take place at all in non-aqueous solvents [for example in CCl_4, $(C_2H_5)_2O$, etc.].

Very often the influence of the solvent on a chemical reaction arises through formation of a substance which is insoluble in it, which displaces the equilibrium towards the formation of the substance. Thus, for example, copper nitrate and sulphuric acid in anhydrous acetic acid react to yield a precipitate of copper sulphate [9,134,135]:

$$Cu(NO_3)_2 + H_2SO_4 \xrightarrow[CH_3COOH]{} \downarrow CuSO_4 + 2HNO_3.$$

In liquid ammonia the reaction of silver chloride with barium nitrate yields a precipitate of $BaCl_2$:

$$2AgCl + Ba(NO_3)_2 \xrightarrow[NH_3]{} \downarrow BaCl_2 + 2AgNO_3.$$

To chemists these reactions are unusual, as is the behaviour of certain electrolytes in non-aqueous solutions, for example that of urea, which is a weak base in water, an acid in liquid ammonia, and a strong base in anhydrous acetic acid. Ethyl alcohol, which is an indifferent (neutral) substance in aqueous solution, shows clearly-defined basic character in liquid hydrogen fluoride and can be titrated with strong acids in the same way as caustic soda in aqueous solution. This is because liquid hydrogen fluoride has a marked influence on the solute and as a result of its marked protogenic nature readily gives up protons even to those substances from which water removes protons. Thus alcohols, and even phenols, which react as acids in water, behave as bases in liquid hydrogen fluoride:

$$ROH + HF \longrightarrow [ROH_2]^+ + F^-.$$

Liquid hydrogen fluoride reacts similarly even with acetic and formic acids, acetates, nitrates, and other compounds, which behave as bases in this medium:

$$CH_3COOH + HF \rightleftarrows [CH_3COOH_2]^+ + F^-,$$
$$HCOOH + HF \rightleftarrows [HCOOH_2]^+ + F^-,$$
$$CH_3COOK + 2HF \rightleftarrows K^+ + [CH_3COOH_2]^+ + 2F^-,$$
$$KNO_3 + 2HF \rightleftarrows K^+ + [H_2NO_3]^+ + 2F^-.$$

The different action shown by different solvents on electrolytes has been studied by many workers [26,130] and was noted by Walden [124-126]. The action of many solvents on a solute is analogous to the action of water. Thus Jander and Kraffczyk [136] concluded that the reaction of ethyl ether as solvent is analogous to that of water:

$$HOH \rightleftarrows H^+ + OH^-,$$
$$C_2H_5OC_2H_5 \rightleftarrows C_2H_5^+ + OC_2H_5^-.$$

Thus the reaction

$$C_2H_5Br + LiOC_2H_5 \longrightarrow C_2H_5OC_2H_5 + LiBr,$$

can be regarded as a neutralisation involving the formation of a molecule of ether (the solvent) and salt.

The reaction of aluminium bromide with ethyl ether

$$AlBr_3 + C_2H_5^+OC_2H_5^- \rightarrow C_2H_5^+ + [AlBr_3 \cdot OC_2H_5]^-,$$

can be regarded as the reaction of a Lewis acid with a base.

Janz and Danyluk [137], who studied the ionisation of HCl in acetonitrile, represented the reactions as follows:

$$CH_3CN + HX \rightleftarrows CH_3CN \cdot HX, \quad (1)$$
$$CH_3CN \cdot HX \rightleftarrows CH_3CNH^+X^-, \quad (2)$$
$$CH_3CNH^+X^- \rightleftarrows CH_3CNH^+ + X^- \text{ (solv.)}, \quad (3)$$
$$HX + X^- \text{ (solv.)} \rightleftarrows HX_2^- \text{ (solv.)}. \quad (4)$$

The overall reaction is

$$CH_3CN + 2HX \rightleftarrows CH_3CNH^+ + HX_2^- \text{ (solv.)};$$

from Eqns. (1) and (2)

$$CH_3C{\equiv}NH^+X^- \rightleftarrows CH_3C^+{=}NH \cdot X^-,$$
$$CH_3C^+{=}NH \cdot X^- + HX \rightleftarrows CH_3\overset{X}{\overset{|}{C}}{=}NH_2^+X^-,$$
$$CH_3\overset{X}{\overset{|}{C}}{=}NH_2^+X^- \rightleftarrows CH_3\overset{X}{\overset{|}{C}}{=}NH_2^+ + X^- \text{ (solv.)}.$$

A detailed study of the influence of acidic solvents on the properties of electrolytes and the theory of the problem have been described by Shkodin [130].

According to Izmailov [26], the observed interactions of an electrolyte KA with a solvent M can be represented as a series of coupled dynamic equilibria:

1. Formation of solvates:

$$KA + nM \rightleftarrows KAM_n$$

with instability constant

$$K^*_{inst} = \frac{a^*_{KA}}{a^*_{KAM_n}}. \quad (1)$$

The asterisk denotes that the activities refer to infinite dilution of the solution, and n is the number of molecules of solvent.

2. Dissociation of the solvated product with the formation of solvated ions:

$$KAM_n + mM \rightleftarrows K^+(\text{solv.}) + A^-(\text{solv.}),$$

$$K^*_{diss} = \frac{a^*_{K^+(solv)}\, a^*_{A^-(solv)}}{a^*_{KAM_n}}, \quad (2)$$

where m is the number of molecules of solvent.

3. The formation (particularly in solvents with a low dielectric constant) of ion pairs:

$$K^+(\text{solv.}) + A^-(\text{solv.}) \rightleftarrows K^+(\text{solv.})\, A^-(\text{solv.}),$$

$$K^{-1}_{ass} = \frac{a^*_{K^+(solv)}\, a^*_{A^-(solv)}}{a^*_{K^+(solv)\, A^-(solv)}}. \quad (3)$$

Bruckenstein[138], who studied the state of electrolytes in anhydrous acetic acid, concluded that even the strongest electrolytes are very slightly dissociated and exist only in the form of ion pairs, and that in a medium with a very low dielectric constant triple and quadruple ions are formed, complicating the quantitative treatment of the processes taking place in the solution. For solvents whose dielectric constant differs from that of acetic acid[6,16] the picture is different.

The ratio of the activities of the different products of these reactions depends on the properties and concentration of the solute and solvent.

The dissociation constant measured by ordinary methods, K_{ord}, expressing the ratio of the activity product of the free ions to the total activity of undissociated electrolyte species,

$$K_{ord} = \frac{a^*_{K^+(solv)}\, a^*_{A^-(solv)}}{\Sigma a^*_{undiss}}, \qquad (4)$$

can be written

$$K_{ord} = \frac{K_{diss}}{1 + K^*_{inst} + K_{conv}}, \qquad (5)$$

where K_{conv} is a conversion constant taking account of the conversion of the undissociated addition product to an ion pair:

$$KAM_n + x\,M \rightleftarrows K^+(solv.)\,A^-(solv.) + y\,M,$$

$$x + y = m,$$

$$K_{conv} = \frac{a^*_{K^+(solv)\,A^-(solv)}}{a^*_{KAM_n}}. \qquad (6)$$

Different processes may predominate, depending on the nature of the solute and the properties of the medium, so that Eqn. (5) may assume a number of particular forms. For example, the acid-base equilibria anhydrous acetic acid, which has marked protogenic character and low dielectric constant (ϵ = 6.16), are expressed by the following scheme[138-141]:

$$HX \underset{(I)}{\rightleftarrows} H^+X^- \underset{(II)}{\rightleftarrows} H^+ + X^-.$$

The equilibrium constant of reaction (I), referred to by Izmailov as the conversion constant K_{conv} and by Kolthoff as the ionisation constant K_i,

$$K_i = \frac{(H^+X^-)}{[HX]},$$

is related to K_{diss} and K_{ass} by the expression

$$K_{conv} = K_i = K_{diss}K_{ass},$$

or

$$K_{diss} = K_{conv}K^{-1}_{ass} = K_iK^{-1}_{ass}.$$

The equilibrium constant of reaction (II), referred to by Kolthoff as K_d, expresses the dissociation of the ion pairs and is equivalent to the reciprocal of the association constant K^{-1}_{ass}, *i.e.*

$$K_d = \frac{[H^+][X^-]}{[H^+X^-]}. \qquad (7)$$

Kolthoff represents the state of equilibrium established between the ions and undissociated molecules HX and H^+X^- as follows:

$$K_{HX} = \frac{[H^+][X^-]}{[HX] + [H^+X^-]}$$

or

$$K_{HX} = \frac{K_i\,K_d}{1 + K_i}. \qquad (8)$$

Eqn. (8) is a particular case of Eqn. (5) with the condition $K^*_{inst} \ll 1$. Substituting activities for equilibrium concentrations in Kolthoff's equation (8) and expressing the equilibrium constants by Izmailov's method, we obtain

$$K_{ord} = \frac{K_{conv}K^{-1}_{ass}}{1 + K_{conv}} = \frac{K_{diss}}{1 + K_{conv}},$$

since

$$K_i = K_{conv};\ K_d = K^{-1}_{ass}.$$

Thus Kolthoff's equation is a particular case of Izmailov's equation.

Analogously it can be shown that the equilibrium schemes and equations of other authors (Ref. 26, p. 603) (Berg and Patterson[142], Smith and Elliot[143], Halban[144], Correy[145], Sadek and Fuoss[146], and others) are particular cases of the general equilibrium scheme put forward by Izmailov:

$$KA + (n+m)\,M \overset{K^*_{inst}}{\rightleftarrows} KAM_n + m\,M \overset{K_{diss}}{\rightleftarrows} K^+(solv.) + A^-(solv.),$$

$$\uparrow\downarrow K^{-1}_{ass}$$

$$x\,M \overset{K_{conv}}{\rightleftarrows} K^+(solv.)\,A^-(solv.) + y\,M.$$

Izmailov's theory is applicable to acids, bases, and salts in non-aqueous solutions and has been described in detail[20,26,147-150].

The modern theory of non-aqueous solutions, according to which the same substance, under the influence of the solvent, may be a strong or weak electrolyte and alter its acidity, basicity, solubility, and other properties, is derived from the above ideas and is based on a quantitative consideration of the affinity between the ions of the dissociating substance *in vacuo* and the energy of their solvation and of the solvation of the solvent molecules.

The dependence of the strength of the electrolyte on the properties of the substance and the solvent is expressed quantitatively by the equation

$$K_{ord} = K_{exp}\frac{\Sigma A_x - A_{mol}}{RT}, \qquad \dagger\ (9)$$

where K is the dissociation constant of the electrolyte *in vacuo*, ΣA_x the total energy of solvation of the ions, and A_{mol} the energy of solvation of the molecules.

By making a detailed analysis of Enq. (9) and introducing the theory developed by Bernal and Fowler[151] and Mishchenko and Sukhotin[152] to define the energy of solvation of ions, Izmailov deduces particular cases[20] of the general equation (9) for salts, acids, and bases and the ionic product of the solvent. From the equations which he gives it follows that:

1. The strength of any acid increases with increase in the basicity of the solvent and the energy of solvation of the lyonium and lyate ions; this energy increases with increase in the dielectric constant of the solvent and the dipole moment of its molecules. The solvent influences not only the absolute strength of acids but also their relative strengths. A differentiating action with respect to acids is particularly characteristic of solvents with a low dielectric constant (Table 1).

2. The strength of any base increases with increase in the acidity of the solvent and the energy of solvation of the

† It appears that this equation should read $K_{ord} = K\exp\left(\frac{\Sigma A_x - A_{mol}}{RT}\right)$ (Ed. of Translation).

TABLE 1. Dependence of the strength of hydrochloric and benzoic acids on the nature of the solvent.

Solvent	Dielectric constant	The logarithm of the reciprocal of the autoprotolysis constant pK_i^*	$pK_{ord(b)}$ (benzoic acid)	$-\log \frac{K_i^*}{K_{ord(b)}}$	$pK_{ord(h)}$ (hydrochloric acid)	$-\log \frac{K_i^*}{K_{ord(h)}}$
Water	78.5	14.0	4.02	9.89	0.80	13.2
Liquid ammonia	21.0	33.7	3.82	29.88	2.90	30.8
50% acetone	24.0	20.3	9.80	10.50	2.47	17.83
Acetone	19.1	—	11.95	—	4.00	—
Ethanol	24.0	19.1	10.13	9.13	1.95	17.35
Anhydrous acetic acid	6.1	14.5	—	—	9.10	5.35

Note. $K_{ord(b)}$ and $K_{ord(h)}$ are the dissociation constants of benzoic and hydrochloric acids respectively.

ions. A differentiating action with respect to bases is also particularly characteristic of solvents with a low dielectric constant (Table 2).

Titration of acids. The influence of different solvents alters not only the absolute strength of acids, but also their relative strengths. This applies particularly to acids of different character, soluble in solvents of low dielectric constant [153-187]. The accuracy of the titration of individual acids and bases is determined by the ratio of the autoprotolysis constant K_i of the solvent to the dissociation constant K_{ord} of the acid or base being titrated, K_i/K_{ord}

The accuracy of the differentiating titration of acid or base mixtures is determined by the ratio of the strengths of the individual components of the mixtures being titrated:

$$K_{ord(II)}/K_{ord(I)}. \qquad (10)$$

The differentiating action of acid solvents on inorganic acids is shown only when the solvent has a low dielectric constant [188]. In acid solvents it is possible to determine separately the concentration of strong acids in mixtures with organic (weak) acids [189]. Even the strongest organic acids are not titrated in protogenic solvents, since the pK values of organic acids are numerically much greater than 10. For example, pK for trichloroacetic acid — the strongest organic acid — is 11.64 (Table 3) [13]. The strongest acid in acetic and formic acids is perchloric acid (Table 3), so that best results in protogenic solvents are given by perchloric acid as titrant.

TABLE 2.

Base	H_2O, $pK_i^* = 14$		CH_3COOH, $pK_i^* = 14.45$		HCOOH, $pK_i^* = 5.7$	
	$pK_{ord(base)}$	$-\log \frac{K_i^*}{K_{ord(base)}}$	$pK_{ord(base)}$	$-\log \frac{K_i^*}{K_{ord(base)}}$	$pK_{ord(base)}$	$-\log \frac{K_i^*}{K_{ord(base)}}$
Diethylamine	2.9	11.1	—	—	0.74	5.0
Urea	13.82	0.18	10.24	4.2	1.25	4.50
Pyridine	8.69	5.31	6.1	8.35	0.95	5.3
Benzidine	9.03	3.97	—	—	0.75	5 0
Aniline	9.4	4.6	—	—	0.44	5.31
α-Naphthylamine	10.8	3.2	—	—	0.88	4.89
Triphenylamine	—	—	5.38	9.07	—	—

Note. $K_{ord(base)}$ is the dissociation constant of the base.

TABLE 3. pK values for some strong acids in different solvents.

Acid	Solvent		
	H_2O	CH_3COOH	HCOOH
Perchloric	—	5.80	0.28
Hydochloric	—	8.85	0.89
Sulphuric	1.92(K_2)	8.20	0.58
Trichloroacetic	0.89	11.64	—

In the titration of acid mixtures, in addition to the value of the ratio $K_{ord(II)}/K_{ord(I)}$, the ratio of their initial concentrations C_1 and C_2 is of great importance. According to Komar', [190] the distribution of alkali between two acids present in a titration mixture may be expressed by the equation

$$\frac{K_1}{K_2} = \frac{\alpha}{(1-\alpha)^2}\left[\frac{C_2}{C_1} - (1-\alpha)\right],$$

where K_1 and K_2 are the dissociation constants of the acids, and α the distribution coefficient of the alkali between the acids. It is possible by solving this equation, to calculate theoretically the ratio C_2/C_1 at which differentiating titration of the acids is possible.

For the accurate separate titration of a mixture of acids when $C_1/C_2 = 0.01$, it is necessary that $K_1/K_2 \geqslant 10^6$, but the ratios of the constants of, for example, sulphuric and lactic acids and sulphuric and citric acids in water are, respectively:

$$\frac{K_{sulphuric}}{K_{lactic}} = \frac{1.2 \times 10^{-2}}{1.4 \times 10^{-4}} = 85.7,$$

$$\frac{K_{sulphuric}}{K_{citric}} = \frac{1.2 \times 10^{-2}}{8.7 \times 10^{-4}} = 13.8.$$

These data show that for this ratio of the constant the separate titration of these acids in aqueous medium is impossible with $C_1/C_2 = 0.01$. This applies to an even greater extent at less favourable ratios of C_1 and C_2, *i.e.* when the concentration of the weaker acid is much greater than that of the stronger acid. Such titrations are possible, however, for mixtures of strong and weak acids in non-aqueous solutions [191].

The use of protogenic solvents in analytical chemistry has extensive possibilities in the analysis of individual acids and bases and their mixtures, including acids and bases whose pK values are > 10.

The processes taking place in the titration of weak acids by bases in protophilic solvents can be represented by the following equations:

$$HCON(CH_3)_2 + HX \rightleftarrows [HC\overset{+}{O}NH(CH_3)_2]^+ + X^-,$$

$$(C_2H_5)_4NOH \rightleftarrows [(C_2H_5)_4N]^+ + OH^-,$$

$$[HC\overset{+}{O}NH(CH_3)_2]^+ + OH^- \rightarrow HCON(CH_3)_2 + H_2O,$$

$$\overline{(C_2H_5)_4NOH + HX \rightarrow (C_2H_5)_4NX + H_2O}.$$

Titration of bases. The strength of bases increases with increase in the acidity of the solvents and the energy of solvation of the ions. A differentiating action

TABLE 4. pK values for some bases in different solvents.

Base	Solvent		
	H_2O	CH_3COOH	HCOOH
Pyridine	8.69	5.96	0.45
α-Naphthylamine	10.08	6.40	0.88
Caffeine	13.39	—	0.78
Urea	13.82	9.8	1.25

with respect to bases is observed particularly for solvents with a low dielectric constant and low acidity [192-200].

In protogenic solvents (for example CH_3COOH) even weak bases with p$K > 10$ are titrated as strong bases. Since the value of $K_{i(CH_3COOH)}$ (2.5×10^{-13}) [*cf.* Table 2, 3.5×10^{-15}? (Ed. of Translation)] is close to that of $K_{i(H_2O)}$, the possibility of titrating bases results from the marked increase in the dissociation constant of bases in protogenic solvents.

In acid solvents reactions leading to the formation of the ions of the base proceed nearly to completion, the degree of completeness being greater, the greater the protogenic properties and dielectric constant of the solvent. For example, in formic acid, which has more marked protogenic properties and a higher dielectric constant than acetic acid, electrolytes exhibit a more marked basic character. Table 4 gives pK values for some bases in acetic and formic acids; data for aqueous solutions are also given for comparison [13]. Table 4 shows that some very weak bases, which cannot be titrated in water or in anhydrous acetic acid, can be titrated quantitatively as strong bases in a solution of anhydrous formic acid.

Acidic solvents, due to their pronounced protogenic properties, have a levelling effect on the strength of bases; this property is particularly characteristic of formic acid, first studied in this connection by Hammett and Dietz [201] and studied in detail later by Shkodin and others [13,130,202-204]. One advantage of formic acid as solvent is that in this acid it is possible to titrate extremely weak bases which cannot be titrated in acetic acid [202,203].

The processes taking place during the titration of weak bases, in protogenic solvents, by perchloric acid dissolved in anhydrous acetic acid, can be represented by the following equations:

$$C_5H_5N + HCOOH \rightleftarrows C_5H_5NH^+ + HCOO^-,$$
$$HClO_4 + CH_3OOH \rightleftarrows CH_3COOH_2^+ + ClO_4^-,$$
$$CH_3COOH_2^+ + HCOO^- \rightleftarrows CH_3COOH + HCOOH,$$
$$C_5H_5NH^+ + ClO_4^- \rightleftarrows C_5H_5NHClO_4,$$
$$\overline{C_5H_5N + HClO_4 \rightarrow C_5H_5NHClO_4}.$$

The fundamental theory of acid-base titrations in non-aqueous media has been described in a number of papers [205-236].

The above review shows the vast possibilities of non-aqueous solutions in analytical chemistry, which are assisting in the development of new, highly-sensitive, rapid, and accurate methods of analysis.

III. SOLVENTS USED IN NON-AQUEOUS TITRATION

1. Solvents for Compounds Titrated as Bases

Conant *et al.* [237-239] first used anhydrous acetic acid as solvent for the titration of weak bases with perchloric acid. Marcunas and Riddick [240,241] showed that many amines, salts of organic acids, choline salts of carboxylic acids, salts of organic bases, oxazolines, amino-alcohols, etc. can be determined in acetic acid. Salts without basic properties are converted to acetates, which can be titrated as bases [242-244]. Titration in anhydrous acetic acid is now widely used to determine a variety of organic substances [245-267]. Pifer *et al.* [268] and Poulos [269] established that the addition of hydrocarbons or their halogeno-derivatives, dioxane, and other solvents to acetic acid facilitates the dissolution of organic compounds and makes the end-point of the titration sharper. Mixtures of acetic acid with chloroform [270,271], benzene [272], nitromethane [256], nitrobenzene [273], and acetonitrile [256], have been used as solvents.

Titration in acetic acid has a number of disadvantages however [274]. Pifer and Wollish's studies [275] showed that the water content of the acetic acid should not exceed 1%. The addition of acetic anhydride eliminates the influence of water and increases the potential jump. Acetic acid can also be dehydrated with triacetyl borate [276]. A number of authors [277,278] have shown that complete removal of water from the acetic acid makes the end-point of the titration much sharper. Many weak bases, which cannot be titrated in acetic acid, can be titrated in a mixture of acetic acid and acetic anhydride [250,279,280]. Some authors have used pure acetic anhydride instead of acetic acid and its mixtures with acetic anhydride [271,279-283]. A method has been developed for the micro- and ultramicro-titration of organic compounds in acetic acid [259,260,271].

Hammett [201,284] and others [285] found that formic acid can also be used for titrating very weak bases: aniline, urea, etc., and the acid has been used for this purpose by Shkodin, Izmailov, Dzyuba, and others [202,203,286]. In formic acid, which is a more acidic solvent than acetic acid, it is possible to titrate weak bases which cannot be titrated in acetic acid. Mixtures of formic acid with acetic acid [285], nitromethane [287], and dioxane [288] have also been used for titrations. The addition of dioxane improves the conductometric titration of weak bases.

A number of workers have titrated bases in propionic [289-294], butyric [295], chloroacetic [188,296,297], trichloroacetic [188], and trifluoroacetic [298,299] acids.

Indian workers [7,300-308] have used glycol solvents. Glycols have also been used as solvents for the titration of bases by other workers [309-311].

Vorländer and others have titrated bases in benzene, chloroform, and carbon tetrachloride [263,269,312-319]. Mixtures of benzene with dioxane have been used as solvents, as well as cyclohexane, ethyl acetate, chlorobenzene, n-heptane, nitrobenzene, nitromethane, ethyl ether, petroleum ether, and other solvents [200,256,263,277,319-325].

Fritz [326] found that anhydrous dioxane is a good solvent for the titration of bases, and this was confirmed by other authors [268,269,327]. Mixtures of dioxane, benzene, and toluene with acetic anhydride have also been used as media for the titration of bases [328].

Solvents with strong acidic properties cannot be used for the differentiating titration of mixtures of bases, due to the levelling action of these solvents on the bases. Solvents with neutral or amphoteric properties have no levelling action on bases, and can be used for the differentiating titration of mixtures of bases which cannot be titrated in water and acidic solvents because their dissociation constants are too similar. Fritz[329] and others[256,321] used acetonitrile as solvent for the differentiating titration of amine mixtures. Chatten[330] titrated bases in a phenol–chloroform–acetonitrile medium.

Acetone[331-333] and methyl isobutyl ketone[334] have been used for the differentiating titration of mixtures of organic bases.

We have studied the differentiating action of acetone, methyl ethyl ketone (MEK), and methyl butyl ketone (MBK) in the titration of mixtures of organic bases[335], and found that MEK is one of the best differentiating solvents. The differentiating titration of multicomponent mixtures of organic bases is possible in MEK.[336,337]

Pyridine[338] and other solvents[323,339-341] have also been used for the titration of organic bases.

2. Solvents for Compounds Titrated as Acids

Inert solvents such as benzene, chloroform, toluene, carbon tetrachloride, and their mixtures were used initially for the titration of acids[342-347], and are still receiving attention[348,349].

Fairly sharp end-points are observed in the titration of acids in alcohols[237]. Acids can be titrated in methyl, ethyl, propyl, isopropyl, butyl, pentyl, isopentyl, benzyl, and other alcohols, and also in mixtures of alcohols with benzene, chloroform, carbon tetrachloride, dioxane, anisole, acetone, and pyridine[350-389]. In 1951 Fritz[390] introduced benzene–methanol mixtures for non-aqueous titration, and these mixtures have since been used by other workers[391-398].

Of the basic solvents, liquid ammonia has the greatest solvent power for organic substances. Reviews on acid–base titration in liquid ammonia have appeared[9,131]. Methods of titration in liquid ammonia have also been described[399,400], but these have limited applications.

Moss *et al.*[401] used anhydrous ethylenediamine (EDA) for the titration of phenols and other weak acids. In EDA it is possible to titrate certain nitro-compounds, which behave as acids[402]. Other authors have also titrated weak acids in EDA.[383,403-411] Many acidic substances have been titrated in butylamine[390,396,404,412,413] and also in ethylamine, diethylamine, and triethylamine[414].

Pyridine[375,391,412,415-424] and piperidine[334] are often used as a medium for the titration of different acidic substances. Marcunas and Cundiff[389,425] found that pyridine is the most suitable solvent for the titration of mixtures of strong acids.

Fritz[404,426,427] suggested the use of dimethylformamide (DMF) as solvent for the titration of acids. Very many compounds of acidic character can be titrated in this medium[408,418,424,428-435]. Diethylamine, diethylformamide, and other amides have also been used for the titration of acidic substances[433,436]. Shkodin and others have used acetic and formic acids as solvents for the titration of mixtures of strong acids[2,191,204,437-439]. Acids have also been titrated in butyric, monochloroacetic, and trichloroacetic acids[130,188,295].

Acids have been titrated in mixtures of ethylene glycol with isopropyl alcohol, acetone, and chloroform[301,440-442], and also in ethers[443,444], dioxane[445-447], and acetonitrile[347,389,448-450].

A number of ketones have been used for the titration of acidic substances. Cray[451] first carried out titrations in acetone. Izmailov and others made use of the differentiating action of ketones (acetone, MEK, methyl propyl ketone) for the titration of acid mixtures[376]. Other authors have also titrated acids in ketones[423,452-457]. Alcohols, ethers, hydrocarbons, and other solvents have been added to acetone to facilitate the dissolution of organic acids[458]. Bruss and Wyld[334] suggested the use of methyl isobutyl ketone for the titration of mixtures of strong, weak, and very weak acids. Cundiff and Marcunas[389,425] showed that methyl isobutyl ketone cannot be used for the titration of mixtures containing strong acids.

We have titrated various organic and inorganic acids in MEK.[459]. It was established that MEK exhibits good differentiating properties with respect to both strong and weak acid mixtures. In this respect MEK is superior to acetone and MBK. Strong acid mixtures cannot be titrated in acetone, and in MBK the salt formed during the titration is often precipitated[460]. Compounds of acidic character have also been titrated in various other solvents[436,453,461-463]

IV. TITRANTS

Titrants of acidic character. A solution of hydrogen chloride in benzene was first used for the titration of bases in non-aqueous media[464]. Subsequently it was established that $HClO_4$ is the strongest acid in non-aqueous solvents: $K_{HClO_4} = 1.6 \times 10^{-4}$ and $K_{HCl} = 1.4 \times 10^{-7}$.[465] [*cf.* Table 3, (Ed. of Translation)] Solutions of perchloric acid in acetic acid are now chiefly used for the titration of bases[253-256,466-479]. Solutions of hydrogen chloride in organic solvents have also been used[249,480-482]. For titration in glycols, glycol solutions of $HClO_4$ and HCl are employed[7,308,309].

Pifer and Wollish[275] found that the end-point is sharper when solutions of $HClO_4$ in dioxane are used. Solutions of $HClO_4$ in dioxane are often used for the titration of bases in inert and differentiating solvents[321,327,329,483]. Solutions of $HClO_4$ are also prepared in acetonitrile[329], nitromethane[484], dioxane–formic acid mixtures[288], and propionic acid[293,294].

We have successfully used a solution of $HClO_4$ in MEK.[336,337] Titration in MEK with a solution of $HClO_4$ in the same solvent gives sharper end-points than $HClO_4$ solutions in anhydrous acetic acid and dioxane.

Udovenko and Vvedenskaya[332] used solutions of H_2SO_4, HNO_3, and β-naphthalenesulphonic acid for titrations in acetone; H_2SO_4 and HNO_3 solutions have also been used by other workers[485,486].

Sulphonic acids have been widely used for the titration of bases, *e.g.* *p*-toluenesulphonic acid[321,487-489]. Hall[321] used perfluorobutyric and methanesulphonic acids. In non-aqueous solvents, the strength of benzenesulphonic acid is comparable to that of sulphuric acid[490]. The strengths of methanesulphonic, ethanesulphonic, benzenesulphonic, and naphthalene-2-sulphonic acids in acetic acid are lower than that of perchloric acid, but the sulphonic acids do not form precipitates and gels during the titration[491,492]. Solutions of diphenyl phosphate and periodic and salicylic acids have also been used as titrants[311,489,493].

Solutions of perchloric acid and other acids in non-aqueous media are usually standardised with sodium carbonate[468,471,472], diphenylguanidine[326,496], potassium hydrogen phthalate[241,497,498], 4-aminopyridine[499], and potassium tetraborate[500].

Titrants of basic character. Potassium and sodium methoxides and ethoxides, and potassium, sodium, and lithium butoxides and amyloxides [342-345,387] were used initially for the titration of acids in non-aqueous media. Many workers have used these titrants[391-393,436,501]. Rubidium and caesium methoxides have also been used for the high-frequency titration of acids[502].

Stronger basic titrants include sodium aminoethoxide in EDA,[401,411] sodium triphenylmethane in an ether–benzene mixture[503], and lithium aluminium hydride and lithium aluminium amide in tetrahydrofuran[504-507]. A solution of sodamide in liquid ammonia has been used as titrant in this solvent[399]. Acids have also been titrated with solutions of pyridine, piperidine, and aniline[204,415,439,440,508], various amines, morpholine, nitron, and dimethylaminoantipyrine[375,402,415,434,441,450,508], alcoholic or ethereal alkali solutions[136,372,384,421,440,441,509], and sodium, potassium, and lithium acetates in anhydrous acetic acid[188,189,191,279,437,441,510,511].

Deal and Wyld[408] titrated mixtures of strong and weak acids with a solution of tetrabutylammonium hydroxide (TBAH) in aqueous isopropyl alcohol. Harlow *et al.*[407] used TBAH dissolved in absolute isopropyl alcohol. Cundiff and Marcunas[418] used a benzene–methanol solution of TBAH. Quaternary ammonium hydroxides have been widely used in analysis[334,374,375,391,425,452,454,456,458,512,513].

We have used a benzene–methanol solution of tetraethylammonium hydroxide (TEAH) for the titration of multicomponent mixtures of compounds exhibiting acidic character in non-aqueous media[459,460]. The titration curves show sharp end-points. Salts of TEAH and inorganic and organic acids are readily soluble in organic solvents.

Benzoic acid is generally used to standardise solutions of the above titrants[7,17,459]. Sulphamic acid has been used to standardise CH_3OLi solutions[514].

V. METHODS OF TITRATING BASIC SUBSTANCES IN NON-AQUEOUS MEDIA

1. Visual and Potentiometric Methods

Primary, secondary, and tertiary aliphatic and aromatic amines. The majority of organic bases can be titrated in non-aqueous solvents. Primary, secondary, and tertiary amines are titrated in anhydrous acetic acid[240,263,468,471,515-517], propionic acid[518], glycols[7,309], and other solvents[263,329,519]. Hall[321] titrated about 70 individual mono- and di-amines in non-aqueous solvents. Micro-[520] and ultramicro-methods[260] have also been used for the determination of amines.

Bloomrich and Bandel[468] developed a method for determining tertiary amines in mixtures with primary and secondary amines by titration in an acetic acid–acetic anhydride medium. Wagner *et al.* extended this method to the determination of mixtures of primary, secondary, and tertiary aliphatic amines[521,522]. The total amine content is first determined by titration. A known quantity of the specimen being studied is then treated with acetic anhydride, after which the tertiary amine is titrated. A third titration gives the sum of the secondary and tertiary amines after treatment of the mixture with salicylaldehyde, which converts the primary amines to Schiff's bases. The glycol titration method has also been used for the quantitative determination of mixtures of primary, secondary, and tertiary aromatic amines[310].

Johnson and Funk[523] have described a modification of the method of Wagner *et al.* Salicylaldehyde in pyridine is added to the amine mixture, after which the excess salicylaldehyde is titrated with sodium methoxide. The primary amine content is found by difference. Critchfield and Johnson[327], in the determination of mixtures of primary, secondary, and tertiary amines, converted the primary amines to the weakly basic imine, by treating the amine mixture with salicylaldehyde, and determined the total content of secondary and tertiary amines. Dioxane was then added to the mixture and the imine was titrated. Primary aliphatic amines have also been determined by titration of the excess 2,4-pentadione which had not reacted with the amine[524].

Kubias[481] developed a method for the analysis of mono-, di-, and tri-ethanolamines. The triethanolamine content was determined by titrating the mixture after treatment with acetic anhydride. The mono- and di-ethanolamines were determined iodometrically, by oxidising them with periodate and titrating the liberated iodine with Na_3AsO_3 solution. Gal'pern and Bezinger[525] developed a method for analysing a mixture of primary, secondary, and tertiary aliphatic amines by titration in glacial acetic acid. The method is based on the quantitative reaction of primary amines with phthalic anhydride, and of the primary and secondary amines with acetic anhydride.

Many Schiff's bases have been titrated in acetic acid[526]. The differentiating titration of amines and Schiff's bases is possible in chloroform and acetonitrile. Primary amides of the sulphonamide series can be titrated in acetic acid[471].

Organic bases containing heterocyclic nitrogen. Many organic bases containing heterocyclic nitrogen have been titrated in non-aqueous solvents. These bases include purines, pyridine, pyridone, thiazole, pyrazolones, quinoline, hydrazones, hydrazides, oxazolines, their derivatives, 5-acetamidomethyl-4-amino-2-methylpyrimidine, aminopyrine, and other compounds[240,241,249,253,277,278,473,476,527-534].

Mizukami and Hirai[535] have developed a method for the separate determination of the hydrazide of isonicotinic acid and sulphoisoxazole. The mixture of these compounds in acetic acid is treated with acetic anhydride; the acetate of the isonicotinic acid hydrazide dissolves in the acid and is titrated with perchloric acid in a mixture of acetic acid and acetic anhydride.

Lieber *et al.*[477] have developed a method for determining the acidic and basic forms of substituted 5-amino-1,2,3-triazoles. The acidic forms are determined by titrating with a benzene-methanol solution of sodium methoxide in dimethylformamide. The basic forms dissolved in acetic acid are titrated with $HClO_4$ solution. The method is applicable to the separate determination of the acidic and basic forms in a mixture.

Hennart and Merlin[294] have titrated benzoimidazole and ethyl-2-benzoimidazole with an alkyl group in the 2- or 5-position, in anhydrous propionic acid.

Alkaloids. A number of authors have used non-aqueous solvents for the titration of many alkaloids which are weak bases: theobromine, caffeine, quinine, brucine, and others[247,262,263,473,536,537]. The titration of the alkaloids is carried out in acetic acid, and also in chloroform, carbon tetrachloride, and their mixtures[268,312-317,538]. The glycol titration method has also been applied to the analysis of strychnine, brucine, narcotine, narceine, papaverine, thebaine, and others[7,301,307,539].

Other organic bases. The nitro-derivatives of aromatic amines exhibit very weak basic properties, even in strongly protogenic solvents. In pyridine these compounds exhibit acidic properties and can be titrated quantitatively with a benzene–methanol solution of triethyl-n-butylammonium hydroxide. Fritz[338] used this method to determine the nitro-derivatives of amines, substituted in the 2,4- or 2,4,6- [2,6- or 4,6? (Ed. of Translation)] positions with two NO_2 groups or one NO_2 group and one or several Cl atoms, diphenylamine derivatives containing NO_2 in position 4, and also trinitrotoluene and trinitrobenzene. Quantitative determination of certain mixtures of nitro-compounds is also possible.

Wimer[282] developed a potentiometric method for titrating different substituted and unsubstituted amides in acetic anhydride. Under these conditions the sparingly soluble diamides of dibasic acids (with the exception of malonamides and tetra-substituted phenylamides), *N*-phenyl- and α-phenyl-substituted amides, and unsaturated amides, with a double bond conjugated with the carbonyl group, are not titrated. The method is applicable to the determination of amide–amine mixtures. The authors also titrated aliphatic, aromatic, and heterocyclic sulphoxides in acetic anhydride.

Certain amides have been titrated in acetic acid[257,540].

Salts of organic acids or bases. Many alkali metal salts of carboxylic acids are readily soluble in acetic acid and can be titrated as bases[240,241,247-249,268,471,541]. The addition of acetic anhydride increases the accuracy of the determination. Titration of salts is also possible in propionic acid[518]. The glycol titration method is applicable to the determination of a large number of salts of monobasic acids and also to the determination of the acetates of all the alkali and alkaline earth metals and a number of other metals[7,301,302,308].

Salts of organic acids and bases are titrated in formic acid[201,203], acetone[331,465,542], butanol[543], and methanol[544]. The differentiating titration of salt mixtures is also possible[545].

Pifer and Wollish[243,275] have developed a method for converting the salts of hydrohalic acids to the acetates, which are titrated in acetic acid as bases. This method is based on the reaction of the salts with mercuric acetate, according to the equation

$$2KA + Hg(CH_3COO)_2 \rightarrow 2CH_3COOK + HgA_2.$$

Pifer and Wollish[242] have titrated a large number of salts of hydrohalic acids, heterocyclic compounds, vitamins, and quaternary ammonium salts. This method has also been used by other authors for the determination of the hydrochlorides of various amines, the hydrohalides of antihistamine bases, certain narcotic and antibiotic salts, and halide complexes[278,546-548]. Anastasi[252] has determined proguanide (paludrine hydrochloride) by potentiometric titration in an acetic acid–acetic anhydride medium in the presence of mercuric acetate. Gyenes[475] has determined salts of tertiary amines in the presence of salts of acetylated amines with the aid of an acetylating mixture of acetic anhydride and mercuric acetate, dissolved in acetic acid.

Bayer[549] has used mercuric acetate for the analysis of organic bases containing sulphur. The method was tested for the determination of $CS(NH_2)_2$ and thiosinamine. Hennart and Merlin[293] determined amine halides by titration in propionic acid. The amine halides are dissolved in propionic acid, mercuric propionate solution is added, and the liberated propionate is titrated with perchloric acid.

Belcher and Berger[259,260] developed a method for the ultramicrotitration of the sodium salts of carboxylic acids, amine hydrohalides, and quaternary ammonium salts. A method has been developed for determining the sparingly soluble salts of phenothiazine bases in acetic acid, acetic anhydride, acetone, nitrobenzene, and acetonitrile[283]. Best results are obtained by titration in acetone and acetonitrile. Salts have also been titrated in non-aqueous solvents by other workers[281,550-555].

Inorganic salts. Many inorganic salts can be titrated in acetic and propionic acids[208,293]. Pifer and Wollish[243] have applied their method for the analysis of organic bases in acetic acid to the analysis of inorganic salts. Many salts of Na, Al, Co, Fe, Pb, Mg, Mn, K, Ag, Sr, Zn, and other metals have been titrated by this method.

Salts of many metals have been determined in glycols[7]. The metals are precipitated as the oxides, hydroxides, carbonates, or salts of a weak organic acid, after which the precipitate is dissolved in acetic acid. The solution is evaporated and the acetate dissolved in the glycol solvent and titrated.

Differentiating titration of bases. Fritz[329] carried out differentiating titration of mixtures of aliphatic and aromatic amines in acetonitrile, which has no levelling action on bases. He carried out differentiating titration of mixtures of di-n-butylamine + pyridine, aniline + *o*-chloroaniline, β-phenylethylamine + aniline, pyridine + caffeine, and aniline + sulphothiazole. The presence of small quantities of water in the solvent decreases considerably the sharpness of the end-point. Schiff's bases and the original amines can be titrated separately in acetonitrile[526].

Pifer *et al.*[556] have carried out differentiating titration of potassium and ammonium acetates, in a mixture with the acetates of a number of other metals, in an acetic acid–chloroform medium. These authors established that differentiating titration of mixtures of potassium and ammonium acetates with n-butylamine or pyridine is possible. Certain alkaloid mixtures, for example codeine and caffeine, have been determined in nitromethane[484]. Differentiating titration of mixtures of bases has also been carried out in ethylene glycol–phenol, methyl isobutyl ketone, and benzene–methanol[334,557,558].

We have carried out differentiating titration of two-, three-, and four-component mixtures of organic bases in MEK:[335-337] tetraethylammonium hydroxide + piperidine, piperidine + acridine, tetraethylammonium hydroxide + triethylamine + pyridine, tetraethylammonium hydroxide + + triethylamine + *m*-tolylenediamine, and tetraethylammonium hydroxide + triethylamine + pyridine + *m*-tolylenediamine.

Acids titrated as bases. Some organic acids exhibit basic properties in acidic solvents. Thus pyridinecarboxylic acids and their derivatives are titrated as bases

in acetic acid[249,470]. A number of amino-acids also exhibit basic properties in acetic acid[246,486,559-561]. The back--titration method gives best results in certain cases, however. The amino-acids are treated with an acetic acid solution of perchloric acid and the excess of the latter is titrated with an acetic acid solution of guanidine acetate or sodium acetate[3,245]. The basic groups of amino-acids have also been titrated in ethyl alcohol[364,365].

Some amino-acids also exhibit basic properties in glycols[7,307]. These include glycine, asparagine, aminosalicylic acid, *p*- and *o*-aminophenols, and others. Anthranilic and hippuric acids exhibit acidic properties in glycol solvents and cannot be titrated as bases.

2. Conductometric Method

The conductometric method has been used for the titration of substances of basic character in non-aqueous media[320,346,562]. Udovenko and Vvedenskaya[332,492] have carried out the conductometric titration of anabasine, lupine, and other alkaloids. McCurdy *et al.*[288] have described a method for the conductometric titration of weak organic bases in a dioxane–formic acid medium. The titration of binary mixtures of bases has also been carried out.

Van Meurs and van Dahmen[322] have studied the influence of the composition, solvating power, and dielectric constant of the solvents, the temperature of the solution, the anion of the titrant (for example $HClO_4$ and HCl), the number of NH_2 groups in the molecules of nitrogen-containing bases, and the distance between them on the results of the conductometric titration of mono- and di-acidic bases and their mixtures in non-aqueous media.

Amino-acids have been titrated as bases by a conductometric method in trifluoroacetic acid[299].

3. High-Frequency Method

Many organic bases which can be titrated in non-aqueous media by visual, potentiometric, and conductometric methods, can be titrated successfully by the high-frequency method. Thus Wagner and Kauffman[563] have titrated aniline, *p*-toluidine, pyridine, and other bases in acetic acid. The high-frequency and potentiometric curves showed good agreement. Sharp end-points could not be obtained for *p*-nitroaniline (K_{H_2O} 1×10^{-12}), *o*-nitroaniline (K_{H_2O} $1.5 \times \times 10^{-14}$), and urea ($K_{H_2O}$ 1.5×10^{-14}).

Ishidate and Masui[564] have titrated a number of alkaloids and weak organic bases in aqueous alcohol solutions. They established that in acetic acid the method is applicable to the analysis of bases with K_{diss} greater than 10^{-9} and alkaloids with $K = 10^{-10}$.[565] Lippincott and Timnick[566] have titrated aniline, its derivatives, and amino-acids in acetic acid. The differentiating titration of two-component mixtures was achieved.

Riolo *et al.*[567] have developed a high-frequency method for the titration of phenazine, hydrooxyphenazine, benzo[*a*]-phenazine, and benzo[*a*]phenazine-1,2-*N*-monoxide in acetic acid. In mixtures of acetic acid and dioxane the authors were able to carry out differentiating titration of mixtures of triethanolamine with quinoline or pyridine, and of triethanolamine in the presence of quinaldine, acridine, and 2-picoline[568]. The high-frequency method in acetic acid as solvent has been used for the analysis of phenylhydrazine, brucine, phenanthroline, *p*-toluidine, and a number of amines. Compounds containing imino- and hydroxyamino--groups, and urea and thiourea could not be titrated[420].

4. Photometric method

The photometric method has been used for the titration of weak organic bases in acetic acid[474,569]. Later Connors and Higuchi[258] put forward an improved method for the titration of weak bases in acetic acid. The titration of acetamide, urea, dimethylpyrone, thiourea, caffeine, antipyrene, and triphenylguanidine has been studied. The quantitative determination of the individual compounds and mixtures (antipyrene + triphenylguanidine, urea + antipyrene) has been carried out.

Hummelstedt and Hume[570] have carried out photometric titration of weak bases in acetic acid and acetonitrile. Two- and three-component mixtures have been titrated in acetic acid. In acetonitrile it is possible to titrate even four-component mixtures (di-n-butylamine + *N,N*-diethylaniline + aniline + *o*-chloroaniline). It has been established that differentiating photometric titration is possible if the p*K* values of the bases differ by 1.5 or more. The photometric method has proved very successful for the differentiating determination of mixtures of components with similar basic strengths.

VI. METHODS OF TITRATING ACIDIC SUBSTANCES IN NON-AQUEOUS MEDIA

The first analytical determination of the higher aliphatic acids in inert solvents was carried out by Folin *et al.*[342-345]. The practical application of non-aqueous solvents for the titration of weak acids began with the studies of Moss *et al.*[401], who used an EDA medium for the titration of phenols, carboxylic acids, and their derivatives, which cannot be titrated in aqueous media.

1. Visual and Potentiometric Methods

Carboxylic acids. Many workers have titrated mono-, di-, and tri-basic carboxylic acids and their derivatives in non-aqueous media[390,395,400,571,572]. Deal and Wyld[408] have titrated *o*-, *m*-, and *p*-phthalic acids in dimethylformamide. Cundiff and Marcunas[418] have titrated hydroxybenzoic acids in pyridine. They found that *m*- and *p*-hydroxybenzoic acids behave as dibasic acids, whereas *o*-hydroxybenzoic acid behaves as a monobasic acid. In the titration of maleic, succinic, *o*-phthalic, oxalic, malonic, malic, and fumaric acids in pyridine, they obtained curves with two inflections. The titration curves for ethylenediaminetetraacetic acid in dimethylformamide and of citric acid in pyridine show three inflections. The authors used pyridine as solvent for the titration of a number of amino--acids (alanine, leucine, threonine, asparagine, etc.), and also acetic, benzoic, salicylic, and nicotinic acids.

Harlow *et al.*[349,370] titrated dibasic carboxylic acids in aqueous isopropyl alcohol, benzene, toluene, and gasoline. Fritz and Yamamura[452,453] titrated carboxylic acids in acetone. They found that unsubstituted carboxylic acids have approximately identical strengths, and that the presence of halogens at the carbon atoms, like the presence of one or more nitro-groups in the benzene ring, increases

the strength of aromatic acids. Van Dahmen[573] studied the titration of aliphatic and aromatic carboxylic acids in pyridine and acetone, and established that each carboxyl group in maleic and fumaric acids has a different strength.

We have titrated carboxylic acids in MEK.[459] The titration curves for the dibasic malic, malonic, succinic, phthalic, oxalic, and maleic acids showed two inflections. The tribasic citric acid gives a titration curve with three inflections. Carboxylic acids have also been titrated in non-aqueous solvents by other workers[376,407,416,423,456,458,461].

Phenols and their derivatives have been titrated successfully in EDA and DMF.[401,404] Substituted phenols containing formyl, keto, ether, or nitro groups in the *o*- and *p*-positions are stronger acids than unsubstituted phenols and can be titrated in DMF. Alkyl- and aryl-substituted phenols are titrated in EDA.

Deal and Wyld[408] titrated mono- and di-hydric phenols and polyphenols. The best results were obtained in DMF. Polyphenolic compounds containing two or more phenolic groups and joined by the carbon atoms in the *o*-position (*o*,*o*'-dihydroxydiphenylmethane) are much more strongly acidic than phenol itself. The titration curves for these compounds show only one inflection. The titration curves for compounds joined in the *p*-position (*p*,*p*'-dihydroxydiphenyldimethylmethane) show two inflections.

Cundiff and Marcunas[418] titrated phenol in pyridine, DMF, acetonitrile, and a mixture of benzene and isopropyl alcohol. Di- and tri-hydric phenols were titrated as monobasic acids. Fritz and Yamamura[452,453] titrated 18 phenol derivatives in acetone. The results show that functional groups increase the acidity of phenols in the order:

$$-NO_2 > -C\begin{matrix}O\\H\end{matrix} > -C\begin{matrix}O\\R\end{matrix} > Cl,\ Br,\ I > -C_6H_5.$$

A group in the *o*- or *p*-position increases the acidity of phenol more than one in the *m*-position.

Streuli *et al.*[416,574] studied the acidity of phenols in pyridine and found that phenols and bisphenols have a higher acidity, that of the *o*-compound being greater than that of the *m*- and *p*-compounds. Only one hydroxy-group in the bisphenols is titrated. Alkylphenols and alkylthiophenols have been titrated in pyridine[573]. Phenols can also be titrated in DMF by a micromethod[424].

We have shown that phenol, *o*-, *m*-, and *p*-cresols, resorcinol, catechol, and α- and β-naphthols show fairly strong acidic properties in MEK and can be titrated as monobasic acids[459,575]. A number of other authors have titrated phenols in non-aqueous media[396,407,430,576,577].

Our studies[578,579] of the properties of *o*-, *m*-, and *p*-nitrophenols, 2,4-, 2,5-, and 2,6-dinitrophenols, and picric acid in MEK show that the mononitrophenols are stronger acids than phenol itself. The strength of dinitrophenol is comparable to that of carboxylic acids, and that of trinitrophenol (picric acid) is the same as that of hydrochloric acid. Others have also titrated individual nitrophenols in non-aqueous media[418,452,456,577].

Derivatives of barbituric acid. Titration in non-aqueous media has found wide application in the determination of derivatives of barbituric acid. The titration is carried out in butylamine, pyridine, DMF, and mixtures of benzene with isopropyl alcohol and chloroform[385,412,435,576,580].

Enols, imides, amides, and other compounds. Enols, imides, and a number of amides exhibit acidic properties in non-aqueous solvents, and this makes it possible to titrate them as acids. Fritz[390,427,452,453] titrated many enols and imides in butylamine, benzene–methanol mixtures, EDA, DMF, and acetone. Certain thiols, which exhibit acidic properties in butylamine, have also been titrated. Enols, imides, and thiols are also titrated in pyridine[418].

The SO_2NH group exhibits fairly strong acidic properties in DMF. Weaker sulphonamides are not titrated in DMF, but they can be titrated in butylamine[581]. Sulphonamides can also be titrated in other solvents[393,452,453,576] Acetamide, benzamide, and acylamino-acids can also be titrated in liquid ammonia[399].

Salts of ammonia, aliphatic amines, and other organic bases can be titrated as acids in DMF and EDA. Sharp end-points are obtained[426].

Brockmann and Meyer[402] titrated certain aromatic compounds which behave as acids in EDA. Thus *m*-dinitrobenzene is titrated as a dibasic acid, and picric acid as a tribasic acid. The authors considered that the substances formed during the titration were addition compounds of the bases and nitro-compounds.

A number of esters have been titrated in non-aqueous solvents[405,427,582-584]. The hydrazide of isonicotinic acid, which is titrated as a base in acetic acid, can be titrated as an acid in dimethylaniline[585]. The titration of benzotriazoles in DMF has been described[586].

Even such weakly acidic substances as alcohols can be titrated in non-aqueous solvents[504,505,587], *i.e.* benzene. This method has been used to titrate not only alcohols, but also certain hydrocarbons. It does not, however, distinguish different functional groups [ketones, aldehydes, ethers (esters), lactones, or alcohols].

Deal and Wyld[408] used EDA and DMF for the titration of dialkyl and trialkyl phosphates, which exhibit weak acidic properties in these media. A number of other organic compounds have been titrated as acids in non-aqueous solvents[458,503,588-590].

Acid anhydrides and acid chlorides. A combination of aqueous and non-aqueous titrations has been used to determine mixtures of anhydrides and acids. The total acid and anhydride content was determined by aqueous titration. In alcohol, the acid is titrated as a dibasic acid, and the anhydride reacts with the alcohol to form the ester, which is titrated simultaneously as a monobasic acid[591]. Anhydrides and acid chlorides in benzene–methanol media behave like monobasic carboxylic acids[390].

Siggia *et al.*[592] applied Malm and Nadeau's method for the determination of acetic anhydride in acetylating mixture[593] to mixtures of other carboxylic acids and their anhydrides. The method has been used to determine propionic, maleic, phthalic, camphoric, and butyric anhydrides in the presence of the corresponding acids. Free maleic and phthalic acids and their anhydrides have been determined by titration in acetone and MEK with solutions of tertiary amines, with which the anhydrides do not react[434].

Johnson *et al.*[594] developed a direct method for determining anhydrides in the presence of carboxylic acids by titration with morpholine solution. The anhydrides of *N*-carboxy-α-amino-acids have been titrated in non-aqueous solvents[595,596].

Patchornik and Rogozinski[375] developed an interesting method for determining anhydrides and acid chlorides in the presence of inorganic and organic acids. The inorganic acid is determined by titration in dioxane. The solution is then heated with ethyl alcohol and the acid chloride is determined by titrating the hydrochloric acid liberated. The organic acid content is found by difference after determination of the total acidity. In the analysis of a mixture of anhydride and organic acid the specimen is titrated in dioxane with an alcoholic solution of sodium methoxide the anhydride being titrated as a monobasic acid. Another portion of the solution is treated with pyridine and titrated with trimethylbenzylammonium hydroxide solution in the presence of thymol blue; in this case the anhydride is titrated as a dibasic acid. The results of the two titrations are used to calculate the concentration of acid and anhydride. Anhydrides in the presence of acids have also been titrated by other workers[597,598].

Per-acids. Wolf[599] has carried out the separate determination of perbenzoic acid in the presence of benzoic acid in benzene. Martin[410] titrated per-acids, primary and secondary hydroperoxides, and hydrogen peroxide in EDA. Mixtures of certain peroxides, for example H_2O_2 and ethyl hydroperoxide, have also been titrated.

Inorganic acids. H_2SO_4,[439,440,450,455,600] HCl,[334,450,454] $HClO_4$,[334,439,510] HNO_3,[334,600] H_3PO_4,[418,440,600] and other acids[7] have been titrated in non-aqueous solvents.

In the titration of H_2SO_4 in certain solvents the titration curve shows two inflections, corresponding to the first and second stages of the dissociation of H_2SO_4.[301,418,455] The curve for the titration of H_3PO_4 in pyridine shows three inflections[418].

2. Differentiating Titration of Acid Mixtures

Non-aqueous solvents can be used to carry out differentiating titration of mixtures of acids whose pK values in aqueous medium differ by at least 2–3 units, or, in some cases, by only 1 unit. It is possible to carry out differentiating titration of multicomponent mixtures of strong inorganic acids, mixtures of inorganic and carboxylic acids, various mixtures of carboxylic acids with one another and with phenols. In some cases the method of high-frequency titration makes it possible to distinguish isomers, for example m-, p-, and o-cresols. Differentiating titrations were first carried out by Lavine and Toennies[449], who titrated a mixture of acetic and perchloric acids in acetonitrile.

Mixtures of strong acids. Izmailov *et al.*[376] titrated mixtures of HCl with mono-, di-, and tri-chloroacetic acids in ketones and alcohols, and showed that the components of the mixture of HCl with monochloroacetic acid, and of HCl with dichloroacetic acid, can be titrated separately in either acetone or MEK. In the titration in acetone of HCl and trichloroacetic acid, which have similar strengths, the first inflection, corresponding to the neutralisation of HCl, is less distinct than in MEK and methyl propyl ketone. Shkodin and Izmailov[189] have titrated mixtures of strong acids in anhydrous acetic acid and concluded that the strongest acid in acetic acid is $HClO_4$, which can be titrated in mixtures with H_2SO_4, HCl, and HNO_3. p-Toluenesulphonic acid is titrated separately in mixtures with HNO_3 but not in mixtures with H_2SO_4 or HCl; HNO_3 cannot be titrated separately in the presence of H_2SO_4 and HCl. The authors found that in acetic acid carboxylic acids exhibit very weak acidic properties and are not titrated, so that it is possible to determine traces of H_2SO_4 in carboxylic acids in an acetic acid–acetic anhydride medium. The method has been used to determine traces of H_2SO_4 in citric, malic, and lactic acids[191,438].

Mixtures of H_2SO_4 with HCl, HNO_3, and p-toluenesulphonic acid have been titrated in acetonitrile[450]. Mixtures of $HClO_4$ with H_2SO_4 and HNO_3, and of HCl with H_2SO_4 and HNO_3, have been titrated in methyl isobutyl ketone[334]. Mixtures of H_2SO_4 with acetic and p-toluenesulphonic acids have been titrated in pyridine[370]. Cundiff and Marcunas[389,425,600] have studied the titration of mixtures containing strong acids, in acetone, isopropyl alcohol, methyl isobutyl ketone, DMF, acetonitrile, and pyridine. They found that mixtures containing strong acids can be determined quantitatively with sufficient accuracy only in pyridine.

Das and Mukhergee[440] published an interesting paper on the analysis of acid mixtures containing sulphuric acid, by titration in a 2 : 1 ethylene glycol–acetone medium. The method was used to determine mixtures of H_2SO_4 with HNO_3, HCl, $HClO_4$, H_3PO_4, p-toluenesulphonic, salicylic, and acetic acids. Titration of some three-component acid mixtures is also possible, for example H_2SO_4 + p-toluenesulphonic acid + acetic acid, H_3PO_4 + H_2SO_4 + salicylic acid, and H_2SO_4 + $HClO_4$ + H_3PO_4. Kalidas and Das[441] titrated strong acids ($HClO_4$, HNO_3, CCl_3COOH) and certain mixtures of acids (H_2SO_4 + phthalic acid; trichloroacetic acid + + monochloroacetic acid) in ethylene glycol. Components of mixtures of H_2SO_4 with H_3PO_4, $HClO_4$, HNO_3, p-toluenesulphonic and benzenesulphonic acids have been titrated separately in methanol[601].

Mixtures of weak acids. One of the most important advantages of non-aqueous solvents is that in these solvents it is possible to carry out differentiating titration of mixtures of various weak acids[348,360–362,432], particularly mixtures of carboxylic acids and phenols[391,407,408,602,603].

Fritz and Yamamura[452,453] titrated two-component mixtures of phenol and its nitro- and halogeno-derivatives, sulphonamide mixtures, mixtures of phenol derivatives with enols and imides, and mixtures of certain carboxylic acid derivatives. For example, dichloroacetic acid can be titrated in mixtures with acetic acid, but not in mixtures with monochloroacetic acid. The authors have titrated the three-component mixture 2,4-dinitrophenol + o-nitrophenol + + methyl salicylate.

The separate determination of components in mixtures of terephthalic and p-toluic acids in pyridine and in a 1 : 4 pyridine–chloroform medium has been described[421,422]. The first inflection corresponded to the neutralisation of terephthalic acid and the second to the neutralisation of p-toluic acid.

We have carried out differentiating titration of mixtures of carboxylic acids and of phenols with carboxylic acids in MEK.[459,460] In this medium it is possible to titrate separately components of mixtures of benzoic acid with malic or malonic acids, and mixtures of phenol with benzoic or oxalic acids. Two-component mixtures of mononitrophenol with di- or tri-nitrophenols, mixtures of di- and tri-nitrophenols, and three-component mixtures of mono-, di-, and tri-nitrophenols have also been titrated[578,579].

Shain and Svoboda[456] have carried out potentiometric titration of mixtures of weak acids in acetone. They established that differentiating titration is possible in the case of

mixtures of two acids, one of which has a strength equal to or greater than that of acetic acid, and the other a strength equal to or less than that of nitrophenols. In pyridine it is possible to titrate mixtures of cyanamide and dicyanamide, catechol and hydroquinone, and 2,4,6-trichlorophenol and *p*-chlorophenol[574].

Mixtures of strong, weak, and very weak acids. In differentiating solvents it is possible to titrate multicomponent mixtures of acids of different character and strength. In ketones it is possible to carry out differentiating titration of mixtures of strong mineral acids, weak carboxylic acids, and very weak acids of the phenol type. Thus a mixture of hydrochloric acid, acetic acid, and phenol can be titrated in DMF.[408]

Cundiff and Marcunas[418] titrated three-component mixtures of strong, weak, and very weak acids in pyridine. Mixtures of *p*-toluenesulphonic acid + benzoic acid + phenol, hydrochloric acid + acetic acid + *p*-cresol, and nitric acid + + malic acid + *p*-cresol have been titrated. The last mixture gives a curve with distinct inflections corresponding to the neutralisation of nitric acid, *p*-cresol, and the two stages of the dissociation of malic acid. The titration curve for a mixture of the four acids nitric, malic, acetic, and phenol shows five inflections. Bruss and Wyld[334] carried out differentiating titration of a mixture of five acids — perchloric, hydrochloric, salicylic, acetic, and phenol — in methyl isobutyl ketone. We have successfully titrated multicomponent mixtures of acids containing 5–6 acids in MEK.[460,575]

3. Conductometric Method

The conductometric method was first used for the titration of acids in benzene by LaMer and Downes[346]. Strong acids[437,511], carboxylic acids and phenols[604,605], and sulphonamides[606] have been titrated by the conductometric method in non-aqueous media.

Van Meurs and van Dahmen[398,607,608] studied the conductometric titration of carboxylic acids, phenols, and their mixtures, in pyridine, DMF, and their mixtures with benzene and xylene. Monobasic aliphatic acids (formic, acetic, and caproic) and aromatic acids (benzoic, salicylic, and *p*-hydroxybenzoic), dibasic aliphatic acids (oxalic, malonic, succinic, adipic, fumaric, maleic, and tartaric) and aromatic acids (phthalic, isophthalic, and terephthalic), and a number of phenols (phenol, cresol, xylenol, and *o*- and *p*-nitrophenols) have been titrated. The conditions for the titration of mixtures of carboxylic acids with one another and with phenol have been studied[391]. For mixtures of monobasic acids with dibasic acids, the best results were obtained by titration in pyridine and a 1 : 3 DMF–benzene mixture. Two-component mixtures of dibasic acids were titrated in pyridine and DMF.

Bruss and Harlow[609] studied the conductometric titration of phenol and its chloro, nitro, dialkyl, and trialkyl derivatives in benzene, toluene, xylene, carbon tetrachloride, pyridine, alcohols, and ketones.

The conductometric titration of aromatic mono- and di--carboxylic acids, mono- and di-hydric phenols, enols, and amides in pyridine, ethylamine, diethylamine, and triethylamine has been studied[414].

4. High-Frequency Method

Ishidate and Masui[392,610] have carried out high-frequency titration of weak inorganic and organic acids in a benzene–methanol medium. Boric acid is titrated with the addition of several drops of ethylene glycol. Dibasic organic acids whose molecules have 1–2 C atoms between the carboxyl groups show very characteristic titration curves. The anhydrides of carboxylic acids are titrated as dibasic acids.

Masui[611] titrated long-chain carboxylic acids (9–18 C atoms) and studied the high-frequency titration curves for dicarboxylic acids in non-aqueous solvents[612,613].

Lippmaa[436] studied the high-frequency titration of acidic groups in basic solvents. The best results were obtained by titration in a mixture of diethylamine, diethylformamide, pyridine, and a small quantity of thymol. Diethylformamide is added to increase the dielectric constant, since that of diethylamine is low. Pyridine is added to increase the solvent power. Thymol improves the equivalence point of the titration of carboxylic acids and polyhydric phenols. The author showed that the method can be used for the titration of individual phenols, enols, and carboxylic acids. The best results were obtained in the titration of monohydric phenols.

High-frequency titration in non-aqueous media makes it possible to analyse certain mixtures of isomers. Thus Karrman and Johansson[397,614] carried out high-frequency titration of phenols and mixtures containing phenols, in a benzene–methanol medium. Phenol, *o*-, *m*-, and *p*-cresols, α- and β-naphthols, catechol, resorcinol, hydroquinone, pyrogallol, and phloroglucinol have been titrated. Each component gives an inflection on the titration curve of mixtures of phenols. Mixtures of *o*- and *m*-cresols, *o*- and *p*-cresols, *o*-, *m*-, and *p*-cresols, and α- and β-naphthols have been titrated.

Riolo and Notarianni[615] studied the high-frequency titration of organic and inorganic acids in a benzene–methanol medium and in pyridine. Monobasic acids (dichloroacetic, bromoacetic, mandelic, benzoic, and acetic), dibasic acids (sulphuric, fumaric, malonic, succinic, and maleic), tribasic acids (citric and phosphoric), and acid mixtures (benzoic + *p*-toluenesulphonic, fumaric + *p*-toluenesulphonic, and maleic + acetic) have been titrated. In the titration of citric acid the inflections are more distinct in pyridine than in the benzene–methanol medium. Jamieson[616] titrated mono-, di-, and tri-hydric phenols, enols, and imides in ethyl alcohol. The high-frequency method has also been used by other authors[420,428].

5. Photometric Method

McKinney *et al.*[617] titrated phenols in butylamine photometrically. Satisfactory results were obtained both for the individual compounds and for their two-component mixtures with one another and with a strong acid. Hummelstedt and Hume[618] titrated phenol and its derivatives in isopropyl alcohol. They were able to carry out differentiating titration of two-, three-, and four-component mixtures of phenols, including mixtures of phenol with *o*-chlorophenol, 2,4,6-trichlorophenol with 2,4-dichlorophenol, *p*-nitrophenol with the *m*-isomer, etc.

VII. PRACTICAL IMPORTANCE OF NON-AQUEOUS TITRATIONS

The titration of organic compounds in non-aqueous media has been widely applied in research and industrial laboratories for the analysis of various industrial materials. Thus the acidic impurities in coal and petroleum products, fats, oils, and asphalt are determined by titration in alcohols, hydrocarbons, and other solvents[272,351-359,371,384,386,387,619-624]. The basic impurities in hydrocarbons and oil and coal products are determined in anhydrous acetic acid[272,472,515,625-628]. Minczewski *et al.*[270] used acetic acid as solvent to determine pyridine bases and ammonia in coal tar products. They also determined guanidine nitrate in the technical product containing ammonium nitrate and dicyanamide[255].

The determination of phenols in coal and its products[270,409,512], lignins[372,429,629,630], oils[631], resins and bitumins[396] has been described. The aliphatic acids in drying oils are determined by high-frequency titration in an alcohol–benzene medium.

Titration in non-aqueous solvents has been used to determine sulphuric and other acids in mixtures used for acetylation, sulphochlorination, and nitration[279,454,510]. Gribova and Levin[455] developed a method for titrating sulphuric acid and sulphonic acids in acetonitrile and acetone. The method is applicable to the determination of sulphuric, anthraquinone sulphonic, anthraquinonedisulphonic, naphthalenesulphonic, and benzenesulphonic acids for the control of the various stages of industrial sulphonation processes. The same authors[333] developed a method for analysing industrial mixtures of cyclo- and dicyclo-hexylamines and aniline, based on two titrations in acetone. The first titration makes it possible to determine the aniline content and the sum of the cyclo- and dicyclo-hexylamines, and the second titration, after preliminary titration with salicylaldehyde, makes it possible to determine the dicyclohexylamine content. The cyclohexylamine content is found by difference.

Titration in non-aqueous media is frequently employed for the analysis of pharmaceutical products[242,248-250,256,275,419,473,633-655]. Much work in this field has been carried out by Izmailov, Shkodin, Dzyuba, and others. They have used non-aqueous solvents to determine alkaloids, their salts, and the salts of organic bases and organic acids, used in pharmaceutical products[12,379,542,656-658]. These products are analysed in acetic acid[659-673], hydrocarbons[674-676], pyridine, acetone, EDA[677-683], and other solvents[684-695].

Sarson[696] used non-aqueous titration for the analysis of explosives. Nitro-compounds are titrated as acids in DMF and methyl isobutyl ketone, and differentiating titration of mono-, di-, and tri-nitrotoluene was described. Hexahydro-1,3,5-trinitro-*s*-triazine in explosives has been titrated[697].

Analysis in non-aqueous media has been widely used for the determination of a number of monomeric and polymeric organic compounds[698-705]. Streuli[287] has determined the basic copolymers of acrylonitrile. The copolymers are dissolved in a mixture of nitromethane and formic acid and the amines and salts of amines and quaternary ammonium bases are titrated. Halides are converted to the acetates by adding mercuric acetate. To determine salts of heterocyclic amines, the polymers are dissolved in DMF. Owens and Maute[706] have titrated the organic acids and bases present in acrylonitrile. To determine weak bases, acetic acid is added to the acrylonitrile, and to determine strong bases and acids, a mixture of ethylene glycol and isopropyl alcohol is used. Non-aqueous solvents have been used in the analysis of polymerisation accelerators[707] and the determination of the concentrations of vinylpyridine, acrylonitrile, and acrylic acid in elastomers[708].

Sergienko *et al.*[378,709] studied high-molecular-weight petroleum compounds. They used a mixture of benzene and alcohol to determine the acid numbers and saponification numbers of petroleum resins. Kirrmann and Daune-Dubois[431] determined the concentration of carboxylic acids and phthalates in polycondensation resins.

Titration in non-aqueous media is used to determine various functional groups in organic and macromolecular compounds: the carboxyl group in polyesters[501,710], the carboxyl and amino-groups in polyamides[699,711,712], the basic group in gelatin[713], and the benzoyl[714] and carbonyl groups[715-718] in other monomeric and polymeric compounds. Epoxy-compounds are also analysed in non-aqueous solvents[719-723].

Kreshkov and Drozdov have developed methods for the analysis of monomeric and polymeric organosilicon compounds, which exhibit acidic or basic properties in non-aqueous solvents[508,724-727]. The analysis of individual products is based on the potentiometric or visual titration in acetonitrile. Differentiating titration of two- and three-component mixtures is possible by a conductometric method in an acetonitrile–ethyl alcohol medium. A method has been developed for the analysis of nitrogen-containing organosilicon compounds with the nitrogen directly attached to the silicon atom, and with nitrogen in the organic radical. Other workers have titrated organosilicon compounds in non-aqueous media[728,729].

Ershov and others[373,730,731] developed high-frequency methods for the analysis of the raw materials, intermediates, and finished products of the plastics industry. Phenol in solutions of synthetic resins and plasticisers, acetic acid in acetylcellulose, 1,3,5-xylenol in the technical product, etc. have been determined.

Analysis in non-aqueous solvents is also used for organic acids and their salts in chrome liquors[732], foodstuffs[733,734], water-insoluble emulsions[735], tobacco[736,737], clays[738,739], wool, wax[395], agricultural chemicals[740], soaps[7], and other substances[741-743].

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