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Review on Analytical and Biological applications of Hydrazones and their Metal Complexes

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Abstract: Hydrazones are very important group of analytical regents for the determination of various metal ions by using various analytical techniques. Besides this use of hydrazones are also having biological activities also. In this paper we first discussed about the chemical nature of hydrazones and their biological activities. We mainly focused on the papers which were published during 1980-2011 on analytical applications (spectrophotometric and spectrofluorimetric) of hydrazones. We gave the total established conditions for the determination of various metal ions with hydrazones.

Keywords: Hydrazones, metal ions, Spectrophotometry, Spectrofluorimetry, Biological applications.

Introduction

Hydrazones as Analytical Reagents

Many organic compounds react with metal ions and form colored precipitates or solutions. Hence, they are extensively used as analytical reagents, even though it is difficult to predict with certainity which organic compound is suitable for the analysis of a particular metal ion. Yoe[1] gave a list of more than twenty ways in which they are used. It has been observed that the reactivity of organic reagents with metal ions in the use of the former as analytical reagents requires the presence of certain acidic or basic groupings[2] and coordinating atoms.

The aim of research in this field is the discovery of compounds possessing a high degree of selectivity and identification of the causes underlying such selectivity. While most of the reagents are not selective, various means are known where by the selectivity of a reagent may be improved. These include adjustment of the pH, and the use of masking agents which form complexes with the interfering elements in the determination of the test ion. Within the organic reagent molecule, there is generally a single acidic or basic group, or a combination of these two, which is the key to the reactivity of the reagent.

Literature survey has revealed that organic compounds capable of forming chelates or inner complex salts give better results than those containing only acidic or basic groupings, in the field of inorganic analysis. The element in the organic molecule through which the metal is bonded is generally oxygen or nitrogen, less usually it is sulphur. The oxygen containing groups most often met in organic reagents were -OH, -CHO, -COOH, >CO. The nitrogen containing groups (-NH_2, =NH, heterocyclic N) met with in general functional groups are amines (usually aliphatic), heterocyclic rings (usually pyridine), oximes (in which bonding tends to be coordinated to the nitrogen instead of replacement of hydrogen) and azo groupings. The aromatic orthohydroxy carbonyl compounds form stable six membered rings with the metal ions. Hydroxy carbonyl compounds derived from benzene and naphthalene, hydroxy quinones of naphthalene and anthracene series have been introduced as analytical reagents.

It is observed, that in many cases the formation of a precipitate or a soluble colored product is dependent on the presence of definite atomic groupings. Such compounds are therefore designated as metal binding groups with specific or selective action. But, it cannot be over looked that the reaction conditions play a definite role in this direction. Hence, proper choice of solvent and other factors are to be given equal importance. Organic compounds can undergo extensive alteration in their structure as a result of condensation and substitution reactions. These may bring in the useful changes in the reagent to make them better organic reagents. A survey of literature[3] shows, that organic compounds containing a phenolic or enolic group and a coordinating group containing nitrogen, oxygen or sulphur forms a variety of complexes with different metal ions.

It is found that -SH group of an organic compound exhibits a higher acidic character than similarly bound –OH group. Thio-keto group (>C=S) also plays an important role compared to its counterpart, keto group (>C=O)[4].

It is observed that aromatic compounds containing nitroso (-N=O) as well as phenolic –OH groups are also useful as analytical reagents. It is clear from the above brief review presented, that many organic compounds containing acidic or basic groups, besides the coordinating groups form chelates easily and have been used extensively as analytical reagents. However, these investigations reveal that sensitivity and selectivity of the reagent should be established, even though a few general guidelines are available to predict the potentialities of a reagent for the said purpose. In view of large and varied demand for the new methods to determine the metal ions, under specific conditions, the search for new reagents is a continuous process. This exercise of finding new and novel reagents as well as methods for inorganic analysis has a special significance in these days in view of the alarming and complex problem of environmental pollution.

Isonicotinoylhydrazones of carbonyl compounds act as good analytical reagents, but they have not been fully exploited. Hence, in the present investigation a detailed study of these reagents has been made with a view to find out their potentialities in inorganic analysis. Hydrazones are usually named after the carbonyl compounds from which they are obtained. Isonicotinoyl hydrazones are the condensation products of isonicotinic acid hydrazide and the carbonyl compounds. These isonicotinoyl hydrazones are prepared by refluxing a mixture of isonicotinicacidhydrazide and the desired carbonyl compound for 2-3 h in slightly alkaline medium. The compound usually crystallizes out on cooling.

Many of physiologically active hydrazones find application[5] in the treatment of diseases like tuberculosis, leprosy and mental disorders. Hydrazones also act as herbicides, insecticides, nematocides, rodenticides and plant growth regulators. Isonicotinicacidhydrazide (INH) is an important antitubercular agent and has potential sites for formation of complexes with metal ions. It is also observed, that isonicotinoyl hydrazones and their metal complexes possess higher activity and lower resistivity to tuberculosis bacteria. These reagents, apart from those specified above are also potential analytical reagents for the determination of several metal ions by different physico-chemical techniques, of which the spectrophotometric determination occupies a special place.

The analytical applications of hydrazones have reviewed by Singh[6] and Katyal[7]. The latest review was published in this area on 1982. In this, the author reviewed the papers published on analytical potentialities of hydrazones up to 1980. After 1980 so many researchers have worked on the analytical potentialities of hydrazones. Hydrazones have both analytical and biological applications, which attract so many researchers. Because of so much of work published in this area, we are interested to review the papers published on analytical applications of hydrazones from 1980 to 2011. We have so much confidence that this three decades survey is very useful to the scientists who are working in this area.

Biological Applications of Hydrazones and Their Metal Complexes

Interest in the study of hydrazones has been growing because of their antimicrobial, antituberculosis and antitumor activities[8,9]. Hydrazones play an important role in inorganic chemistry, as they easily form stable complexes with most of the transition metal ions. The development of the field of bioinorganic chemistry has increased the interest in hydrazone complexes, since it has been recognized that many of these complexes may serve as models for biological important species[10]. Coordination compounds derived from aroylhydrazones have been reported to act as enzyme inhibitors and are useful due to their pharmacological applications[11,12]. Hydrazones possessing an azomethine –NHN=C<u>H</u>- proton constitute an important class of compounds for new drug development. Therefore, many researchers have synthesized these compounds as target structures and evaluated their biological activities. Hydrazones and their metal complexes are biologically very active compounds. For example Ragavendran et al[13] have reported anticonvulsant activity of 4-aminobutyricacidhydrazone, Abdel-Aal et al[14] have reported antiviral activity of N-arylaminoacetyl hydrazones against *Herper simplex virus-1* and *Hepatitis-A virus(HAV),* Walcourt et al[15] have reported antimalarial activity of 2-hydroxy-1-naphth-aldehydeisonicotinoyl hydrazone and Savini et al[16] have reported antitumor activity of 3- and 5-methyl -thiophene-2-carboxaldehyde α-(N)-heterocyclic hydrazone derivatives.

Some important hydrazones and their complexes with antibacterial activity reported earlier are discussed here under.

Singh et al[17] have reported the antibacterial activity of cobalt(II), nickel(II), zinc(II), copper(II) and cadmium(II) complexes of acetophe- none-4-aminobenzoylhydrazone and 4-hydroxyacetophenone-4-aminobenzoylhydra- zone. The authors have evaluated the antibacterial activity of these complexes against *Escherichia coli* and *Aspergillus niger*. They reported that at each concentration, copper(II) complexes are more active than zinc(II) complexes with both the hydrazones. The antifungal activity of the metal complexes is less than their parent ligands. Nora H. Al-Sha'alan[18] has reported the antibacterial activity of 7-chloro-4-(benzylidenehydrazo)quinoline and its complexes with copper(II), nickel(II), cobalt(II), manganese(II) and iron(III). The author has evaluated the antibacterial activity against gram positive bacteria (*Staphylococcus aureus*), gram negative bacteria (*Escherichia coli)* and antifungal activity against *Candida albicans*. The ligand is also proved to be biologically active. The author reported that chelation tends to make ligand to act as more powerful and potent bactericidal agent.

Kucukguzel et al[19] have reported antibacterial activity of 2,3,4-pent aneotrione-3[4-[[(5-nitro-2-furyl)methylenehydrazide]carbonyl]phenyl]-hydrazone against *Staph -ylococcus aureus* and *Mycobacterium tuberculosis* at a concentration of 3.13 μg mL^{-1}. Gulerman et al[20] have reported the in vivo metabolism of 4-fluorobenzoic acid (5-nitro-2-furyl)methylene hydrazide which is effective against *Staphylococcus aureus.*

Sah and Peoples[21] synthesized hydrazones by reacting isonicotinicacid hydrazide(INH) with various aldehydes and ketones. These compounds are reported to have inhibitory activity in mice infected by various strains of *Mycobacterium tuberculosis.* Ersan S et al[22] have reported antimicrobial activity of N-[(α-methyl) benzilidene]-(3-substittuted 1,2,4-triazol-5-yl-thio) acetohydrazides. The authors synthesized eighteen new hydrazones by reacting ortho- or para- substituted acetophenones with (3-substituted-1,2,4-triazol-5-ylthio)acetohydrazide in ethanol.

The synthesized hydrazones are tested for antimicrobial activity. These compounds exhibits only poor activity against gram positive bacteria (*Staphylococcus aureus* and *Enterococcus faecalis*) and gram negative bacteria (*Escherichia coli* and *Pseudomonas aeruginosa*). Moderate activity is observed against fungi *Candida albicans, Candida parapsilosis* and *Candida krusei.* Ulusoy et al[23] have reported the synthesis and antimicrobial activity of N^2-substituted alkylidene /arylidene-6-phenylimidazothiazole-3-aceticacidhydrazides. These compounds show antibacterial activity against *Staphylococcus aureus* and *Staphylococcus epidermidis.* (MIC is 25.0 μg mL^{-1}). Kucukguzel et al[24] have reported the synthesis and antibacterial activity of ethyl-2-arylhydrazone-3-oxobytyrates. These compounds show significant activity against *Staphylococcus aureus.*

Rodriguez-Arguelles et al[25] have reported the antibacterial activities of cobalt(II), copper(II), nickel(II) and zinc(II) complexes of 2-thiophenecarbonyl hydrazone and isonicotinoylhydrazones of 3-(N-methyl)isatin. 2-thiophene carbonyl hydrazone metal complexes exhibits a strong inhibition on the growth of *Haemophilus infrenzae* (MIC is 0.15-1.50 μg mL^{-1}) and good antibacterial activity towards *Bacillus subtilis* (MIC is 3.0-25.0 μg mL^{-1}). The authors have reported that the antimicrobial activity of thiophene carbonyl hydrazone derivatives is greater than that of the isonicotinoyl hydrazone derivatives. Loncle et al[26] have reported the antifungal activity of tosylhydrazone-cholesterol derivatives against *Candida albicans* at a concentration of 1.0-5.0 μg mL^{-1}. Recently critical reviews have been published by various authors on antibacterial activity of hydrazones[27,28].

Analytical Applications of Hydrazones

Spectrophotometric Applications

Katiyer et al[29] employed Orthohydroxybenzaldehydeisonicotinoylhydrazone for its chelatometric properties with several metal ions. It precipitates nickel(II), zinc(II), palladium(II), cerium(II) and copper(II) and also form soluble complexes with lead(II), iron(II), cobalt(II), tin(II), vanadium(II), stabium(III), aluminium(III), iron(III), zinconium(IV) and thorium(IV).

Salicylaldehydeisonicotinoylhydrazone[30] is used for the spectrophotometric determination of gallium(III) and indium(III). Dimethylaminobenzaldehydeisonicoti -noyl hydrazone[31] was employed for its selective detection of mercury(I) and mercury(II). All the reagents which were used for the spectrophotometric determination of metals ions and the established conditions like λmax, pH, validity of Beer's law, molar absorptivity and the composition are presented in Table 1[32-103]. In this table we described about the reagents which were used in the period of 1980 - 2011. Kavlentis employed Phthalaldehyde bisguanylhydrazone[104] for the determination of Co(II), Cu(II) and Ni(II), 4-Dimethyl aminobenzaldehdye isonicotinoylhydrazone[105] for Arsenic(III) and Antimony(III), Salicylaldehyde isonicotinoylhydrazone[106] for Molybdenum(VI). Hydrazones were extensively used for the spectrophotometric determination of metal ions during this period. We gave the total description about all the papers published during this period about the spectrophotometric determination of metal ions by using hydrazones as analytical reagents.

Table 1. Analytical applications of Hydrazones in the determination of metal ions by using Spectrophotometer.

Reagent	Metal ion	λmax (nm)	pH	Beer's law ppm	ε ×10[4]	M:L	Solvent	Ref
2-Aceto-1-naphthal-N-salicylhydrazone	Mn(II)	-	-	-	-	-	-	32
α-(Benzimidazolyl)-α',α''-(N-5- nitro-2-pyridylhydrazone)-toulene	Cu(II)	410	6.0	0-2.50	3.81	1:2	-	33
2-Benzimidazolyl-3-sulphophenyl methanone-5-nitro-2- pyridyl-hydrazone	Co(II)	517	2.7-9.4	0.02-1.00	6.65	1:2	-	34
Benzyl-2-pyridylketone-2-quinolyl hydrazone	Hg(II)	475	9.0-10.4	-	5.01	1:2	-	35
Biacetyl mono (2-pyridyl) hydrazone	Zn(II)	440	10.0	0.04-1.20	5.20	-	-	36
2,2'- Bipyridyl -2-pyridylhydrazone	Co(II)						Iso amyl alcohol	37
	Pd(II)	585	0.5-1.5HCl	0.5-1.5	1.90	1:1	chloroform	38
2,2'- Dipyridyl-2-quinolylhydrazone	Pd(II)	-	-	-	-	-	-	39
Bis (isonicotinoylhydrazone)	Zr(IV)	410	2.8	-	-	-	-	40
N-cyanoacylacetaldehydehydrazone	Mo(VI)	790	Phospharic acid	-	-	1:1	-	41
1,2-Cyclohexanedione bis(benzoyl) hydrazone	Ti(IV)	477	1.75 - 3.00	-	1.05	1:2	-	42
1,2-Cyclohexanedione-2-oxime-1-guanylhydrazone	Cu(II)	-	-	-	0.52	-	-	43

Diacetylmonoxime-p-hydroxybenzoyl hydrazone	Sn(II)	430	-	0.25-2.76	3.2	1:2	-	44
2,6-Diacetylpyridine bis (furoyllhydrazone)	U(VI)	400	3.0	0.72-10.80	1.5	-	-	45
2,6-Deacetylpyridine bis(benzoyl hydrazone)	U(VI)	420	-	-	-	-	Dichloro methane	46
2,6-Diacetylpyridine bis (benzoylhydrazone)	V(V)	335	2.6-4.0	-	2.74	1:1	-	47
2,4-Dihydroxyacetophenone benzoylhydrazone	Mo(VI),	440	3-6	0.5-8.0	0.9	1:2	-	48
	V(V)	410	3-6	0.22-4.9	0.77	1:1	-	
3,4-Dihydroxybenzaldehyde guanylhydrazone	MoVI)	550	5.0	0.3-9	0.88	-	-	49
	Fe(III)	355	3-6	0.03-1.1	4.13	-	-	
	Co(II)	355	-	0.5-16.00	0.29	-	-	
2,4-Dihydroxybenzaldehyde isonicotinoylhydrazone	Ti(IV)	430	1.5	0.09-2.15	1.35	1:2	-	50
3,4-Dihydroxybenzaldehyde isonicotinoylhydrazone	V(V)	360	5.5	0.5-5.3	1.29	1:1	-	51
	Cr(VI)	400	5.5	0.5-7.7	1.35	1:1	-	52
	Pd(II)	380	3.0	0.5-20.0	0.53	1:1	-	53
2,4-Dihydroxybenzaldehyde isonicotinoylhydrazone	Th(IV)	390	2.0-8.0	0.30-7.00	2.20	1:1	-	54
	Zn(II)	390	4.0-10.0	0.10-1.50	3.55	1:1	-	55
2,4-Dihydroxybenzophe	Ce(IV)	400	10.0	0.7-7.0	2.0	1:1	-	56

none benzoichydrazone								
Diphenylglyoxal bis(2-hydroxybenzoylhydrazone)	Ca(II)	432	7.80	-	1.76	1:1	-	57
	Sn(II)	455	3.5-7.0		2.25	-	MIBK	58
	Ti(IV)	500	0.1H $_2SO_4$	-	1.5	1:3	Benzyl al	59
2,2’- Dipyridyl-2-benzothiazolylhydrazone	Fe(II)	427	4.5-8.4	-	3.41	1:3	-	60
		615	3.0-9.6		1.23	-		
Di-2-pyridylketone benzoylhydrazone	Pd(II)	455	-	0.00-10.00	0.94	-	Benzene	61
	Fe(II)	379	4.7-6.0	0.00-30.00	1.59	-	Benzene	62
	Ni(II)	-	-	-	-	-	-	63
Di-2-pyridylketone benzylhydrazone	Fe(II), Fe(III)	-	-	-	-	-	-	64
	Co(II)	-	-	-	-	-	-	65
	Ni(II)	-	-	-	-	-	-	66
Di-2-pyridylketone-2-furan carbothiohydrazone	Re(VII)	546	-	0.70-14.00	1.64	-	MIBK	67
2,2’-Dipyridylketone hydrazone	Cu(II), Hg(II)	-	-	-	-	-	-	68
Di-2-pyridylketone isonicotinoylhydrazone	Fe(II)	-	-	-	-	-	-	69
Di-2-pyridylketone-2-pyridylhydrazone	Pd(II)	-	-	-	-	-	-	70
	V(II)							71
Di-2-pyridylketone salicyloylhydrazone	Fe(II), Fe(III)	-	-	-	-	-	-	72
Di-2-pyridylketone thiophenylhydrazone	U(VI)	-	-	-	-	-	-	73
Di-2-pyridylmethanone-2-(5-nitro) pyridylhydrazone	Pd(II)	560	-	-	3.78	-	1,2-dichloro ethane	74
	Fe(II)	505	2.0-7.5	0.00-0.84	5.83	1 :2	-	75
2,2’-Dipyridyl-2-pyridylhydrazone	Fe(II), Fe(III)	535	-	0-30 mg/L 0-50	-	-	-	76

				mg/L				
	Pd(II)	560	-	Upto 12 mg/L	-	-	Chloroform	77
2-Hydroxybenzaldehyde-5-nitropyridylhydrazone	Co(II)	470	-	0.02-1.50 mg/L	6.5	-	-	78
2-Hydroxy-1-napthaldehyde benzoylhydrazone	V(V), Fe(III)	465 540	5.0	0.12-2.50 0.14-4.20	-	-	-	79
Isoniazid-p-diethylaminosalicylaldehyde hydrazone	Fe(III)	-	-	-	-	-	-	80
5-Nitro-2-pyridinecarbaldehyde-5-nitro-2-pyridylhydrazone	Ni(II)	561	5.5-10	-	13.5	1:2	benzene	81
N,N'-oxalyl-bis (salicylaldehyde) hydrazone	Cu(II)	422	2.0	0.4-1.8	0.92	1:1	-	82
Orthohydroxypropiophenone isonicotinoylhydrazone	U(VI)	380	3.0	0.47-17	1.15	1:1	-	83
p-Methylisonitrosoacetophenonehydrazone	Co(II)	520	7.5	0.2-6	1.83	-	Chloroform	84
p-Methylisonitrosoacetophenonehydrazone	Cu(II)	510	7.0	0.1-1.0	0.63	-	Chloroform	85
Phenyl-2-picolylketone-2-pyridyl hydra zone	Pd(II)	-	-	-	-	-	-	86
Pyridine-2-acetaldehydesalicylhydrazone	Pd(II)	425	2-4.5		1.3	1:2	Chloroform	87
Pyridine-2-acetaldehydesalicyloyl hydra zone	Pb(II)	380	8.6-9.3	1.5-6.2	1.93	1:2	Chloroform	88
2-Pyridinecarbaldehyde-3,5-dinitro-2-pyridylhydrazone	Ni(II)	-	-	-	-	-	-	89
Di-2-pyridylmethanone-1-phthalazinyl-	Ni(II)	500	4.3-10.7	0.1-1.0	5.40	1:2	Benzene	90

hydrazone								
Pyridine-2-carboxaldehyde-2-hydroxybenzoylhydrazone	U(VI)	375	3.5-4.6	1.0-5.6	4.74	1:2	MIBK	91
Pyridine-2-carboxaldehydeisonicotinoyl hydrazone	Fe(II)	-	-	-	-	-	-	92
Pyridoxalsalicylalhydrazone	Ti(IV)	440	0.9-2.5		0.55	1:1		93
2-Pyridylketonebenzoylhydrazone	Fe(II), Fe(III)	-	-	-	-	1:1	-	94
1-(2-Pyridylmethylildene-5-salicylidene) thiocarbohydrazone	Pd(II)	505	8.3	-	1.65	-	-	95
2-Pyridyl-3’-sulphophenylmethanone-2-pyrimidylhydrazone	Fe(II)	379 580	7.3-10 -	- -	4.79 1.18	1:1 -	- -	96
Pyruvaldehyde-2-benzothiazolylhydrazone	Cd(II)	-	-	-	-	-	-	97
Salicylaldehydecarbohydrazone	Zn(II)	445	5.4	-	-	1:1	-	98
Salicylaldehydeguanylhydrazone	Mn(II)	505	-	8-80µg/l	-	-	-	99
2-(3-Sulfobenzoyl) pyridinebenzoylhydrazone	Fe(II)	-	7-11	0-4	-	1:2	-	100
Thiazole-2-carbaldehyde-2-quinolylhydrazone	Pd(II)	625	acidic	0-4.2	1.93	1:1	benzene	101
2-Thiophenaldehyde-2-pyridylhydrazone	Cu(II)	435	8.5	0.017-1.2	-	1:3	-	102
2,3,4-Trihydroxy acetophenone phenylhydrazone	Th(IV)	370	3.5	-	32.46	1:1	-	103

Spectrofluorimetric Applications

Most of the researchers were used the hydrazones as analytical reagents for the determination of metal ions by using spectrophotometer, but few researchers considered these hydrazones as spectrofluorimetric reagents also. β- cyclodextrin-o-vanilinfurfural hydrazone[107] was used as spectrofluorimetric reagent for the determination of traces of cadmium(II). Di-2-pyridylketone-2-furoylhydrazone was used for the determination of gallium(III) by Salgado et al[108]. Orthovanillin furfural -hydrazone[109] and

Orthovanilinfuroylhydrazone[110] were used for the spectrofluori -metric determination of Os(VIII) and Cd(II), respectively.

Conclusions

Based on the above information about the biological and analytical applications of hydrazones it is concluded that the hydrazones are precious reagents in the determination of metal ions in various environmental matrices. We believe that the three decades survey is very useful to the future studies in this respect.

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