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Review on Azo-dyes and their corrosion Inhibitory Potentials in Aggressive Media- Experimental and Theoretical approach

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Abstract

In a bid to eradicate/reduce corrosion of metal parts in home appliances and industrial machines, the use of corrosion inhibitors have been employed in recent times. This is necessary in order to reduce production cost and improve productivity. This work reviewed the corrosion inhibitory capacity of azo-dyes. The review showed that most Azo-dyes are synthesized majorly by diazotisation and coupling reactions. They showed very strong inhibitory capacity owing to the presence of the azo group(s), substituent group(s) like –OH, -SH, NH_2 and the plane conjugated system of their moieties. These azo compounds protect metals against corrosion by adsorbing on the metal's surface forming a protective layer with their adsorption following at least one of the classical adsorption isotherms. They showed excellent inhibition efficiencies which depend on the concentration and the temperature of the environment. Azo-dyes are therefore good prospective inhibitors against most metals corrosion in an aggressive environment.

Keywords: Azo-dyes, corrosion inhibitors, acidic media, heteroatoms.

Introduction

Organic compounds play important roles in the inhibition of corrosion of metals in aggressive media – acidic and basic media [1-4], while, the most effective organic inhibitors are organic compounds possessing π-electron system, atoms with lone pair electrons (phosphorus, sulphur, nitrogen, Oxygen), and the plane conjugated systems including all aromatic rings [5-8]. These groups of atoms or bonds aid electronic interactions between the organic corrosion inhibitor and the metal surface which make the adsorption of the inhibitors onto metal surface possible. Study revealed that the inhibition of corrosion by most organic inhibitors is by the formation of complex compounds between the metal-ions and electron donating atoms, such as nitrogen atom, of the organic compound [9]. The organic molecules usually adsorb at the metal-solution interface to inhibit the corrosion rate on the metal surface [10]. The efficiencies of these organic inhibitors, in most cases, depend on the characteristics of the environment in which it acts viz; the nature of the metal surface, electrochemical potential at the interface of the metal and the inhibitor, structure of the inhibitor, its molecular size, mode of adsorption, protected area of the metal surface and the concentration of the inhibitor [11,12]. On the other hand, the choice of any inhibitor depends strongly on its economic availability, its efficiency in inhibiting the substrate material and its environmental side effects [9]. Most of the used organic compounds as inhibitors are Azo-dyes [9, 13, 14], benzaldehydes [15], furans [16], triazoles [17-19], pyridines [20], isoxazolidines [21, 22], thia-diazole [22-24], oxadiazoles [25], and imidazolines [1]. Among these, azo-dyes have been proved to show excellent inhibitory properties with significant inhibition efficiencies [9, 26, 14, 27]. The inhibition process of these compounds have been proved to follow inhibitor adsorption isotherm [9, 28], while, their efficiency depends on the structure and chemical characteristics of the adsorbed inhibitor layer formed on the metal surface. This present work, therefore, aimed at reviewing the synthesis of azo-dyes and its usage as organic corrosion inhibitor for metals in corrosive media.

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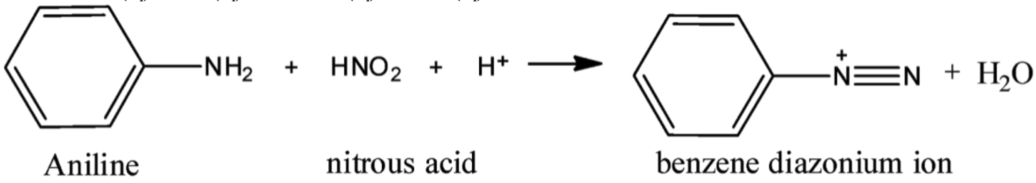
Azo-Dyes

Azo dyes are organic compounds having azo group (-N=N-) in their moieties conjugated with two, distinct or identical, mono- or polycyclic aromatic or hetero aromatic systems [29]. The chromophoricazo group enhances the stability of the azo compounds by extending the delocalised system of the arenes [30]. Due to this well delocalised electron system, they are often brightly coloured with some of them showing orange, red and yellow colours as they absorb light having its wavelength at the visible region of the electromagnetic radiation. They are the largest and the most versatile class of organic dyes with applications in pharmaceuticals [31], cosmetics, food and dyeing or textile industries as well as in analytical chemistry owing to their distinct physico-chemical properties and biological activities [29-31]. They are the most studied class of organic dyes with major application as dyeing pigment [29]. In addition, studies have also shown that this class of organic compounds showed various medicinal activities such as antidiabetics [32], antiseptics [33]. antineoplastics [34], antibacterial [35, 36],and antitumor [37]. Many of the azo dyes serve as chromogenic reagents used for the determination of several metal ions [38, 39]. They have also been reported to show good inhibitory capacity for the corrosion of many materials in both acidic and basic media [40-42]. These activities shown by this class of organic compounds may probably result from the presence of the azo functional group in their molecular backbone.

Synthesis of Azo dyes

Azo-dyes are mostly prepared by coupling reaction between diazonium salt and a coupling agent generally called the coupler [43, 44]. Firstly, diazonium salt is prepared in ice-cold solutions, as it decomposes explosively at temperature above 5^{o}C, by diazotisation reaction. In this reaction, a cold solution of sodium nitrate is added to a solution of arylamine in concentrated acid kept below 5^{o}C. The sodium nitrate reacts with the acid first to generate unstable nitrous acid in-situ (Equation 1) which then reacts with the arylamine to produce the diazonium salts (Equation 2) which is used immediately [30].

$$NaNO_{2(aq)} + HX_{(aq)} \rightarrow HNO_{2(aq)} + NaX_{(aq)} \qquad 1$$



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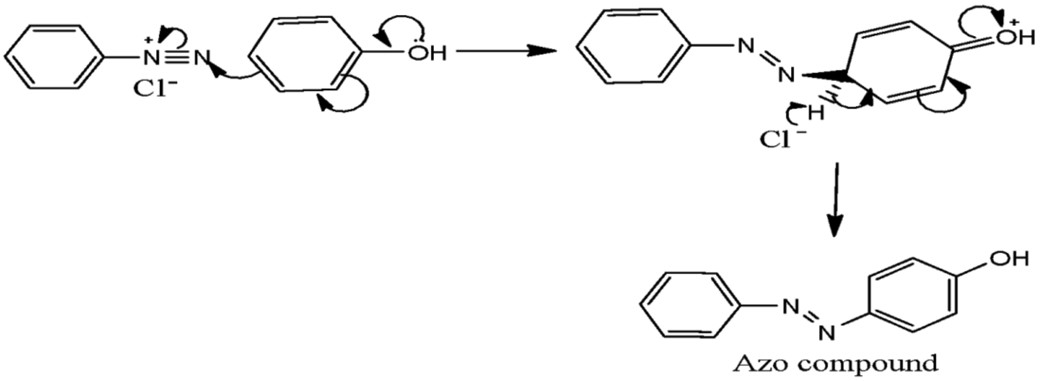
Scheme 1
Synthesis of aromatic diazonium salt [30]

The second phase of the azo-dyes synthesis is the diazo coupling reaction which involves the reaction of the generated diazonium salt with another arene known as the coupling agent. The diazonim salt acts as an electrophile reacting with the aromatic ring of the coupler, the nucleophile. If the functional group of the coupling agent is in position one, the coupling agent will react in either at the two (i.e. ortho) position or four (i.e. para) position of the benzene ring but the para position takes precedence or else been already occupied [30]. Although the diazoniumcation is a weak electrophile, it is very reactive towards the coupler due to the activation of the aromatic ring of the coupling agent by an attached activating group like -OH or -NH_2 group. Coupling reaction is an electrophilic type of substitution reaction which gives the azo-dye whose colour is determined by the coupler. The pH of the solution must be mildly acidic or neutral as the reaction will not take place if the pH is too low [45, 30]. Scheme 2 illustrates this mechanism as the benzenediazonium chloride salt is coupled with phenol.



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Scheme 2
Coupling of benzene diazonium chloride with phenol [30]

Azo-dyes as corrosion inhibitor

The use of organic inhibitors is one of the most efficient and robust methods for combating corrosion of metals [27]. Most of the organic compounds acting as inhibitors contain electronegative atoms like nitrogen, oxygen and or sulphur atoms [46, 47]. The presence of these atoms aids the adsorption of the inhibitors to the metal surface via their lone pairs. Azo dyes is the most widely used class of dyes [9, 48] and have been shown to have excellent inhibitory capacity for corrosion of metals due to their ability to form complex compounds between the metal-ions and the nitrogen of the azo group binding at the electrode surface [49]. As revealed in literature, the adsorption of any corrosion inhibitors depends mainly on physico-chemical properties of the molecules such as the functional group, steric factor, molecular size, molecular weight, molecular structure, aromaticity, electron density at the donor atoms and orbital character of donating electrons [50-52], and also, on the electronic structure of the molecules [53-55]. Most azo compounds possess these properties and hence, the greater adsorption capacity for them to act as efficient corrosion inhibitors. Azo dyes are used in the past and in recent times as corrosion inhibitor, among which include:

- [N-substituted] p-aminoazobenzene corrosion inhibitors reported in 2014 [9]. The various derivatives used in the work showed excellent corrosion inhibition efficiencies with percentage inhibition up to 82.48 % reported.
- Mono Azo Dyes Compounds as Corrosion Inhibitors for Dissolution of Aluminium in Sodium Hydroxide Solutions with the percentage inhibition efficiencies up to 73 % reported [28].
- Corrosion inhibitive potentials of Mordant Green 17, Mordant Orange 37, Trypan blue (Direct Blue 3B), DIAMINE GREEN B (Direct Green B) and Woodstain Scarlet (Acid Fast Red RN) on mild steel dissolution in hydrochloric acid solution using weight loss method and showed that Woodstain Scarlet (Acid Fast Red RN) possessed the highest inhibition efficiency (83.1 %) at its highest concentration [26].
- BenzonitrileAzo dyes (benzonitrileazo-dyes synthesised include (Z)-2-(cyano((4-methoxyphenyl) diazenyl)methyl) benzonitrile, (Z)-2-(cyano (p-tolyldiazenyl) methyl) benzo-nitrile, and (Z)-2-(((4-chlorophenyl)diazenyl)(cyano)methyl)benzonitrile. Study of the inhibitory properties of these azo-dyes using potentiodynamic polarisation method revealed (Z)-2-(cyano((4-methoxyphenyl)diazenyl)methyl)benzonitrile, with the percentage inhibition of 99.5 %, as the most efficient [27].
- Azochromotropic acid dye on carbon steel in 0.5 M sulphuric acid using both weight loss and potentiodynamic polarisation methods. The azo compound studied yielded up to about 82.3 % inhibition efficiency with weight loss analysis [14].
- Corrosion inhibition of Carbon Steel in Acidic Solution by Alizarin Yellow using both gravimetric and electrochemical measurements techniques. The studied compound showed good corrosion inhibitory characteristics for carbon steel in the tested acid solution with its efficiencies (up to 97.3 % by gravimetric measurement) shown to increase with the increase both its concentration and the experimental temperature [56].
- Allura red, sunset yellow and Amaranth were investigated as corrosion inhibitors of mild steel in sulphuric acid with the efficiencies up to 90%, 80% and 78% recorded respectively for each of them [42].

Dependence of Corrosion Inhibition Efficiency on Concentration of Inhibitor and Temperature

The corrosion inhibition efficiencies of many organic inhibitors have been shown to increase with the increase in the concentration or amount of the inhibitor used [3, 60, 56] except for the use of *Caesalpiniapulcherrima* [57]. For most azo-dyes, the inhibition efficiencies increase with increase in their concentration [60, 9, 27, 28, 48]. This may be due to the increase in adsorption of the inhibitor molecules on the metal surface as the inhibitor’s concentration increases thereby preventing further attack of the aggressive medium on the metal. On the other hand, the inhibition efficiencies were shown to decrease with increase in temperature for most organic inhibitors [60, 27, 28] except in few cases [3, 58, 59, 61] where the inhibition efficiencies increase with the increase in temperature. In most azo-dyes used as inhibitors, their inhibition efficiencies decrease with increase in temperature [60, 28, 48, 62]. This is attributed to the easy desorption of the inhibitors from the metal surface as the experimental temperature increases [62].

Theoretical Investigations on Corrosion Inhibition

A lot of researches have been carried out and reported on the use of theoretical methods for predicting and investing the corrosion inhibitive potentials of molecules, including azo dyes, in past and in recent times [7,40, 60, 63]. Many of these researches establish the fact that organic molecules with extensive π-conjugation and with heteroatoms like N, O, S, P etc, whose lone pairs could bind to metal surfaces are strong candidates for corrosion inhibition applications, including azo dyes [60, 64]. Some azo dyes have been investigated for their inhibitive potentials via theoretical methods [60, 65-67] and they have been reported to be potent corrosion inhibitors. The density functional theory (DFT) has been mostly used for investigating the molecular and electronic properties of organic molecules and azo dyes [60, 67]. Global reactivity descriptors like the molecular hardness (η), softness (S), global electrophilicity (ω) etc are usually calculated in a bid to determine the reactivity of molecules and binding ability [60, 67]. These properties are a direct consequence of the frontier molecular orbital energies, E_{HOMO} and E_{LUMO} (energies of the highest occupied molecular orbital and those of the lowest unoccupied molecular orbital).

Conclusion

From the above overviewed, it could be inferred that azo compounds possess unique characteristics that give them corrosion inhibitory power in different media. This unique property could be attributed to the possession of the azo group that enhanced both the extensive delocalisation of the pi-electron system of the compound as well as aiding the physical and chemical adsorption of the compounds on the adsorbent in the corrosive media. Their corrosion inhibition efficiencies, in most cases, increase with increase in their concentration and decrease with the increase in temperature.

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