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Review On Ligands Containing Phenolic Unit Used As Fluorogenic Chemo-sensors During The Last Decade

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Abstract: This review focuses on recent developments arising from studies of fluorescent sensors for different biologically important metal ions and anions, utilizing ligands containing phenolic unit during last decade. Emphasis is given to phenol based ligands owing to their phenol to phenolate conversion with respect to pH and in consequent generation of fluorescence property. A number of chemo-sensors based on phenolic chromophore have recently been introduced in reputed journals.

Index Terms **- Fluorogenic, Chemo-Sensor, Phenol Based Ligand**

I. INTRODUCTION

For the last two decade there has been a significant growth in research related to molecular recognition and molecular signaling. Chemists, environmental scientist and biologists pay incredible strategic attention to design and synthesis of chemical sensor for detecting ionic and neutral species in meaningful way.[1]Chemo-sensing performance based on fluorescence spectroscopy may be considered as primary research tool owing to its simplicity, high sensitivity, rapid responsive ability and obviously non-destructive facility.[2]Such technique is further advantageous not only for ratiometric responses of the analyte but put in extra feather to find out the precise low detection limit of the sensing ion/molecule.[3]Since after rapid modernization, ions and molecules have increasingly been redistributed in the environment with sufficient accumulation in terrestrial and aquatic habitats, that may often invite adverse effects on the human health.[4]On the other hand, some biomolecules in their excessive or acute storage may alter the actual metabolic pathway and in consequent undesirable physiological activities are often observed. Undoubtedly, designing and synthesis of fluorescence chemo-sensor, operable under physiological conditions is a challenging task. In continuation, the fluorescent probes could effectively be utilized to study living cells including living organism using bio-imaging studies.[5]In this short review some phenol based ligands were discussed as fluorogenic chemo sensor mentioned in the existing literature.

1.1 General approaches for designing of fluorescent sensors

An ideal fluorescent chemo-sensor must fulfill three basic requirements – first of all, binding selectivity that is the receptor must have the strongest affinity towards the relevant target. Secondly the signal selectivity that is the fluorescence property should remain unaffected from the environmental interferences such as sensor concentration and photo-bleaching tendency *etc*. Finally, it must be stable under illumination. The fluorescence signal reflected by different pathways like as Photo-induced electron transfer (PET), [6] Photo-induced charge transfer (PCT), [7] Intra and intermolecular charge transfer (ICT), [8] Exited state proton transfer (ESIPT), [9] Fluorescence resonance energy transfer (FRET) [10] etc. Three different approaches are generally employed to construct optical sensors for the detection of different analytes (**Fig. 1**). The most popular strategy involves the chemodosimeter approach which is based on the irreversible binding phenomenon, usually centered on a specific reaction induced by the analyte of interest.[11]The process is associated with significant chemical modification involving both breaking and making of the covalent bonds results the formation of products differing from the starting material concomitantly with optically different properties. Secondly, the strategic use of sensors in which the binding sites and signaling subunits are linked covalently. In this case, interaction of analyte with the binding site makes a change in fluorescence of the signaling subunit. Last but not the least; a coordination complex may be used through displacement approach.[12]In these case, the introduction of analyte leads to decomplexation and regeneration of fluorophoric behavior of the signaling unit. However, the development of fluorescence chemo-sensors for ion/molecule in pure aqueous media is much more advantageous compare to those function only in organic solvents.[13]Among aforesaid strategies, fluorophoric ligand connected coordination complex-based displacement approach is probably the best way to develop chemo-sensor for its simple and straight forward mechanism along with readily operable suitability in physiological condition.[14] Hence, design of fluorescent chemo-sensors is an interesting field towards the chemists, not only because of the potential practical applications in cell physiology and analytical and environmental chemistry, but also as a proving ground for manipulation and/or engineering of various photo-physical processes toward an ultimate goal of selective and sensitive signaling of targeted species.

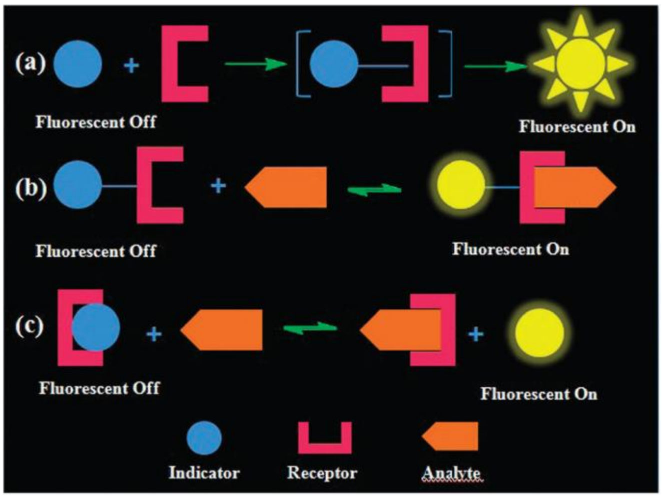

Fig.1. The three main approaches for sensing purpose: (a) chemodosimeter; (b) Binding site-signaling approach; (c) displacement approach.

1.2 Brief review of phenol based ligands for chemo-sensoning

A number of chemo-sensors based on phenolic chromophore have recently been introduced in reputed journals. Some of them are reported as Al^{3+} sensor. Development of simple probes to sense Al^{3+}*in vivo* and *in vitro* in real time is highly desirable since imbalance accumulation of Al^{3+} often correlates with variety of neuro-degenerative diseases. A phenol-based chemo-sensor (**P1**) with dual PET processes by simultaneous introduction of both nitrogen and sulfur donors were synthesized by Y. Lu *et al*. The fluorescence signal of the free chemo-sensor is in its *normal-off* mode due to sulfur and nitrogen donor mediating PET process. Added advantage of the probe is that it may operative over a wide pH range from 3–11. A large fluorescence enhancement was observed for the complexation with Al^{3+} owing to the inhibition of PET processes from both the sulfur and the nitrogen donors of the fluorophore.[15]

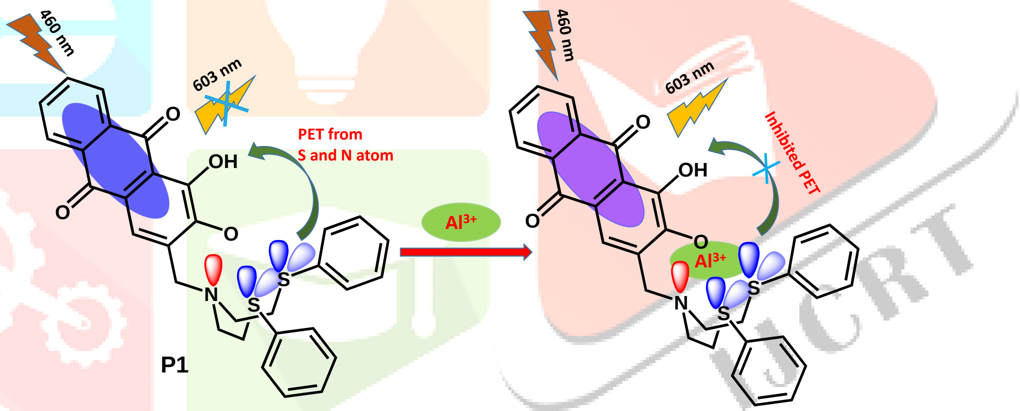

Fig.2. Scheme for Al^{3+} sensing mechanism

Another interesting phenol-based ligand (**P2**) was reported by X. J. Zheng *et al*. as Al^{3+} sensor. Here, inhibition of the ESIPT process by Al^{3+}ion selectively shows fluorescence enhancement.[16]Again, P. Ghosh and co-workers developed an ESIPT based highly sensitive ratiometric fluorescence sensor (**P3**) for selective Al^{3+}detection in acetonitrile as well as in mixed aqueous medium. About 2.3-fold enhancement in emission intensity and lower detection limit ~ 0.5 nM are mentionable features for aforesaid chemo-semsor.[17] Besides, M. Sukwattanasinitt and co-workers have prepared a series of phenolic ligands (**Chart-1**) for *turn-on* Al^{3+} sensors, based on metal chelation-enhanced fluorescence (CHEF) effect that inhibit the non-radiative PET and ESIPT processes.[18] Recently H. Hou and his group synthesized 8-hydroxyquinoline based a fluorescent chemodosimeter (**P4**) for detection of Al^{+3}ion. The probe exhibits *turn-on* fluorescence response to Al^{3+} owing to Al^{3+}promoted hydrolysis of carbon-nitrogen double bond.[19]Another highly sensitive chemo-sensor based on Schiff-base phenolic ligand (**P5**) for Al^{3+} detection in HEPES buffer medium wasre ported by S. Goswami and co-workers.[20] In this connection, organic dye chromone connected phenolic Schiff-based ligand (**P6**) deserves special attention for exhibiting Al^{3+}chemo-sensingin eco-friendly aquatic environment.[21] Another phenol containing Schiff-base ligand (**P7**) acts as Al^{3+} sensor in 100% aqueous media, reported by H. Wu and co-workers, renders novelty for its easy applicability to find out unknownAl^{3+} concentration in river and tap water.[22] Notably, in the contemporary period, anotherAl^{3+}chemo-sensor (**P8**) highlighted in literature for excellent bio-imaging study owing to its excellent cell permeability and low cytotoxicity.[23] In the same Year, D. K. Das *et al*. reported a novel condensation product (**P9**) of 2-hydroxy-1-naphaldehyde and 2-aminophenol to detect Al^{3+}exclusively in aqueous medium. The sensor was well explored for intracellular bio-imaging purpose using live rat L6 myoblasts cells.[24]

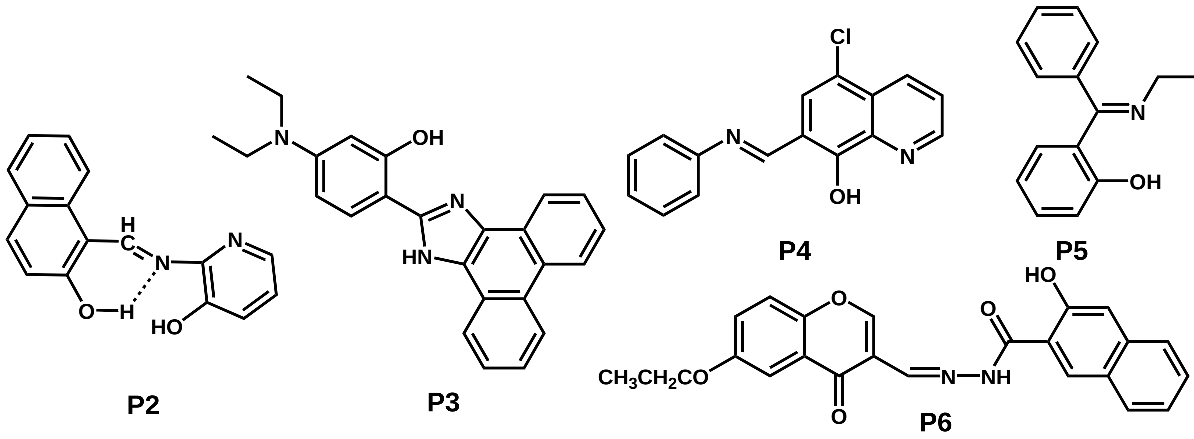

Fig.3. Structural representation of different ligands (P2, P3, P4, P5 and P6)

Phenol-based ligands are also exploited for precise Zn^{2+} detection. M. Ali and co-workers utilized *p*-cresol containing a Schiff-base ligand (**P10**) that was highly selective and sensitive to Zn^{+2} in mixed aqueous medium at physiological pH.[25] A colorimetric and fluorometric dual signaling probe (**P11**) for Mg^{+2} and $Zn^{+2}$was prepared by the same group in 9:1 acetonitrile/water (V/V) medium. The *turn-on* fluorescent enhancement for Mg^{+2} and Zn^{+2} was reported as 40 fold and 53 fold respectively and sensing response was nicely explored *in-vivo* cell imaging.[26] R. G. Harrison *et al.* have synthesized a metal ion sensor (**P12**) containing quinoline and pyridylamino phenolic precursor. It was acted as Zn^{2+}sensor in acetonitrile medium and Cd^{2+} sensor in aqueous medium respectively. The unique combination of pyridine and phenol group appended with quinoline plays the pivotal role for imparting fluoro-sensing selectivity.[27] T. K. Mondal and co-workers reported a coumarin based chemo-sensor (**P13**), in dual switching *turn-on* mode for $Zn^{+2}$and HSO_4^-. Gradual addition of Zn^{+2} to the probe in acetonitrile-water mixture (1:1, V/V) showed an excellent fluorescence emission intensity enhancement ~ 27 fold, whereas that for $HSO_4^-$was 17 fold.[28]

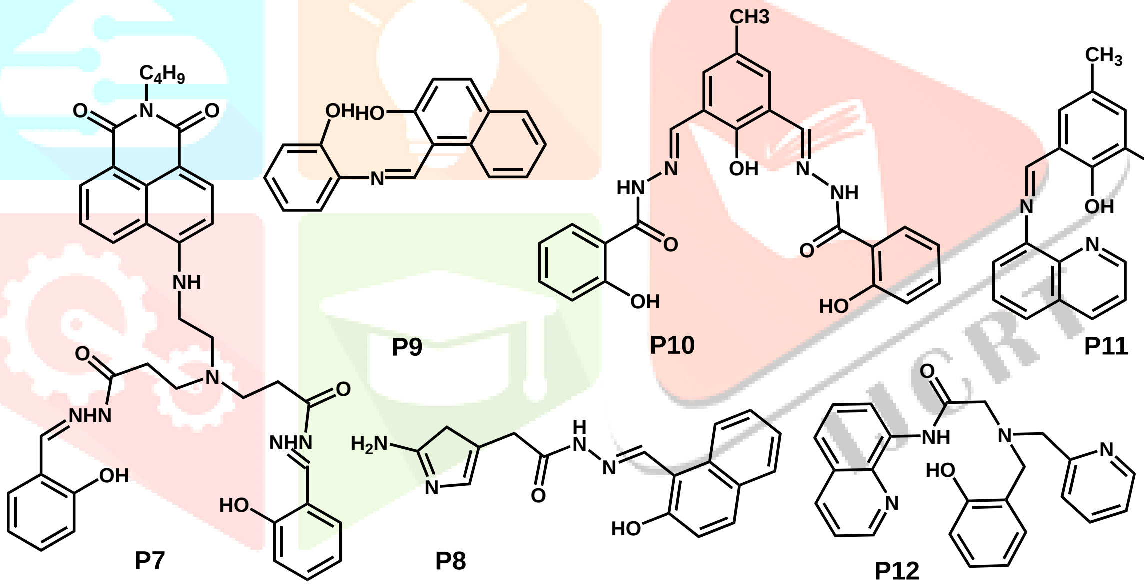

Fig.4. Structural representation of different ligands (P7, P8, P9, P10, P11and P12)

A phenol containing Schiff-base ligand (**P14**) acts as a chemo-sensor for Zn^{+2} over other competitive metals ions in methanol water (4:1) mixture was reported by S. C. Bhattacharya *et al.* using ITC mechanism.[29] In contemporary, M. Hosseini and co-workers have synthesized a water soluble phenol-based ligand (**P15**) for Zn^{+2} sensor at 7.2 pH. Interestingly the sensor was successfully utilized for fluorometric Zn^{+2}determination in water.[30] A physiologically compatible pyridoxal based Zn^{2+} fluorescence chemo-sensor, (**P16**) was reported by S. Goswami and co-workers. It exhibits a *turn-on* fluorescence response for Zn^{2+} in ethanol/water mixture. The combined effect of proton transfer between the prevailing tautomeric forms, C═N isomerization and in consequent metal induced CHEF are responsible for such sensing activity.[31] A water soluble Schiff-base (**P17**), prepared by the same group using salicylaldehyde and 2-amino-1-ethanol, also acts as Zn^{+2} fluorescence sensor at physiological pH.[32] An interesting multi-metal colorimetric and fluorescent sensor (**P18**) based on Schiff base bearing an "O-N-N"-coordination site was developed by S. Jiang and his group. This newly designed sensor is a *turn-on* fluorescence chemo-sensor towards Zn^{2+}. The sensor is also advantageous for cell-imaging. In addition color changes were observed from colorless to yellow, orange and purple respectively for selective binding with Cu^{2+}, $Zn^{2+}$and Ni^{2+} respectively. The corresponding color changes may be utilized for colorimetric determination. The sensor could simultaneously detect and differentiate three transition metal ions through fluorogenic (Zn^{2+}) and chromogenic (Cu^{2+}, $Zn^{2+}$and Ni^{2+}) pathways in aqueous medium.[33]

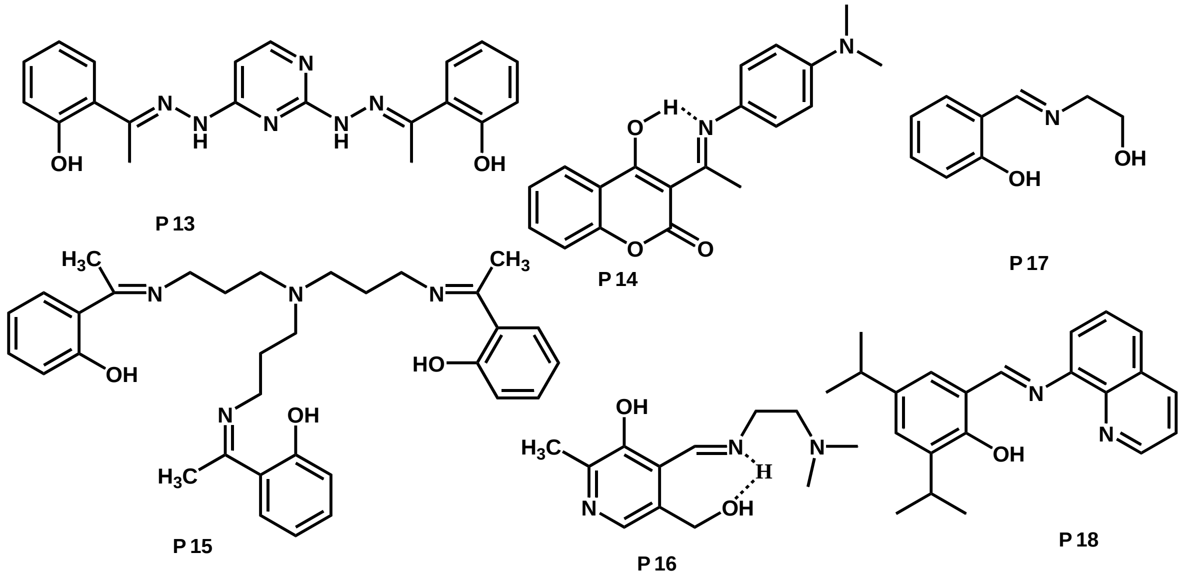

Fig.5. Structural representation of different ligands (P13, P14, P15, P16, P17 and P18)

An amino acid appended Schiff base ligand (**P19**) nicely exhibits its sensing ability for Zn^{+2} was reported by M. A. Neelakantan *et al*. Zn^{+2} ion after complexation in aqueous medium avoids the loss of energy *via* non-radiative transition, resulting fluorescence enhancement.[34] A. Misra and co-workers reported a phenol based new simple and inexpensive fluorescent probe, (**P20**) for Zn^{2+} detection, exploiting its promising CHEF/AIEE features. Sensing ability for Zn^{2+} was observed in dual path way in terms of both colorimetric and fluorometric response in DMF-water (9:1, V/V) medium.[35] P. Rossi *et al.* have synthesized the ligand (**P21**) containing a *bis*-phenolic moiety linked as side arm to an N,N′-dimethylethylenediamine scaffold. The deprotonated form of the ligand selectively coordinates to $Zn^{+2}$and Cd^{+2} ions in 50% ethanol/water medium, resulting sharp increase in emission intensity.[36]

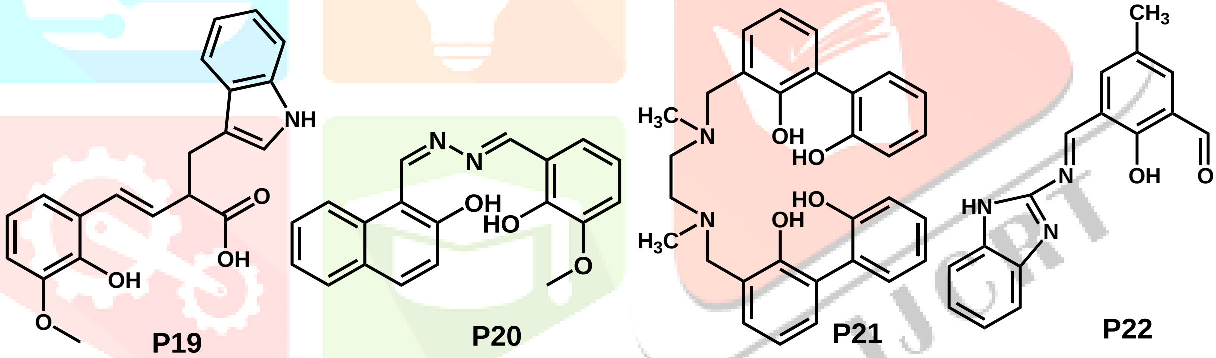

Fig6. Structural representation of different ligands (P19, P20, P21, and P22)

In contemporary, S. Dey and his group have synthesized a novel phenol-based ligand (**P22**), acting as Zn^{2+} sensor by inhibiting the ligand assisted PET in mixed aqueous solvent with very low LOD.[37] M. Shahid and A. Mishra have reported a light stimulated photo-enolization in phenol containing imidazole system by ESPT induced *turn-off-on* fluorescence and ratiometric responses for acetate in mixed aqueous medium.[38] Later, H. Li and co-workers have synthesized a novel cationic fluorescent probe for simultaneous detection of SO_3^{2-}/$HSO_3^{-}$and HSO_4^{-}ion with different emission channels. They also reported the fluorescence response in bio-imaging studies for aforesaid ions in HeLa cells.[39] The extracellular pH is often acts as a key factor in many biological processes. In this regard a pioneer work was reported by P. Roy and co-workers.[40] The phenol containing Schiff-base molecule, acts as a pH sensor and exhibits a strong emission band at 464 nm in Britton Robinson buffer solution (pH 2.0) for400 nm excitation. While increasing pH, the intensity at 464 nm decreases gradually and at the same time, a new fluorescence peak emerges at 530 nm and 435 nm in acid and basic region respectively. The probe nicely exploited for unknown pH determination for river water and successfully utilized in bio-imaging studies. It is mentionable that the same group has also designed another Schiff base probe by slight alteration of the mother ligand to monitor the unknown pH both calorimetric *as-well-as* fluorometrically with better sensitivity.[41]

During this period a new 2-(2′-hydroxyphenyl)thiazole-4-carboxaldehyde based fluorescent probe was reported in the literature for sequential detection of Al^{3+} and F^{-} ions in methanol medium. Al^{3+} was selectively detected through a "*switch on"* response driven by the selective complexation of Al^{3+} ions with ligand by ESIPT and CHEF mechanism followed by F^{-} ions detected sequentially through a "*switch off"* response.[42] In the same period a new diketopyrrolopyrrole -based dual-responsive colorimetric and fluorescent "turn-on" chemosensor developed for detection of Fe^{3+} ions with high sensitive and selective response based on the inhibition of PET effect and CHEF effect. It also showed a dramatic color change from purple to red with blue shift in absorption maxima and a 9.5-fold fluorescence enhancement response to analyte in water medium.[43] In next year a considerable amount of quality research was published in the literature. Phenyl-ethynyl-phenyl based Schiff base sensor was used to detect Cu^{2+} ions in water medium with high level of selectivity and sensitivity.[44] A novel terthiophene-derived chemo- sensor was synthesized by a group for colorimetric and fluorescent dual-channel sensing of Fe^{3+} and Cu^{2+} ions with low detection limits.

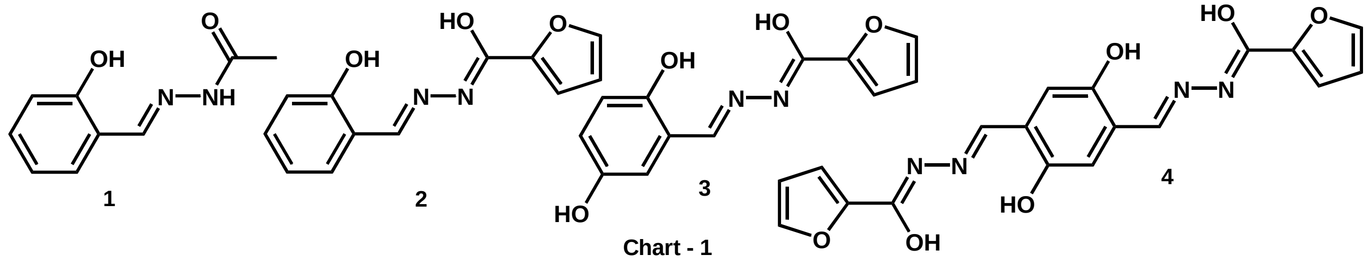

Chart - 1

The chemo-sensor showed a significant fluorescence “turn-on” response to Fe^{3+} and obvious fluorescence turn-off response to Cu^{2+} with high sensitivity, ultrafast response time and high reversibility. They employed the probe to rapidly and visually determine Fe^{3+} and Cu^{2+} in water based on paper test strips and solid silica gel with good results.[45] Based on salicylaldehyde and imidazo[2, 1-b]thiazole a simple Schiff base was exploited to detect Al^{3+} ions through a significant fluorescence enhancement respone via CHEF and inhibition of PET process and then to identify Cu^{2+} selectively through paramagnetic quenching.[46] Two new fluorescence sensors were published by G. Wang and group exploiting benzothiazole moiety identify Fe^{3+} through selective fluorescence quenching response. They employed the probes to monitor the existence of Fe^{3+} ions in living cells.[47] Contemporary a simple sensor was come to the literature to detect various ions and PPi through “turn on-off” response in mixed medium with paper strip test in tap water.[48]

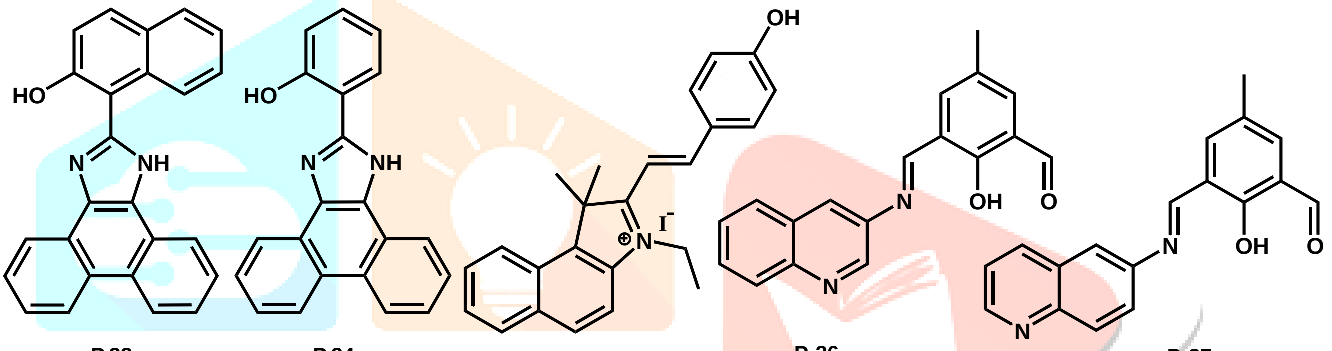

Fig7. Structural representation of different ligands (P23, P24, P25, P26 and P27)

A group of Chinese scientists synthesized a fluorescent chemo-sensor based on a bis(salamo)-like tetraoxime to detect selective metal ions in water medium. It used in double N_2O_2 cavities as sensing elements, which can be combined with specific metal ions to achieve ion recognition. It can detect Cu^{2+} ions by fluorescence quenching response, and a test strip loaded with the sensor is used to quickly and accurately identify Cu^{2+}. Besides, the chemical sensor also can continuously recognize Al^{3+} in the system and realize the interference-free identification effect of other trivalent metal ions on aluminum.[49] Recently, S. K. Chattopadhyay reported a Ni-complex containing phenolic unit to detect most toxic cyanide ion in water medium.[50]

3. Conclusions and Future Outlook

Among many different types of binucleating ligands and more generally polynucleating ligands, the phenol-based ligands and their metal complexes have attracted particularly a great number of researchers. This is due to the key role played by the phenolic group which has many useful electronic and structural characteristics such as: (i) charge as function of pH, spanning from lower pH to higher pH values; (ii) bridging capability, often the phenolic donor atom binds two metal centers in close proximity; (iii) the benzene ring present allows a great synthetic flexibility, (iv) the phenolic chromophore, itself often exhibits fluorescence. Consequently tuning the various characteristic modes to exhibit different physicochemical properties are a great challenge and render special attention. On the other hand, the acid-base properties of the phenol in aqueous solution depends on the ligand topology in which it is inserted, in fact although the phenol loses its acidic proton at pH >10, giving the phenolate anionic species, the process can occur also at lower pH values when the phenol takes part of an amino-phenolic ligand. Often the phenolate oxygen atom takes part to bridge two metal centers. Considering the immense applications in diversified field, phenol containing ligands and their polynuclear complexes demand further exploration. Designing and synthesis of new application oriented phenol based ligands is also a new challenge in this field. Moreover, synthesis of new homo or hetero polynucleating macrocyclic or acyclic metal complexes and isolation of their single crystals not only significant for structural determination that mimick the various metalloenzymes but also important to generate supramolecular motif to understand interesting physicochemical phenomenon.

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