



Review

Aromatic Metal Corrosion Inhibitors

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Abstract: Molecular inhibitors added to the corrosive medium attacking metallic materials are a well-established way of combating corrosion. The inhibitive action proceeds via adsorption of the inhibitor on the surface to be protected. Aromatic building blocks in the inhibitor play a major role in its protective action, and further details like substituents, heteroatoms, and molecular geometry contribute. An overview focused on aromatic inhibitors is provided, aiming at the identification of particularly promising inhibitors and their mode of action. Directions for further research and development are pointed out in the conclusion.

Keywords: corrosion; soluble corrosion inhibitors; molecular inhibitors; aromatic inhibitors; substituent effects

1. Introduction

Attempts and methods to meet the huge challenges posed by corrosion of all forms, but particularly of metals, by corrosion protection are numerous and have been well developed into many different approaches as described in several monographs and extended reviews [1–16] and as frequently highlighted in contributions in this journal. Depending on the details of the material or system needing protection, corrosion inhibitors may be a practically relevant or even attractive option; sometimes their use may be the only option.

For systems with large amounts of circulating water as a heat transfer medium (power stations, air conditioners, heat exchangers, water-cooled engines [17]), dissolved inhibitors added to the circulating medium at concentrations as small as possible can slow down the metal dissolution (anodic protection) and/or the hydrogen evolution/dioxygen reduction (cathodic protection). Possible inhibitors must be sufficiently soluble in the circulating medium (water); they should be economically viable, must have sufficient chemical stability under operating conditions (i.e., at elevated temperatures), and must be environmentally compatible. This obviously includes a level of toxicity that is as low as possible (see also [18]). For the latter reason, “green inhibitors” [11,19,20] obtained from natural sources (plants) have attracted growing attention in recent years; their natural origin is assumed to be an indicator of inherent environmental compatibility. The latter term is also applied to inhibitors prepared following “green synthetic routines” or prepared from precursors or raw materials claimed to be green [21]. Possibly, an “eco-addition inhibitor” prepared as a plant extract may also be called a green inhibitor [22].

In some applications, major changes in concentration of the inhibitor must be taken into account; e.g., in a cooling tower, some of the heat to be dissipated into the environment is released by evaporation of water, leaving a significantly higher concentration of the



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inhibitor (which is assumed not to be volatile under the operating conditions), which should not precipitate or cause any other undesirable effects. Similar reasons must be considered when looking for inhibitors suitable for temporary protection by spraying or dipping of large items during manufacturing and construction (see e.g., [23]). They also must be kept in mind when application as a volatile inhibitor is intended [24–26] as well as when packing paper impregnated with volatile inhibitors is considered [27]. Although corrosion in aqueous environments should generally be inhibited, there are further applications in industry where corrosion in oil or other hydrocarbons circulating in a refinery (see e.g., [28]) poses a challenge for inhibitors with slightly different properties better adapted to the different solvent environment [29]. The influence of the inhibitor on the wetting behavior is important; several metal salts of sulfo- and nitro-alkyl-aromatic compounds were found to be most effective. In particular, aromatic compounds may pose problems as contaminants in runoff water [30–32], in water produced in the oil industry [33], and in the environment [34].

Inhibitors mostly act by adsorptive interaction with the surface to be protected [35–38]; some of them form coatings, i.e., passivating layers, on the surface [39]. In a review called by the authors advanced and critical for reasons hard to find, many aspects of corrosion inhibitors have been addressed [40].

Whether frequently invoked films are just adsorbate layers or multilayer arrangements is left open in most cases; thus, this detail is addressed below only when the presented evidence warrants this. The interaction needed for adsorption is supported by electron-rich heteroatoms, functional groups, and moieties at the molecules to be adsorbed. According to a literature survey, electron-withdrawing groups/substituents seem to decrease inhibition efficiency [41]. Given these general observations, it is not surprising to note that Hammett parameters have been considered in regard to any correlation between their values for a given molecule and its inhibition efficiency [41,42]. The authors conveniently ignore that Hammett parameters are derived from kinetic measurements and are primarily intended for understanding of reaction kinetics in organic chemistry. Despite these inherent limitations, Hammett and Taft substituent constants have been discussed with respect to inhibitor performance [43].

Inhibitors containing aromatic units [44] like substituted benzenes have attracted attention because of their various adsorptive interaction possibilities, as indicated in Figure 1, where *p*-phenylenediamine is taken as an example.

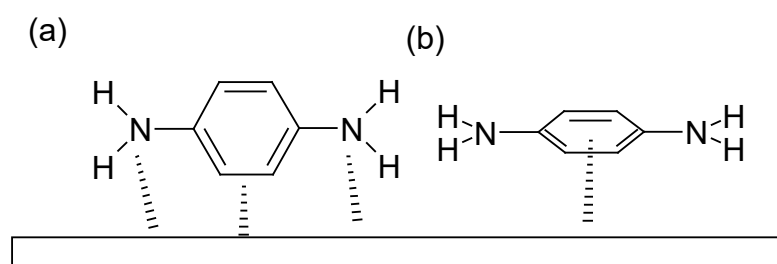


Figure 1. Conceivable modes of interaction of *p*-phenylenediamine with a surface, for details see text.

The lone electron pairs of the nitrogen atoms in the amino substituents, the double bonds of the aromatic ring (both in (a)), and the π electrons of the aromatic ring system (b) may interact with the surface to be protected. In case of e.g., iron, this may involve vacant d-orbitals, but empty d-orbitals are no prerequisite for adsorption (see e.g., zinc) The importance of aromatic electron systems as well as the presence of heteroatoms supporting adsorptive interactions has been highlighted elsewhere [45–47]. In a typical example, inhibition of aluminum corrosion by a linear (linalool) and an aromatic (eugenol) inhibitor molecule were compared; the aromatic one performed significantly better [48].

Depending on the composition of the corrosive medium (the electrolyte solution), a further option may be relevant. Anions, in particular halide ions, may be adsorbed on the metal surface first. Now positively charged cation species including inhibitor molecules

may adsorb on the surface. Depending on the specific conditions, electrostatic interactions as well as chemisorption may be active [49]. Such interactions have been studied with numerous experimental methods, in particular with spectroelectrochemical techniques [50]. Despite the broad availability of these tools, they have rarely been used in reported corrosion studies, leaving the explanation of observed effects and efficiencies on a rather speculative level. This is particularly regrettable when particular substituents known for adsorptive interactions (as “anchoring sites” [51]) may be inspected easily with in situ vibrational spectroscopies. A notable exception has been reported [52]. Adsorption of several sulfur-containing aromatic corrosion inhibitors was examined with surface enhanced Raman spectroscopy (SERS). Strong sulfur–iron σ bonds were observed, in addition to evidence of interaction between π electrons of the aromatic ring systems and the iron surface. Another application of SERS on CO_2 corrosion has been reported [53]. Quasi-complementary in situ infrared spectroscopy was used to monitor adsorption of aliphatic and aromatic carboxylic acids suggested as corrosion inhibitors for magnesium [54]. In both cases, the carboxylate groups acted as anchoring sites, but whereas the aliphatic acids were oriented perpendicularly at the surface, the aromatic ones were oriented in plane with the metal surface. This orientation provided a further interaction option for the π electrons of the aromatic system, enhancing adsorption and thus inhibition. Further non-electrochemical (sometimes called non-traditional) methods have been used in corrosion inhibition studies, like the electrochemical quartz crystal microbalance [55,56]. As with traditional methods, their results provide only a quantitative picture, not more.

More recently, tools of theoretical chemistry have been applied to investigate possible and the most effective adsorbate geometries; for an example unfortunately lacking experimental verification, see [57]. This molecular view of adsorptive interaction can be put into a broader perspective when considering these interactions as follows:

- The attraction between charged as well as neutral molecules and metal through electrostatic forces;
- The interaction of the metal with lone electron pairs in the molecule;
- Interactions of π electrons with metal;
- A combination of the first three possibilities.

Further considerations apply to corrosion inhibitors in more general terms:

- Sufficient solubility;
- Environmental compatibility;
- Nontoxicity;
- Cost and sustainability;
- Chemical stability in the particular environment.

Aromatic molecules with functional substituents supporting these various modes of interaction have attracted attention accordingly, although in a report on molecular structural aspects of organic corrosion inhibitors, this detail appears to be barely noteworthy [58]. The particular function of the $-\text{SH}$ substituent has been reviewed [59]. In addition, the substituents frequently also affect solubility. Typical examples of application have been reported, see e.g., benzoic acid and its derivatives in [60]. An early general discussion of inhibition mechanisms included iron, addressed among other compounds also including aromatic ones and their electronic properties [61]; further compounds and mixtures were studied later [62]. As a result, for a given group of amino- and thiobenzenes, relative inhibition efficiencies were estimated. In a follow-up attempt searching for correlations between molecular properties and inhibition efficiencies, high-resolution NMR spectra of some amine inhibitors in a cyclohexane solution were recorded [63]. A correlation between the chemical shift of the amine protons and inhibition efficiencies was noticed and tentatively related to electron density at the nitrogen and subsequently adsorptive interaction with the metal surface. On the molecular structure level, substituents and their relative position at an aromatic ring were used to explain the observed NMR shifts. Correlations between chemical shifts in ^{13}C and ^1H NMR spectroscopy with four aromatic

heterocyclic copper corrosion inhibitors were studied [64]. Conclusions were diffuse; apparently, the method may just help to distinguish effective from non-effective compounds. A similar study focusing on ^{15}N unfortunately yielded a similarly diffuse output [65].

In the following overview, proposed and investigated compounds are presented. Relevant data and results, in particular inhibition efficiencies expressed as a percentage of the corrosion current with/without inhibitor present and associated inhibitor concentrations, as well as solubility and stability data are reported as far as available. Obvious statements like increasing inhibition with increasing inhibitor concentration are not repeated; the same applies to the practically always-communicated temperature dependency of inhibition efficiency. In cases where the authors provide any argument for the selection of a particular molecule or its synthesis, these are specifically mentioned; quite obviously, such considerations elsewhere called rational design are highly unusual in corrosion research. For a typical example marred by even more flaws and riddles, see [66]. Compounds are organized following the identity of the main building block, i.e., substituted benzenes first, followed by six-membered heteroatom-containing compounds, and finally, five-membered aromatic rings. The majority of reported aromatic corrosion inhibitors have been studied with respect to their corrosion inhibition efficiency on iron and its alloys; there are also examples for other metals like copper and aluminum. Examples not pertaining to iron and its alloys (Designations of alloys are apparently not stated in an internationally accepted format, sometimes they are just called by naming the majority metal. To avoid confusion the term alloy is always inserted when required irrespective whatever in the original report has been stated.) are collected at the end of the respective section or subsection dealing with a specific class of aromatic compounds, when possible. An overview of corrosion inhibitors for copper and brass is available [67].

Several literature searches executed in preparation of this review, always using the term “aromatic”, certainly and most unfortunately must have failed to identify any reports on such compounds wherein the term was mentioned nowhere in the title, abstract, or keywords. In many cases, rather complicated new molecules have been synthesized without providing any argument for this approach. Similarly disturbing is the observation that differences in inhibition efficiency were noticed without even the slightest attempt to understand and to explain them. The plain fact that in most studies, no reason is provided for the synthesis of further molecules considered to be corrosion inhibitors (not always the outcome of the experimental study) is equally surprising. Because corrosion is always the combination of anodic metal oxidation and dissolution combined with a cathodic reaction, either being the electroreduction of dioxygen or the reduction of protons from the corrosive environment, the presence or absence of dioxygen in a corrosive environment may be relevant in case an inhibitor influences these cathodic processes in distinctly different ways. Accordingly, a statement in the experimental part of a report stating clearly whether solutions have been saturated with air or (less frequently) plain dioxygen or with an inert gas would be welcome. The majority of studies pertain to the latter case, but even in an article with a title suggesting a study with aerated solution [68], this relevant detail is nowhere mentioned in the experimental part.

Inhibition of stress corrosion cracking of stainless steel by aromatic and heterocyclic inhibitors has been studied [69]. The use of corrosion inhibitors in self-healing protective coatings on magnesium alloys has been addressed [70]; aromatic ones performed slightly better, although the reasons were not specified. Organic acids as corrosion inhibitors in engine coolant fluids have been reviewed [17].

The influence of several inhibitors including aromatic ones on current oscillations has been studied [71]. Corrosion of aluminum by methylene chloride has been studied [72]; aromatic compounds did not show particularly high inhibition effects, possibly due to the completely different reaction mechanism. Corrosion protection for a magnesium alloy possibly of interest in the automotive industry was studied [73]. From the wide selection of inhibitors including aromatic ones, a mixture of sodium phosphate and dodecylben-

zenesulfonate was finally identified as being best, for reasons not exactly revealed to the reader.

In a wide-ranging study of the influence of chemisorbed organic monolayers on the oxidation of electrode surfaces, a sequence of strength of chemisorption with aromatic compounds featuring prominently was reported [74]. Since the studied metals were all noble metals, the conclusions may be of limited value with respect to corrosion of non-noble metals.

An attempt to correlate electronic structure and inhibition efficiency was reported for mild steel in acidic solutions [75]; see also [76]. In an earlier study, these authors noticed a correlation with the HOMO-LUMO energy difference [77]. Now, as an additional parameter, planarity of the inhibitor molecules is taken into account. Planar molecules—and these are frequently aromatic—protect better than saturated ones (which are frequently twisted, i.e. non-planar). A critical reexamination of suggested correlations between many electronic molecular parameters and corrosion inhibition efficiency was provided [78]. The conclusion, based on a consideration of a significant number of empirical examples, was devastating; no correlations could be verified.

The briefly reviewed considerations basically follow an approach commonly known as quantitative structure–Activity relationships (QSARs) [79]. A similar study looking for structure–activity correlations with aluminum as the subject was conducted [80].

1.1. Substituted Benzenes

Benzene itself is practically immiscible with aqueous solutions at all pH values. Required solubility must be afforded by suitable substituents, preferably with heteroatoms like in amino or carboxylic groups. The anionic surfactant dodecylbenzenesulphonate showed considerable inhibition with pure aluminum and some of its alloys, up to 95% at 10 mM concentration with significant agreement between the results of weight loss, impedance, and PPM measurements [81]. Although the free enthalpy of adsorption was low, suggesting physisorption only, the typical surfactant-like behavior may have contributed to this efficiency. Aromatic dicationic surfactants with various chain lengths have been tested as corrosion inhibitors for carbon steel; up to 97% inhibition at 250 ppm inhibitor concentration was found at an intermediate chain length [82]. The quaternary ammonium sites, heteroatoms, and the aromatic electron system certainly contributed to adsorption and inhibition, but the authors remained silent on this. Substituted ammonium cationic surfactants were inspected for the influence of wettability and molecular structure [83]. Somewhat confusingly, inhibitor coatings—in a slight deviation from the commonly established meaning of inhibitor in corrosion science—are mentioned; the statement “the synergistic inhibition effect and the anticorrosion efficiency” remains completely mysterious.

Benzene para-substituted with various groups and with S-CH₂OCH₃/ Se-CH₂OCH₃ showed generally higher inhibition with the selenium compounds, although reasons were not provided [84,85]. Several substituted benzoic acids were compared as corrosion inhibitors for aluminum in alkaline solution [86]; *m*-bromo benzoic acid performed best at 500 ppm concentration, with 81% inhibition.

Nitrones with two substituted phenyl rings were tested as corrosion inhibitors for mild steel in organic acid media at inhibitor concentrations ranging from 50 to 150 ppm [87]. Although no inhibition efficiencies were stated, it could be derived from the displayed figures that in formic acid at 750 μM inhibitor concentration (different from the numbers stated in the text), efficiencies reached with the best inhibitor were 93%, in acetic acid 81%. Chalcones with further azo linkages were tested as corrosion inhibitors [88]; the highest inhibition (87% at 400 ppm inhibitor concentration) was observed with a nitro-substituted chalcone.

In a report on corrosion inhibition by an inclusion product of dibenzylthiourea DBT (more precisely 1,3-dibenzylthiourea) with hydroxypropylated cyclodextrins, solubility issues overcome by the inclusion were stated as a reason for this added complication [89]. Surprisingly, such solubility issues had not been observed by earlier authors (see e.g., [90]),

and some of them were conveniently ignored in the report. Somewhat surprisingly, later in the report, inhibition by plain DBT was reported (despite the claimed solubility problems) with an efficiency of 85%, only slightly lower than with the inclusion material at 92%. There are more inconsistencies in the report. Elsewhere, a group of 1-benzoyl-3,3-disubstituted thiourea derivatives was studied for corrosion inhibition performance [91]. Solubility issues were not mentioned. The highest inhibition of 97% was found at 100 ppm inhibitor concentration for 1-benzoyl-3,3-dibenzylthiourea. The fairly diffuse explanation of both physisorption and chemisorption being operative and an unspecified correlation with aliphatic and aromatic substituents is not really helpful. Upon closer inspection, it appears that benzyl instead of phenyl substituents are more flexible, enabling better adsorptive interaction with the metal surface. This is supported by the larger absolute value of ΔG_{ad} .

A mixture of cationic ammonium surfactants synthesized from reformat (a petroleum fraction) provided corrosion inhibition by adsorption for carbon steel in aqueous 1 M HCl [92].

Among several amides and amines studied for hydrogen sulfide corrosion protection, aromatic benzylpropanediamine performed best [93]. Disubstituted aminobenzenes had been studied earlier [94]; selected compounds are depicted in Figure 2.

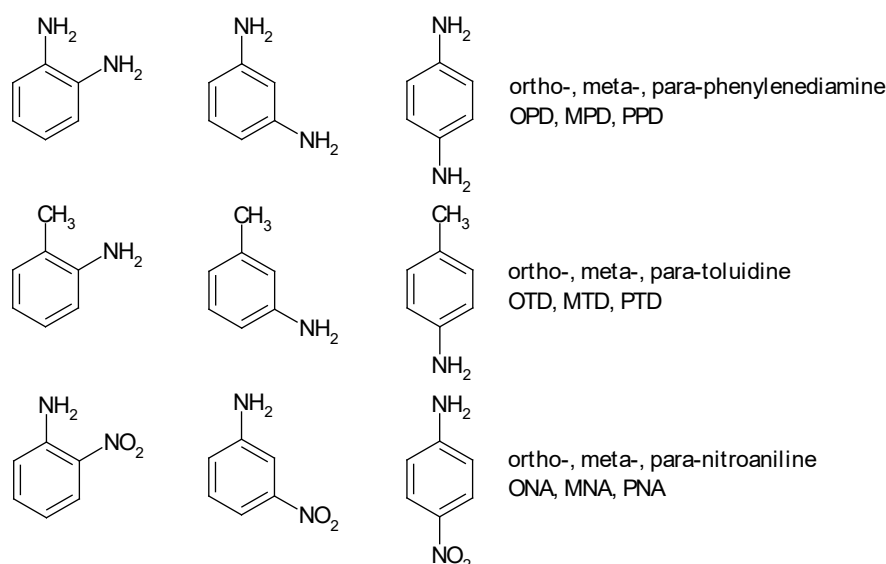


Figure 2. Disubstituted benzenes studied by Babu et al. [94].

The amino group was protonated in the acidic 1 M HCl-electrolyte solution used in this study. According to Hackerman and Makrides [95], deprotonation happens upon adsorption; thus, the amines are adsorbed instead of the ammonium ions. Similar observations have been reported for e.g., the adsorption of aniline on various metals from acidic solutions [96,97]. This differed from a report on the inhibition activity of 18 aromatic amines on mild steel, wherein adsorption of the ammonium (i.e., anilinium) ion via the nitrogen atom of the protonated amino group was claimed [98]. Depending on the actual electrode potential of the studied metal and the composition of the electrolyte solution, adsorption of e.g., chloride ions may happen. On top of this adsorbate layer, anilinium ions were reported to be adsorbed in a rather perpendicular orientation based on spectroelectrochemical evidence; the surface Raman spectrum showed mostly in-plane vibrational modes [49]. Contributions from the aromatic ring system(s) were not invoked; instead, the surface area actually covered by an adsorbed molecule was stated as the main influence, with larger areas affording better protection. Blomgren and Bockris instead suggested adsorption of the ammonium ions, with the interaction between π electrons of the aromatic ring system providing the driving force [99]. Results based on Tafel evaluation (see below and [100,101]) suggested the highest inhibition efficiency of 90% for OPD at

12 mM concentration. This compound also performed best at the lowest studied concentration of 2 mM. Unfortunately, earlier predictions [61] regarding a higher efficiency of *o*- than *m*-toluidine could not be verified. Specific health risks associated with *o*-toluidine have been discussed in detail [102]. Neither was the predicted [41,42] particularly poor performance of nitro-compounds noticed. In subsequent studies aimed at verification of these results via further standard techniques (linear polarization resistance and impedance measurements), the concentration effect for PPD was verified [103], while the substituent effects suggesting PPD as the most efficient among the para-substituted compounds was confirmed in [104]. In both studies, results obtained with the Tafel approach did not agree very well with those of the other methods. In a comparison of six *o*-substituted anilines as corrosion inhibitors for copper, *o*-ethylaniline performed best [105]. This was attributed to the higher repelling power of the ethyl group in comparison to the other substituents. In an earlier study with iron and a slightly different selection of substituted anilines, 2-ethoxyaniline performed best [106]. This was ascribed to the presence of oxygen in the second substituent enhancing adsorption and thus protection. Substituted anilines were examined as corrosion inhibitors for an Al-Mn-alloy in phosphoric acid [107]. Plain aniline performed best; adsorptive interaction via the electrons at the amino nitrogen and the aromatic π electrons was suggested as the mode of action. At low pH values, some substituted anilines performed better than aniline. Isomeric toluidines were examined as corrosion inhibitors for aluminum alloy 3S in chloroacetic acid [108]. *m*-toluidine performed slightly better than the other compounds, and this was also observed with aluminum alloy Al-57 S [109]. Several *p*-substituted aromatic amines were compared as inhibitors for aluminum alloy 1060 [110]. *p*-amino benzoic acid performed best because of the electronic effect of the carboxylic acid in the *p* position. In a comparative study of aliphatic and aromatic amines as inhibitors for aluminum 2S alloy, it was noticed that ternary amines with an ethyl instead of a methyl group showed higher inhibition effects [111]. Further effects of alloy ingredients in another aluminum alloy were examined with these inhibitors; no significant effects of the alloy ingredients were reported [112], and results were corroborated with a further study with a wide selection of aromatic and non-aromatic inhibitors that also included corrosive aqueous alkaline solutions [113]. Secondary aromatic amines (actually *N*-substituted *p*-toluidines) were examined as copper corrosion inhibitors [114]; a methyl group in the 5 position of the 2-furfuryl substituent yielded the most efficient inhibitor. Whether type and position of the substituent had any effect of the electronic properties of the furfuryl ring and its contribution towards adsorption on the copper surface was not addressed. In aqueous extracts from cigarette butts containing many other ingredients, aromatic amines showed high levels of protection [115]. Results for further, more highly substituted amines were reported in a study ignoring the state of the art [116], in which 5-nitrobenzene-1,2,4-triamine performed best, providing 94% inhibition at 10 ppm inhibitor concentration.

Epoxidized linseed oil modified with aniline and various substituted anilines was tested for corrosion protection of mild steel in solution and in the gas phase [117]. Inhibition efficiencies were as high as 86 to 95% at concentrations of 100 ppm in the gas and 50 ppm in solution. *p*-anisidine-modified material performed best, and efficiency correlated with electron density on the nitrogen atom, as derived from NMR measurements. Since protection proceeded via adsorbed molecules, this suggested a relation between electron density and adsorption. The electronic effect is based on the conjugated electron system of the aromatic ring. Chemically related epoxidized soybean fatty acids/*p*-substituted aromatic amines have been suggested as corrosion inhibitors in UV-curable steel coatings [118].

In a study with steel QD36, *p*-anisidine showed the highest inhibition efficiency, greater than that of aniline and *p*-toluidine [119]. For copper in nitric acid thioglycolic acid, *o*-anisidine and quinoline showed very similar inhibition efficiencies [120]. Why quinoline was called a weak inhibitor when it performed better than *o*-anisidine remains mysterious. Some corrosion inhibition by *o*-toluidine (the authors call it "ammonium iron" and "2-amino-1-methylbenzene") with steel C35 in aqueous 0.5 M H₂SO₄ was observed [121].

Anisidines and phenylenediamines were compared as corrosion inhibitors for mild steel in aqueous HCl [122]. *p*-phenylenediamine performed best among its isomers, as confirmed in [104], and *m*-anisidine was most effective among its isomers; there is no straight disagreement with [119] because in that report, only *p*-substituted compounds were compared. Condensation products (i.e., Schiff base, discussed further below) of *p*-phenylenediamine and various aromatic aldehydes were studied as corrosion inhibitors for mild steel and oil-well steel at 105 °C [123]; 2,4-dicinnamylidene aminophenylene (Figure 3) performed best at 5000 ppm inhibitor concentration, which was attributed to the conjugated double bonds in the linker groups. Flat or linear structures in turn enabled by the aromatic building blocks also support good adsorption.

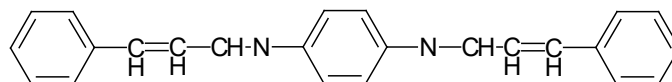


Figure 3. 2,4-dicinnamylidene aminophenylene.

In a similar report by the same authors, further condensation products of thiosemicarbazide and aromatic aldehydes were examined, and 1-cinnamaldehyde thiosemicarbazide was most effective [124]. No attempt was made to explain this finding.

The corrosion inhibition properties of aniline unit-containing epoxy monomers with either an oxygen or a sulfur linkage at the aromatic ring have been examined [125]. The difference in efficiency, already small in the Tafel evaluation and even smaller (about 1 percentage point) from the impedance measurements, was explained with the results of the very extensive theoretical calculations. For further details, see the companion reports [126–129], and for similar examples by the same authors, see [130,131]. Inhibition by another epoxy polymer was studied [132]. The inhibition by the fraction of the prepared polymer soluble in the corrosive medium was examined, and this solubility was found to be in the micro- to millimolar range. At 1 mM, potentiodynamic polarization yielded 98% inhibition. Why a value of 96% found with the impedance measurements was better remains mysterious.

Inhibition by four amines with two differently functionalized phenyl substituents was reported [133], and the compound with a nitrile substituent performed best. Although theoretical data suggest flat adsorption of the molecules on an idealized Fe(110) surface, steric hindrances seem to limit the interaction between the two aromatic ring systems and the substituents with the surface, given the moderate efficiency values. Aminostyrene provided only moderate protection [134]. Compared with plain aniline, the vinyl group provided some improvement, possibly due to enhanced adsorption supported by the additional unsaturated bond. Among several substituted quinolin-6-amines, 7-(1-vinylhex-5-en-1-yl)quinolin-6-amine performed best with 97% inhibition at 0.2 wt.% inhibitor concentration [135]. The performance was better by about 10% compared with that of aminonaphthalene, which was attributed to extended substituents with double bonds supporting stronger adsorption.

Inhibition activity of several amines for zinc in aqueous HCl has been studied; beyond general observations like a lower rate of corrosion at higher inhibitor concentration, no conclusions have been reported [136]. Corrosion protection for brass was studied, with a wide variety of substituted anilines supporting the applied cathodic protection [137]. Significant effects of applied cathodic potential protection on the actual efficiency of inhibitors were noticed, some corrosion accelerators turned into inhibitors, and vice versa; these effects were explained with inhibitor adsorption as a function of electrode potential. Inhibition of brass corrosion in aqueous nitric acid without externally applied potential was compared for several aromatic amines [138]. Anthranilic acid protected best; this was attributed to the electronic effects of the substituents that in turn influenced adsorption on the metal surface. Protection of brass by several aromatic amines without applied cathodic protection has been compared [139]. The protection works in a very different way by inhibiting formation of nitrous acid which in turn attacks the alloy. *o*-substituted compounds like *o*-chloroaniline perform best because they accelerate the diazotization rate.

Adsorption of the amine may play a role also. Particular effects of the aromatic moiety were not addressed. In a further study of protection for brass with aromatic amines, *o*-anisidine, *o*-toluidine, and *o*-phenetidine were compared [140]. With a longer substituent chain, the inhibition increased.

Corrosion inhibition on nickel by several amines including three aromatic ones was studied, although the results did not suggest specific effects of the aromatic moieties [141].

In a theoretical study of the corrosion protection provided by a few *o*- and *p*-substituted anilines, higher protection by *p*-substituted ones was claimed based on calculated differences in dipole moments that were hardly visible with the small selection of compounds [142]. A further substituted aniline 3-amino-2-methylbenzylalcohol has been tested as a corrosion inhibitor for mild steel [143]. Inhibition of 50% at 500 ppm inhibitor concentration was not spectacular and deteriorated after 24 h, and addition of up to 5 mM NaI increased this value to 80%. Although further results of theoretical considerations were presented, the reason for the beneficial effect of the iodide ions was not mentioned. Presumably, iodide is adsorbed (like chloride in similar systems) first and provides interaction sites for increased inhibitor adsorption. Quaternary ammonium salts with a palmitic acid chain and ethoxylated aniline and two further $-\text{CH}_2\text{CH}_2\text{OH}$ -rests provided 98% inhibition at 200 ppm inhibitor concentration, according to a hard-to-understand report [144,145]. Experimental methods for determination of quaternary ammonium salts have been developed [146].

Protective properties of oligomers of several alkyanilines with formaldehyde have been compared [147]. These are soluble in hydrocarbons as encountered in refineries. They performed better than the respective monomers, and oligomers of 4-vinylpyridine also performed better than similar monomers [148]. This may have been due to stronger adsorption of the oligomer and thus, more extensive coverage of the metal surface. Oligomers with anilines having long-chain alkyl substituents showed up to 99% inhibition at 100 ppm inhibitor concentration. Polyaniline as well as polymers prepared from substituted anilines (see e.g., poly(*p*-toluidine) in [149] and further polyheteraromatic samples in [150]) including copolymers have been examined as protective coatings; these materials are beyond the scope of this report. In a comparison of the polymers of isomeric toluidines applied in solution with a mild steel electrode at concentrations ranging from 100 to 1000 ppm [151], poly-*p*-toluidine showed the comparatively highest protection at 69%. Possibly, given the poor solubility of intrinsically conducting polymers, only some oligomers were dissolved. Coatings, i.e., paints, containing these compounds (for an example, see [152], for an overview on coatings, see [39]) certainly utilize the aromatic entities within them for enhanced protection once these groups are close enough to the metal surface to be protected. Although many of these intrinsically conducting polymers (ICPs) can be deposited electrochemically, coating with chemically prepared ICPs appears to be more effective because the coatings are homogeneous and are not more or less rough or cauliflower-like [150]. A sulfonated and thus water-soluble aromatic polyamide has been proposed as a corrosion inhibitor for copper, because of a claimed much better film-forming capability [153]. At 500 ppm inhibitor concentration, an inhibition efficiency of 92% was found, not better than values reported with molecular inhibitors. In a similar approach, two chemically different polyesters containing aromatic substituents, being presumably water soluble (not stated in the report), was tested as a corrosion inhibitor for steel rebars [154]. Depending on the actual chemical composition of the polyester, inhibition efficiencies up to 64% at 1000 ppm inhibitor concentration were reported. For a very similar further example by the same researchers, see [155]. Apparently very water-soluble polyester derivatives were studied as carbon steel corrosion inhibitors [156], and the phthalate-based compound was more efficient than the maleate-based one. How π electrons in the benzene rings of the maleate-based (!) compound helped in adsorption remains a mystery; in the case of the phthalate-based compound, they certainly contributed to the better performance. Further polyesters based on the indazole moiety provided significantly higher protection ranging from 86 to 97% [157].

In a study dealing with hydroquinone with various groups and hydrophobic motifs as an inhibitor, *N*-[6-(4-propargyloxyphenoxy)hexyl]-*N,N*-dimethyl-*N*-dodecylammonium

bromide provided 97% inhibition at 15 ppm concentration [158]. Alkyne, hydroquinone, quaternary ammonium, and a hydrophobic alkyl chain on one molecule were claimed as reasons for the performance. Three diquaternary ammonium bromide surfactants of the general structure shown in Figure 4 provided inhibition efficiencies up to 99% at 200 ppm concentration, showing significant decay within a few days [159].

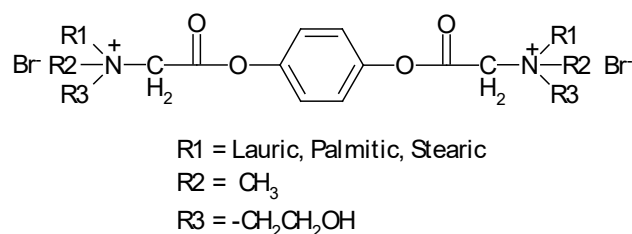


Figure 4. Diquaternary ammonium bromides.

Plain 1,4-naphthoquinone as a corrosion inhibitor for aluminum was tested in both aerated and de-aerated 0.5 M NaCl solutions [160]. Inhibition efficiencies or equivalent results cannot be found in the report. Chromeno naphthyridines (see Figure 5), being rather low in toxicity, have been suggested as corrosion inhibitors, reaching 98% inhibition at 300 mg·L⁻¹ inhibitor concentration with R = -CH=CHC₆H₅ [161]. The numerous heteroatoms and the extended aromatic π -electro system suggest a flat orientation of the adsorbed molecule, also concluded from molecular dynamic simulation.

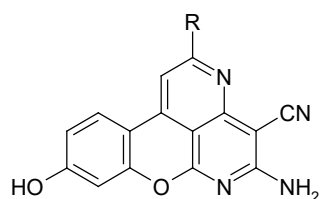


Figure 5. Chromeno naphthyridines.

Aromatic aldehydes were compared as corrosion inhibitors for aluminum alloy Indal 57S in HCl solution, showing rather similar efficiencies and no particular evidence regarding specific effects of the aromatic moiety present in all of them [162]. In a study with an alkaline solution of 1 M NaOH, aromatic aldehydes performed better, which was attributed to the π electrons of the aromatic groups [163]. A correlation between inhibition and molecular dipole moment claimed elsewhere [142] was explicitly excluded. Although the aromatic system was identified as the major cause of the much better efficiency of the aromatic aldehydes, double bonds in substituents were obviously not helpful (as claimed elsewhere), and this was tentatively attributed to extended conjugation reducing electron density in the aromatic system, possibly weakening adsorption. Corrosion inhibition by several aliphatic and aromatic aldehydes for aluminum–manganese alloys in aqueous KOH solution was evaluated, and salicylaldehyde performed best [164]. Adsorption via the oxygen of the aldehyde group was proposed, but the obvious beneficial effect of an aromatic ring was not addressed. Aromatic aldehydes were compared as corrosion inhibitors for 70/30 brass [165]. At fairly high inhibitor concentrations of about 2 mL per liter of electrolyte solution, benzaldehyde was slightly more efficient. In a comparison of several aldehydes for corrosion inhibition on mild steel, formaldehyde performed better than benzaldehyde because it formed a protective polymer film [166]. Compared with the other non-polymerizing aliphatic aldehydes, the much better inhibition by benzaldehyde was attributed to the presence of the aromatic system in addition to the carbonyl function further enhancing adsorption and thus inhibition. Two aromatic aldehydes with an azo linkage connecting a further aromatic system were compared, with a methyl instead of a nitrile substituent providing a higher inhibition of 93% at 200 μ M inhibitor concentra-

tion [167]. This may have been due to the poorer chemical stability of the latter compound, with the nitrile group undergoing hydrolysis, yielding a less effective carboxyl group.

Aluminum alloy 56S corrosion protection by azomethines (i.e., Schiff bases) derived from aromatic aldehydes has been compared [168]. $-OCH_3$ -substituted compounds performed better, while all compounds were highly efficient and much better than the corresponding amines. Results obtained with *t*-cinnamaldehyde (Figure 6) and steel yielded 99% inhibition but were only at first sight at variance regarding the detrimental effect of a double bond in the substituent [169]. Closer examination revealed a film-forming reaction as the mode of protection, instead of adsorption as assumed in the preceding aldehyde-related examples. Nevertheless, adsorption played a significant role in keeping the film attached to the steel surface.

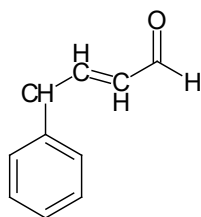


Figure 6. *t*-cinnamaldehyde.

Benzoic acid and ω -benzoyl alcanoic acid were compared as corrosion inhibitors for steel [170]. The presumed effects of further anions from the electrolyte solution on the inhibition effects of carboxylic acids were studied [171], and a ranking taking into account the stability of formed passivating films was suggested, with perchlorate affecting the stability least and sulfate being most detrimental. The influence of iodide in the corrosive medium on corrosion protection by various aromatic carboxylic acids has been studied theoretically and experimentally [172]. A small improvement was observed with iodide ions present; presumably, adsorbed halide anions enhanced inhibitor adsorption by the aromatic ring system as well as the carboxylate group. It is noteworthy that the iodide already provides substantial inhibition.

A much wider selection of aliphatic and aromatic carboxylic acids as inhibitors of the corrosion of aluminum has been studied [173]. Neither a clear ranking of inhibition efficiencies nor an explanation for the observed differences were provided. Further related compounds, described in a somewhat unusual terminology as aromatic acid derivatives, have been examined for the same purpose [174]. Rankings were proposed, but given the somewhat unsystematic mix of compounds, the absence of an explanation is no surprise.

Corrosion of an aluminum–copper alloy in phosphoric acid by substituted anilines was inhibited best by *p*-toluidine [175]. This was attributed to both resonance and the inductive effects of the amino and methyl substituents. With increasing acid concentration, the efficiency decreased as presumably, the protonated form of the inhibitor was adsorbed less strongly. The same result has been reported elsewhere [176].

Nitro-substituted aromatic compounds hardly protective to iron and its alloys showed high inhibition efficiency with titanium [177] (for a further example, see [178]), due to the different mode of protection by passivation of the surface (instead of inhibition of electrode reactions). For strange reasons, this conclusion escaped the latter authors who carefully ignored the earlier report. A review of oxide formation on titanium and the conceivable effects of inhibitors has been published [179]. Nevertheless, a general conclusion regarding the lack of inhibition properties of nitro-substituted compounds in the protection of steel is not justified, as demonstrated in the study of a selection of different aromatic nitro compounds [180]. A protection mechanism invoking the formation of metal–inhibitor complexes different from simple inhibition by adsorption of an inhibitor is possibly the explanation. Unfortunately, this suggestion is somewhat disturbed by a concluding statement in the report claiming plain adsorption as the operating mechanism.

Schiff bases incorporating methoxyphenyl substituents have been tested as corrosion inhibitors for mild steel [181] and copper (up to 99% inhibition at millimolar inhibitor concentrations) [182]. By comparison of the studied molecules and taking into account results of theoretical calculations in the former case, the better performance of the inhibitor with an additional aromatic unit was attributed to the enhanced adsorption due to this presence. Further aromatic Schiff bases have been compared, although structure–efficiency correlations were not reported [183–189], and while further examples of moderately [190–195] as well as highly efficient compounds have been presented [196–203], these reports are sometimes purely theoretical without any experimental verification. In a further study, much higher efficiencies with mild steel were already observed at low concentrations of 40 ppm [204]. In a study with copper and copper alloys, Schiff bases prepared from salicyl aldehyde and aliphatic or aromatic amines were compared; unfortunately, in the report, only data pertaining to aromatic amines can be found [205]. Schiff bases with aliphatic amines accelerated corrosion (possible a reason for dismissing them in the report) because of their decomposition in water, yielding effective corrosion stimulators, whereas those with aromatic amines turned out to be stable [206]. Those with –SH moieties were more efficient, possibly because of the capability of this moiety to interact more strongly with copper and its ions. A Schiff base 2-oxo-3-hydrazonoindoline showing structural features of hydrazones and indoline (a relative of indole, see above) provided 92% inhibition for corrosion of mild steel at 500 μM inhibitor concentration [207].

Triazole-based Schiff bases were particularly efficient, reaching 95% inhibition at 25 ppm concentration [208]. Attempts to correlate inhibition efficiency of Schiff bases obtained from *p*-anisaldehyde and *p*-substituted anilines with solvatochromic parameters remained as inconclusive as the attempt with Hammett parameters [209]. Inhibition efficiency of 90% at 90 ppm inhibitor concentration for the Schiff base (E)-N-(2-chlorobenzylidene)-2-methylaniline on steel was attributed to the aromatic moiety and the azomethine group [210]. There appears no rational connection between the many calculation results of theoretical considerations and observed inhibition. Schiff bases of the type *m*-substituted aniline-*N*-salicylidenes (see Figure 7) are very efficient in terms of corrosion protection (99% and better at the highest concentration of inhibitor) for zinc [211]. The high efficiency was attributed to the salicylidene part. An extended study of salen as a Schiff base dye included some considerations of its corrosion inhibition properties [212].

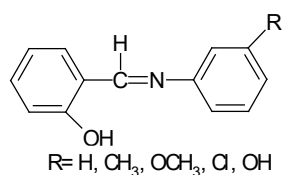


Figure 7. *m*-substituted aniline-*N*-salicylidenes.

Schiff bases prepared with L-lysine and aromatic aldehydes have been recommended as green corrosion inhibitors for mild steel [213]. To achieve 95% inhibition required 400 $\text{mg}\cdot\text{L}^{-1}$ inhibitor concentration. Also taking into account the results of theoretical considerations, adsorption on the chloride covered metal surface with electrons at the nitrogen atoms and the aromatic ring systems participating was suggested as the mode of operation. These inhibitors have been claimed to be green because they are less toxic or not toxic at all compared with many other Schiff bases. In addition, the present bases can be prepared from green starting materials. These considerations apply to many reported Schiff bases.

The sometimes indicated concern regarding the lower efficiency of Schiff bases due to hydrolysis in acidic solution was addressed in a study of a particularly hydrolysis-stable compound [214]. High efficiency for mild steel in aqueous 1 M HCl was attributed to adsorptive interaction of the imine moiety, the other heteroatoms, and the aromatic π

electron system, and no deterioration within 24 h was observed. Sometimes, addition of e.g., surfactants enhanced inhibition efficiency [197].

Two more Schiff bases have been synthesized and characterized; their corrosion inhibition performance was praised but was not even estimated, even less so experimentally verified [215].

Corrosion inhibitors for aluminum and its alloys have been reviewed [216]. Phenol was studied an inhibitor in an acidic solution, with a combination of experimental and theoretical methods showing at best 71% inhibition efficiency [217], confirming earlier observations with phenol and some substituted phenols [218]. Phenol and further aromatic hydroxy compounds have also been compared as corrosion inhibitors for zinc, used as a zinc pigment ingredient in corrosion-protection paint; for comparison, an aluminum pigment was included [219] (see also [220] for more on aluminum pigment). Several alkyl gallates (Figure 8), in particular octyl gallate, performed best; this applied also to the aluminum pigment. The capability to form chelate complexes with zinc ions was suggested as the relevant inhibitor property.

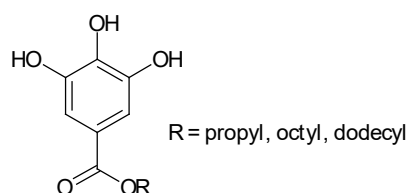


Figure 8. Alkyl gallates.

For the same purpose, substituted aromatic 2-hydroxyoximes were compared [221].

p-thiocresol showed significantly higher inhibition, possibly due to additional adsorption-enhancing interactions between the –SH moiety and the metal. The inhibition by benzene-1,2,4,5-tetracarboxylic dianhydride was very low [222]. In a comparative study of the inhibition efficiencies of some organic acids for aluminum in aqueous NaOH solution, it was observed that aliphatic acids interacted, i.e., adsorbed, via the carboxylic acid group whereas aromatic acids lay flat on the surface [223]. The former mode is less effective, resulting in poor inhibition, whereas the latter mode is much more effective, yielding a correspondingly much higher inhibition. Protection of aluminum alloy Al-57S in aqueous HCl by various aromatic amines (substituted anilines) has been compared [109]. Neither the effects of the type of substituent (methyl vs. chlorine) nor the position (*o*- vs. *m*- vs. *p*-) were correlated with the reported inhibition efficiency.

It was reported that 4-phenyl-3-thiosemicarbazide (Figure 9) showed only moderate inhibition activity with aluminum in 0.1 M HCl solution [224]. Because of the acidic solution, the nitrogen atoms in the inhibitor were protonated, leaving only adsorptive interaction with chloride ions already adsorbed on the metal. This interaction appears to have been relatively weak, as also expressed in the free enthalpy of adsorption with numerical values (depending on temperature) indicative of physisorption only.

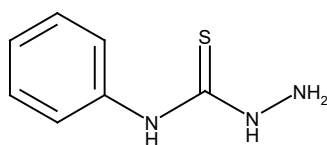


Figure 9. 4-phenyl-3-thiosemicarbazide.

Corrosion inhibition with aluminum by 3-(10-sodium sulfonate decyloxy)aniline and its water-soluble polymer has been examined [225]. At 10 ppm concentration, the monomer and polymer showed roughly the same inhibition efficiency at around 50%. Amides obtained from aromatic amines and the fatty acid 13-docosenoic acid were examined [226]. Inhibition efficiencies reached 97% at 500 ppm inhibitor concentration, and the aromatic

compounds performed better than aliphatic ones studied for comparison. Surprisingly, the expected influence of the long aliphatic chain was not addressed.

Two different aromatic systems have been combined in 1-(2-fluorophenyl)-2,5-dimethylpyrrole and 1-(2-fluorophenyl)-2,5-dimethylpyrrole-3-carbaldehyde (Figure 10), suggested as inhibitors for aluminum [227]. The higher efficiency of the carbaldehyde was noticed, although an attempt to explain it was not provided. Stronger adsorption caused by the additional oxygen-containing substituent may possibly have been the reason. Replacing fluorine with other halogens reduces the efficiency [228]. In one report, the higher efficiency provided by the carbaldehyde group was attributed to additional condensation on the electrode (formation of dimers); elsewhere, later authors claimed stronger adsorption instead [216].

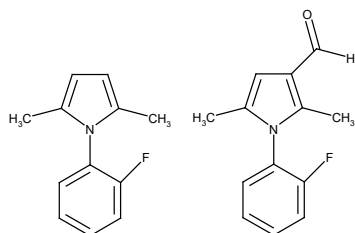


Figure 10. 1-(2-fluorophenyl)-2,5-dimethylpyrrole and 1-(2-fluorophenyl)-2,5-dimethylpyrrole-3-carbaldehyde.

A particularly efficient compound is 5-bromosalicylaldehyde ethanesulphonylhydrazone (Figure 11), yielding 100% inhibition at 10 ppm concentration [229]. Unfortunately, further experimental data do not enable a coherent interpretation of this effect, and in particular, of the apparently striking effect of the bromine substituent; with chlorine instead, efficiencies drop to values around 92%. Attribution of this difference to the higher molecular weight of the former compound remains unclear.

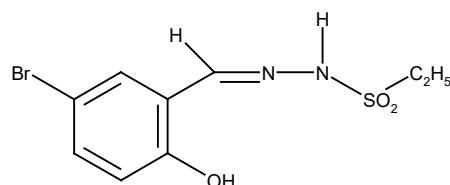


Figure 11. 5-bromosalicylaldehyde ethanesulphonylhydrazone.

Hydrazones with a furfural moiety at one end and benzene or a substituted benzene at the other were tested as corrosion inhibitors for C-steel [230]. At micromolar concentrations, inhibition was poor to moderate. The claimed influence of substituent position (*o*- vs. *p*-) at the benzene end in a fragmentary final statement remains mysterious, because no comparable data were communicated. Further aromatic hydrazones with inhibition properties less impressive than the examples mentioned above have been synthesized [231,232]. This also applies to three other hydrazone derivatives, showing slightly inferior inhibition of up to 95% at 5 mM inhibitor concentration, despite the considerable numbers of three aromatic rings and several heteroatoms (see Figure 12) [233]. The confusion between hydrazides and hydrazones prevailing in that report can easily be resolved by consulting a handbook of chemistry.

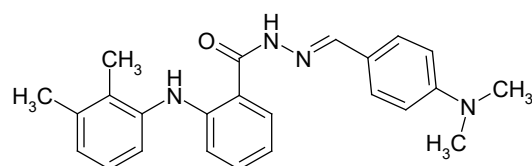


Figure 12. N'-[(E)-4-Dimethylaminobenzylidene]-2-(2,3-dimethylanilino)benzohydrazide.

The aromatic hydrazide derivative 2-(3,4,5-trimethoxybenzylidene)hydrazinecarbothioamide tested as a corrosion inhibitor for mild steel provided 90% inhibition at 0.8 mM inhibitor concentration [234]. Given the number of heteroatoms in the molecule and the presence of an aromatic system, the speculation of the authors about adsorptive interactions involving these contributions appears to be reasonable. Further hydrazides with aromatic moieties have been compared [235], with the highest inhibition of 94% at 500 ppm inhibitor concentration of cinnamic acid hydrazide. A further 2-hydroxy-*N'*-((thiophene-2-yl)methylene)benzohydrazide with even better 98% inhibition at 400 μM inhibitor concentration has been reported [236]. Slightly lower inhibition was observed with another benzohydrazide derivative [237], and much lower inhibition of 71% at 1 mM inhibitor concentration was provided by 4-hydroxyl-*N'*-[(3-hydroxy-4-methoxyphenyl)methylidene] benzohydrazide [238]. In the absence of any explanation in the report, it may be assumed that the additional double bond in this compound provided a major boost to adsorption and inhibition, better than the amino group in the much poorer performing anthranilic acid hydrazide. Further benzohydrazide derivatives have been evaluated, yielding in the best case 92% inhibition at 150 μM inhibitor concentration in the presence of 10^{-2} M added KI [239]. According to the authors, added iodide adsorbed on the metal surface enhances cationic species adsorption, although which cationic species are meant remains open; possibly, the inhibitor molecules can be protonated, yielding cationic species. Corrosion inhibition by two sulfonohydrazide derivatives has been studied; unfortunately, the benefit of the added sulfonyl group was not discussed and neither was the claimed strong correlation between theoretical and experimental data evident [240]. That report contained an overview of hydrazides as corrosion inhibitors. An extended theoretical study of many hydrazides, including a comparison with some Schiff bases, provided long lists of numerical values and calculated inhibition efficiencies but unfortunately, no experimental support [241]. The same limitations apply to theoretical considerations of “new possible biobased gemini corrosion inhibitors derived from fatty hydrazides” [242].

Phenylcarbomodithioate suggested as an inhibitor provided 86% inhibition at 500 ppm concentration [243].

In a comparison of corrosion inhibition on steel by several sulfoxides, several aromatic ones (see Figure 13) were included [244].

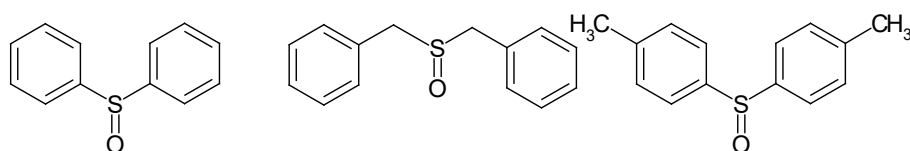


Figure 13. Aromatic disubstituted sulfoxides.

These compounds act as secondary inhibitors, i.e., the sulfides formed by their chemical conversion are the actual inhibitors. Because of conjugation effects of aromatic substituents the electron density on the sulfur is slightly lowered and adsorption thus weaker, but possibly electronic interaction between one of the two aromatic ring systems with the metal surface may contribute to stronger adsorption [244]. Nevertheless, alkyl-substituted sulfoxides are more efficient.

In aluminum–air batteries, inhibition of corrosion and hydrogen evolution are of considerable interest. Benzoic acid, isophthalic acid, and trimesic acid were compared [245]. Why benzoic acid performed best was not revealed.

Self-assembled monolayers (SAMs) have been suggested as possible corrosion protection [246–252]. Because of the high affinity of the frequently sulfur-containing molecules forming SAMs on copper, studies have focused on protection of this metal [253]. A comparative study of SAMs formed from benzenethiol, 2-naphthalenethiol, or 4-acetamidothiophenol (see Figure 14) copper reported some inhibition [254–256]. In another hard-to-follow report with numerous internal contradictions, inhibition of copper corrosion by SAMs of 2-naphthalenethiol, *p*-chlorothiophenol, and *p*-toluenethiol was compared [257]. Somewhere

in the report, all inhibition efficiencies were >99%, while elsewhere, much lower values were listed. The thiols were present in the electrolyte solution during the corrosion studies, although with an SAM, there is no need for this. A comparison with plain thiophenol was missing. Although SAM formation is hardly a simple reversible adsorption, the authors applied a “Langmuir thermodynamic kinetic model”.

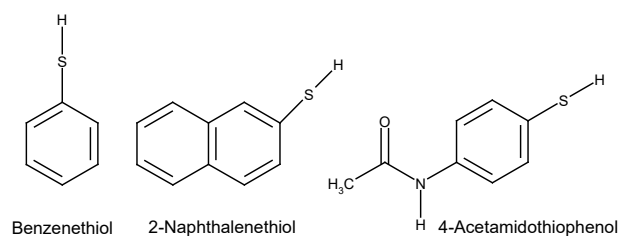


Figure 14. SAM-forming molecules studied for copper corrosion protection.

SAMs of aromatic aldehyde Schiff bases on carbon steel have been proposed as corrosion protection [258]. Inhibition around 93% was found. Even higher inhibition of 99% was provided by an SAM of the Schiff base 2-[(2-sulfanylphenyl)imino]methyl phenol [259]. In cases of Schiff bases proposed for corrosion protection of copper, some theoretical considerations have been reported [260]. The concept of SAMs has been expanded to ionic copolymers [261]. Aromatic moieties play basically the same role as with molecular SAMs regarding intermolecular interaction when organizing the SAM and thus, corrosion protection is not affected by them directly. Different from the molecules depicted in Figure 14, the 2-benzylthiopyrimidine BTP (see Figure 15) and its parent compound 2-mercaptopyrimidine that were used for the preparation apparently did not form SAMs [262]. Instead, both molecules presented a perpendicular adsorbate orientation with nitrogen and sulfur interacting with the iron surface. At 200 μM concentration of BTP, 99.8% inhibition was reported. Taking into account the value of free enthalpy of adsorption ($\Delta G_{\text{ad}} = -50.9 \text{ kJ}\cdot\text{mol}^{-1}$) a flat adsorption yielding better surface coverage and participation of one aromatic electron system and the N and S atom(s) appears to be more likely.

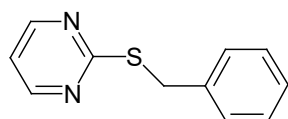


Figure 15. 2-benzylthiopyrimidine.

Tetraphenylphosphonium bromide was identified as a highly efficient inhibitor for zinc in aqueous 1 M HCl [263]. A highly complex mechanism of protection was suggested, while further experiments appear to be necessary to specify the role of the aromatic substituents. Similar high protection effects were reported for aluminum [264]. Overviews on corrosion inhibition by phosphoroorganic compounds are available [265,266]. Considerations of theoretical chemistry highlighting the influence of the electron density on the phosphorus atom and the contribution of aromatic electron systems are available [267]. Further phosphonium compounds of the type $\text{Ph}_3\text{PY}^+\text{X}^-$ with Ph = phenyl, X = Br⁻ or Cl⁻, and Y = propyl, propargyl, cyclopropyl, allyl, 1,3-dioxolanyl, or cinnamyl have been tested as corrosion inhibitors for mild steel in aerated acidic solutions [68]. Allyltriphosphonium bromide was found to be the most effective, but the difference to the other compounds was rather small except for tetrabutylphosphonium bromide—which was actually an accelerator. Improved protection after replacing a methyl substituent with an aryl substituent in dimethyl-aryl-telluronium cation was attributed to the increased number of adsorption centers and effective charge density on the heteroatom [268].

1.2. Substituted Heteroatom-Containing Six-Membered Rings

Heteroatoms like N, S, or P in an aromatic ring system have been frequently identified as adsorption-enhancing and thus inhibiting promoting options in aromatic six- and five-membered rings [269]. Numerous substituted pyridines have been examined as corrosion inhibitors for various types of steel; for examples, see [270]. Apparently, no correlation between molecular structure, type of substituent, or any other inhibitor property and actual inhibition efficiency could be found. Generally, it seems that whatever enhances adsorptive interaction will increase inhibition. DFT studies of several pyridine derivatives as corrosion inhibitors for aluminum yielded a perpendicular orientation of the adsorbed molecules via the nitrogen atom; for iron, a flat orientation involving the aromatic electron system is also conceivable [271]. A linear correlation between chemical (or electronic chemical?) potential and inhibition efficiency was found, and the same was noticed for the extent of charge transfer to the metal. In a comparison of inhibition efficiencies of pyridine, quinoline, and 1,10-phenanthroline (see Figure 16) with mild steel, the latter compound performed best with 80% at 10 mM inhibitor concentration [272]. This was attributed to the heteroatoms and the aromatic rings. Presumably, the more extended aromatic system of the latter inhibitor helped too. Corrosion protection by quinoline-based coatings has been studied [273]. A further comparison of these *N*-heterocycles and their *N*-hexadecyl derivatives as inhibitors for iron and steel was reported [274]. Better inhibition was found with a growing number of aromatic ring systems. The derivatives showed even higher inhibition, attributed to a blocking effect of the hydrocarbon chain hindering access of corroding species to the metal surface and to an increased electron density on the nitrogen atom enhancing adsorption. Optical measurements suggested a more dense structure of the protective layer on steel than on iron, possibly explaining the slightly higher corrosion resistance at the steel electrode.

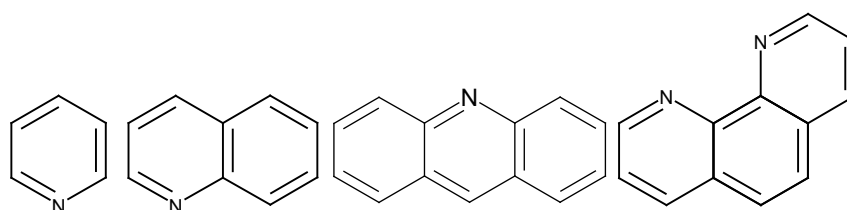


Figure 16. Pyridine, quinoline, acridine, and 1,10-phenanthroline.

Substituted *N*-decylpyridinium derivatives have been compared as corrosion inhibitors for iron [275]. Chemical identity of the substituent in position 3 influences efficiency; at room temperature, the hydroxyl group was best, and at 70 °C, the carboxyl substituent was most beneficial. Adsorptive interaction by the oxygen-containing substituents was suggested as the reason. A wide comparison of quaternary ammonium ions including substituted pyridines and quinolines as corrosion inhibitors for pure iron has been reported [276]. Inhibition increases with the number of aromatic substituents. Adsorption of 2-((3-methylpyridine-2-imino)methyl)phenol as a potential corrosion inhibitor for mild steel has been extensively described, although inhibition efficiencies have not been reported [277]. A large set of 2-amino-4-aryl-6-substituted pyridine-3,5-dicarbonitrile derivatives attractive because of their simple chemical synthesis was tested for corrosion inhibition, with only poor to moderate results [278].

In a comparison of corrosion inhibition on steel by several pyridine and pyran derivatives based on experimental as well as theoretical studies, 2-amino-6-(4-methoxyphenyl)-4-phenyl-pyridin-3-carbonitrile performed best [279]. The smallest HOMO-LUMO energy difference corresponds to the highest efficiency. In a comparative study of corrosion inhibition by several substituted pyridines, a beneficial effect of a vinyl substituent was noticed, as already seen for the respective substituted anilines [134].

In a theoretical study of three aminopyrimidine derivatives (presumably pyrimidine in the report means pyrimidine) [280], better protection by fewer aromatic compounds was concluded without any experimental evidence. A further pyrimidine derivative, 5-(4-

hydroxy-3-methoxyphenyl)-2,7-dithioxo-2,3,5,6,7,8-hexahydropyrimido [4,5-d]pyrimidin-4(1H)-one, provided 90% inhibition of CO₂ corrosion at 20 ppm inhibitor concentration [281]. Given the aromatic system and the many heteroatoms, the result surprises slightly. Furthermore, 6-phenylpyridazine-3(2H)-thione provided 96% inhibition for carbon steel in 3 M H₃PO₄ at 0.1 mM inhibitor concentration; this was presumably the optimum concentration, which the authors forgot to specify [282].

In a comparative study of quaternary amines with various aromatic moieties, naphthyl methyl quinolinium chloride performed best on carbon steel under H₂S exposure (sour corrosion) [283]. This was attributed to the two large (bicyclic) aromatic ring systems with associated higher electron density. Quinoxaline-based inhibitors with different substituents for a zinc–aluminum alloy [284] and for bronze [285] were prepared and compared. No significant differences between the four inhibitors were found in the former case; in the latter case, small differences apparently did not suggest speculations about structure–efficiency relationships. The status of quinoxaline derivatives as corrosion inhibitors has been reviewed [45].

In a comparative study of substituent effects on the corrosion protection capabilities of substituted pyridines, those with acceptor substituents were recommended as being most likely to be more efficient [286]. Three oxaldehydes (i.e., pyridoin, benzoin, and benzyl) were compared as corrosion inhibitors for mild steel [287]. Beyond the bland statement that these molecules are good inhibitors, the claimed advantages of the theoretical approach followed is noteworthy because it actually convolutes most of the experimental results with some calculated molecular properties, leaving common perceptions about the possibilities of theoretical methods rather disfigured. Several quinazoline derivatives have been synthesized, starting with benzoxazinone, and have been evaluated as corrosion inhibitors [288]. Protection was claimed; unfortunately, communication of any numerical result was overlooked. Several 1,3-benzoxazines have been compared as corrosion inhibitors and biocides [289]; the best inhibition reached 97% at 100 mg·L⁻¹ inhibitor concentration.

In a comparative study of N-heterocyclic aromatic amines, simple piperidine and piperazine were found to be more effective inhibitors than their substituted derivatives [290]. The better inhibition by piperazine (88% instead of 82% at the same inhibitor concentration) was attributed to the presence of two nitrogen atoms in the ring instead of only one. Heteroatom(s) always play a major role in adsorption and thus, inhibition. More moderately efficient piperazin-based inhibitors have been reported [291].

Several pyrazolopyrimidine derivatives (see Figure 17) have been studied as corrosion inhibitors for copper, with the mercapto compound yielding highest inhibition [292]. According to theoretical calculations, the smallest HOMO-LUMO energy difference corresponds to the highest efficiency.

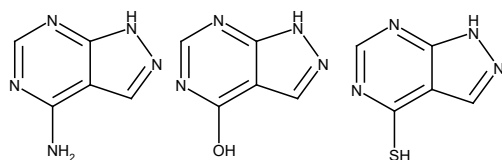


Figure 17. Pyrazolopyrimidine derivatives.

Further substituted pyrazolones have been prepared and characterized but not tested as corrosion inhibitors [293].

Schiff bases incorporating pyridine (see Figure 18) have been studied as corrosion inhibitors for mild steel [294]. At 10 mM inhibitor concentration, inhibition efficiencies were practically the same at around 99%, only at the lowest studied concentration was the chlorine-substituted compound slightly better, presumably because of somewhat stronger adsorption due to the chlorine substituent.

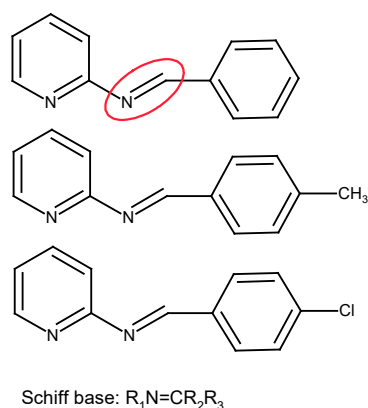


Figure 18. Benzylidene-pyridine-2-yl-amine and substituted derivatives; the structural motif of a Schiff base is highlighted.

Schiff bases (for examples as studied in [295], see Figure 19, for further examples, see [296–304]) have been studied as corrosion inhibitors for aluminum and its alloys in acidic solutions.

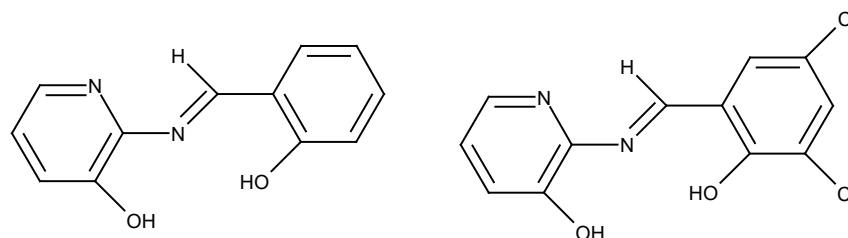


Figure 19. Representative Schiff bases as studied in [295].

No reasons for different inhibition efficiencies were provided, and the theoretical tools employed sometimes yielded suggestions like “adsorption proceeds by a chemical mechanism” again, without even indicating a cause–effect relationship. Biodegradation of Schiff bases has been studied [305].

In other research, 8-hydroxyquinoline further substituted in position 5 was tested as a corrosion inhibitor [306]. Since the plain 8-hydroxyquinoline was not included as a reference, any substituent effect could not be concluded beyond the observation that chlorine at the end of the substituent chain caused a slightly better performance than bromine.

Research compared 1,3,5-oxadiazinium salts with aliphatic and aromatic substituents as corrosion inhibitors for copper [307]. Aromatic substituents caused better inhibitor performance, attributed to their stronger adsorption.

Quaternary ammonium salts with a wide variety of aromatic moieties have been compared as corrosion inhibitors for pure iron and mild steel using impedance measurements [276]. Most inhibitors were more effective on pure iron than on the alloy. According to reports, 1-naphthylmethylquinolinium chloride performed best because it had the largest number of aromatic units in the inhibitor molecule and favorable dielectric properties in the inhibitor adsorbate layer. In a comparative study of the inhibition efficiency of *N*-propargyl derivatives of aromatic amines on steel 10, the amine with a *p*-toluidine group performed best [308]. This was attributed to an increased electron density on the nitrogen atom caused by the influence of the methyl group on the toluidine.

Some azine and thiazine dyes (for examples, see Figure 20) have been examined experimentally and theoretically as corrosion inhibitors [309]. Why the authors call inhibition efficiencies well below 90% “excellent” is only one of the mysteries in this report.

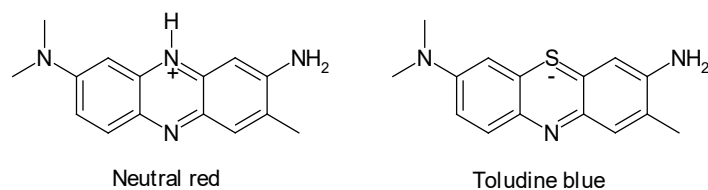


Figure 20. Some azine and thiazine dyes.

The QSAR-approach employed also only confirmed the experimental observation that sulfur-containing dyes performed slightly better. A theoretical investigation of several substituted triazines was reported [310]. The active sites for adsorptive interaction with the surface to be protected were the nitrogen atoms and the aromatic rings of the substituents. Hexahydro-1,3,5-*p*-aminophenyl-s-triazine was proposed as the most efficient, based in particular on the DFT-based relatively strongest adsorption; this has been verified experimentally elsewhere [311]. In another study, 1,4-diazines (pyrazines) were reviewed as potential corrosion inhibitors [312].

Studies of corrosion inhibition by SAMs built from aromatic molecules 4- and 2-mercaptopyridine (see Figure 21) on iron alloy C-60 have been reported [313]. The stronger adsorption and higher corrosion inhibition of 2-mercaptopyridine was attributed to formation of a chelate-like adsorbate structure.

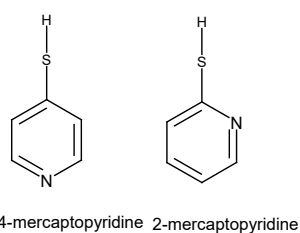


Figure 21. 4- and 2-mercaptopyridine.

1.3. Substituted Five-Membered Hetero-Atom Containing Rings

Corrosion of stainless steel 304 can be inhibited by thiophene derivatives (see Figure 22) [314]. Both cathodic and anodic reactions were slowed down but only to a rather moderate extent. [315].

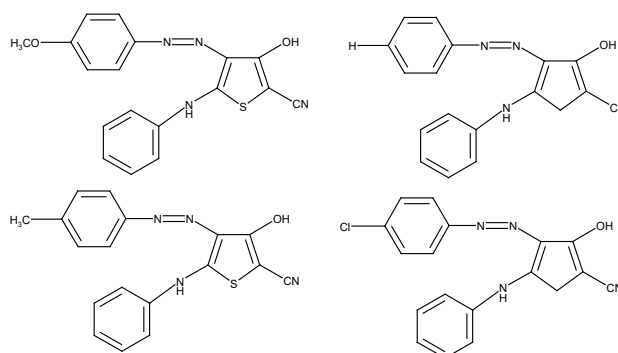


Figure 22. Some substituted thiophene corrosion inhibitors.

Highly fluorinated quaternary bisammonium surfactants with a furan building block showed high inhibition up to 98% at 2 mM inhibitor concentration [316]. Why these highly fluor-containing compounds are claimed to be green and sustainable remains mysterious.

In a study reporting the antibacterial effects of a new pyrrole derivative, its corrosion inhibition was examined [317]. At 600 ppm inhibitor concentration in sulfuric acid, 82% inhibition was found, with 84% in hydrochloric acid. The difference was not even

addressed; possibly, it was due to enhanced adsorption of the inhibitor on top of adsorbed chloride anions.

Azole-based corrosion inhibitors, i.e., compounds with two or three nitrogen atoms in a five-membered ring, were reviewed, and some were synthesized to support the conclusions finally drawn [318]. Triazoles are relative simply to prepare and were able to reach inhibition >90% at concentrations around 850 μM , imidazoles performed only slightly worse at 85%. The contributions of the aromatic electron systems and the nitrogen atoms to adsorptive and thus inhibitive interaction were stressed, and the same observation was reported for further substituted imidazolines with only moderate inhibition efficiency [319]. Further studied substituted triazoles showed inhibition of up to 99% at 500 ppm inhibitor concentration [320]. Synergistic effects of two inhibitors 2-amino-4-methylthiazole and 2-(methylthio)imidazole for the protection of galvanized steel were observed [321]; the effect was attributed to a favorable combination of the two different protection mechanisms of both inhibitors.

In a comparison of five azoles ranging from indole to 2,1,3-benzothiadiazole, all compounds except for the latter one, which acted as a stimulator (or accelerator), showed inhibition efficiencies around 90% at millimolar inhibitor concentration [322]. Three carbazole derivatives were compared as corrosion inhibitors, and 6-methyl-2,3,4,9-tetrahydro-1H-carbazole (Figure 23) performed best [323]. The accompanying theoretical calculations suggested participation of heteroatoms and aromatic ring systems in adsorptive interactions, providing corrosion protection.

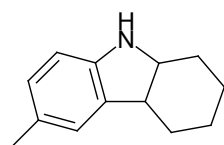


Figure 23. 6-methyl-2,3,4,9-tetrahydro-1H-carbazole.

In a comparison of two corrosion inhibitors for gas and gas condensate environments, an imidazole-based inhibitor was superior to an amine salt-based inhibitor at concentrations below 100 ppm [324]. This was attributed to a stabilizing effect of the aromatic imidazole structure. Substituted imidazoles are frequently encountered as cations in ionic liquids [325,326]. Some of them have also been examined as corrosion inhibitors for various metals with frequently very high inhibition efficiencies; for examples, see [327–333]. The influence of the chain length of aliphatic substituents at the imidazole moiety on corrosion inhibition has been studied theoretically [334]. Results suggest a major contribution of the aromatic electron system in the imidazolium ring on adsorption; the electron density is influenced by the length of the alkyl chain. At $n = 18$, an optimum was found, and this has been confirmed experimentally [335,336]. The beneficial effect of a benzyl substituent in 1-benzylimidazole compared with only alkyl substituents in 2-ethyl-3-methylimidazole was noticed [337]. Supporting results from theoretical considerations in terms of lower band gap energy and lower global hardness were stated, while the actual difference in terms of inhibition was small. It remains unclear whether the claimed enhanced adsorption was provided by the aromatic substituent, given that the free enthalpies of adsorption listed in a table entitled “2-I-Imz and 2-Cl-Imz” (?) were practical identical. An application-related detail of imidazole as a corrosion inhibitor was revealed with a combination of electrochemical impedance measurements and XPS [338,339]. Adsorption from an aqueous solution, e.g., the corrosive environment, yielded an adsorbate with the imidazole nitrogen behaving like a “pyrrole nitrogen” with the aromatic ring more parallel to the metal surface. Adsorption of imidazole before exposure to the corrosive environment left the nitrogen in a “pyridine type” state with the aromatic ring more perpendicular to the surface. The better protection in the former case was obvious in a polarization resistance (i.e., the charge transfer resistance of the corrosion electrode reaction) that was larger by an order of magnitude. Effects of further substituents on corrosion inhibition via imidazole 2,4,5-trisubstituted with aromatic phenyl units were compared [340]. At 1 mM inhibitor

concentration, inhibitions varied between 92 and 96%, suggesting only minor effects, although no attempt was made to interpret this. A further comparison of the effects of much simpler substituents for imidazole was reported, yielding a wide range of effects and with an aromatic substituent in 4-(1*H*-imidazol-1-yl)benzaldehyde showing 92% inhibition at an unspecified inhibitor concentration [341]. A water-soluble zeolitic imidazole framework, ZIF-8 has been suggested for copper corrosion inhibition [342]. At 60 mg·L⁻¹ inhibitor concentration, 75% inhibition was achieved, and the adsorption of the inhibitor on the copper was attributed to the aromatic electron systems in the imidazole units.

Based on results of a theoretical study of several benzimidazole derivatives, 2-(2-bromophenyl)-1-methyl-1*H*-benzimidazole) was suggested as the most promising inhibitor [343]. In a comparison of three substituted benzimidazoles, 2-aminobenzimidazole (see Figure 24) was identified as being most effective, in agreement with results of theoretical considerations [344]. The beneficial effects of both nitrogen heteroatom(s) and aromatic electron systems in substituted benzimidazole inhibitors have been highlighted [345]. In a mostly incomprehensible theoretical comparison of five substituted imidazolines, the one with an aromatic substituent was claimed to be most effective without even a trace of experimental evidence [346].

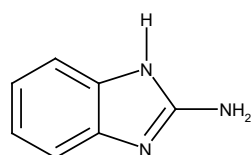


Figure 24. 2-Aminobenzimidazole.

In other work, 2-aminobenzimidazole (see Figure 24) was studied as a corrosion inhibitor for copper in chloride-containing solution via several electrochemical methods [347] and compared with further imidazole-based inhibitor molecules.

Moreover, 2-mercapto-5-methoxybenzimidazole has been studied as corrosion inhibitor for copper and has been compared with 5-amino-2-mercaptobenzimidazole [348]. Using microscopic methods, formation of self-assembled monolayers (see above) was observed, and these results were in agreement with further spectroscopic evidence.

The iron corrosion inhibitive properties of a wide selection of heterocyclic diazoles have been compared [349]. Inhibition efficiencies at millimolar inhibitor concentrations in aqueous 1 M HCl were around 80 to 90%, except for a nitro-substituted compound showing actually corrosion acceleration at increasing concentrations. Theoretical calculations did not yield conclusive results supporting the experimental observations. Three aromatic oxadiazoles were compared as corrosion inhibitors [350]. Inhibition efficiencies at 500 ppm inhibitor concentration reaching 98% for 2-cinnamyl-5-mercapto-1-oxa-3,4-diazole were larger in sulfuric than in hydrochloric acid as corrosive medium. Inhibition proceeded via adsorption via the aromatic electron system and the heteroatoms, and the lower efficiencies in HCl were tentatively attributed to “higher aggressiveness”.

Copper corrosion inhibition by tautomeric forms of 2-aminino-5-mercapto-1,3,4-thiadiazole added in very small concentrations to rolling oils yielded 92% inhibition at 50 ppm inhibitor concentration [351]. (Presumably, mercato and mercatpo mean mercapto; further mysteries remain in the report.)

Corrosion inhibition for aluminum by alkylimidazolium-based ionic liquids with alkyl-substituents of different lengths in the 4-position was examined [352]. The longer octyl chain corresponded to slightly higher inhibition efficiency than the hexyl chain, and the butyl-substituted performed weakest. The slight decrease of the capacity (part of the constant phase element used in impedance data evaluation) with growing chain length suggested a more or less perpendicular adsorbate orientation, perhaps in line with the slight growth of inhibition efficiency. Plain imidazole and methylimidazole were tested as corrosion inhibitors for aluminum [353]. The methyl substituent barely increased the moderate inhibition efficiency.

Pitting corrosion protection for aluminum in neutral salt solution afforded by indole and some of its derivatives (tryptophane and tryptamine) was examined [354]. The highest efficiency of tryptophane was attributed to enhanced adsorptive interactions by both nitrogens. Some oxindoles showed inhibition of copper corrosion [355]. The structurally related molecule isatin has attracted attention as a corrosion inhibitor; some of its derivatives have been examined. For an overview, see [356].

Pyrazole-4-sulfonate (see Figure 25) as a ligand forms layered solids on copper surfaces, inhibiting corrosion [357,358]. At pH = 2, no corrosion inhibition was observed; at pH = 3 and even more so at 4, the corrosion rate decreased depending on the cation. In comparison with simple pyrazole, the increased efficiency was attributed to interaction of the nitrogen atoms enhanced by the sulfonate group. The protective coating on the copper surface was identified as a salt of the ligand containing both copper ions and ions of the metal cations of the used ligand salt.

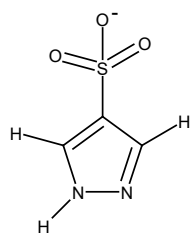


Figure 25. Pyrazole-4-sulfonate.

Aluminum corrosion inhibition by imidazole and some substituted imidazole in 1 M HCl acidic solution was attributed to film formation at higher (presumably 5 mM) inhibitor concentration [359]. Three thiazole-containing inhibitors for zinc were tested [360], and the best one provided 85% inhibition at 21 μ M concentration. The roles of the numerous heteroatoms and aromatic entities in the molecule were not addressed.

Corrosion inhibition on mild steel by 3,5-bis(*n*-methoxyphenyl)-4-amino-1,2,4-triazole (It should be noted that some triazoles are not aromatic.) (*n*-MAT) has been studied [361]. After invoking the general importance of aromatic rings and heteroatoms for adsorption as a step towards protection, the better performance of 4-MAT than of 3-MAT was attributed to a mesmeric effect. Protection of mild steel by 1(benzyl)1-H-4,5-dibenzoyl-1,2,3-triazole was studied, 95% at 50 ppm concentration of inhibitor was reported [362]. The number of electrons on the nitrogen atoms in the triazole ring was invoked as a reason for the good performance. In a comparative study of four substituted 1,3,4-thiadiazoles as corrosion inhibitors for silver, it was concluded that adsorptive interaction mostly involved the aromatic ring system and heteroatoms in polar substituent groups; thus, 5-methyl-2-amino-1,3,4-thiadiazole performed best [363]. Phenyl and diphenyl substitution were much less effective and were even detrimental because of steric hindrance and lack of contribution towards adsorption. Structure–efficiency relationships were investigated for three substituted triazoles and the parent compound 1(benzyl)1-H-4,5-dibenzoyl-1,2,3-triazole on mild steel [364]. The unsubstituted parent compound performed best, and further conclusions remained tentative. Further triazoles and isoxazoles have been studied as corrosion inhibitors for copper-nickel alloys and galvanized steel [365]. Inhibition efficiency was similar to that of already available commercial products. The low efficiency with copper–nickel alloys was attributed (not surprisingly) to weak adsorption on copper. In rather general terms, free electron pairs at the heteroatoms, chain length (presumably of some substituents), and the aromatic electron systems were invoked as possible reasons for corrosion protection on galvanized steel.

The biodegradability of triazole-based corrosion inhibitors for yellow metal has been examined [366]; for further details, see [367]. Transformation and the fate of organic compounds including corrosion inhibitors in groundwater have been discussed more generally [368]. Some aspects of microbial inhibitor degradation have previously been discussed with par-

ticular attention to degradation of aromatic constituents and moieties [369–371]. Corrosion inhibition on copper and copper alloys by aromatic triazoles has been studied [372]. Even concentrations of e.g., benzotriazole BTA as low as 2 ppm afforded corrosion protection for copper and brass fittings in household applications. The vapor pressures of some compounds are high enough to enable gas phase protection, and in addition, low toxicity and biodegradability are further advantages. With mild steel, BTA was also best compared with other nitrogenous aromatic inhibitors [373]. In the presence of chloride ions, the other inhibitors performed slightly better, while BTA did not show any change. This was explained by the assumption that the other inhibitors were adsorbed as cations better on the surface charged negatively by adsorbed chloride, whereas BTA was not adsorbed as a cation. For details of exemplary conceivable bioactivities of BTA, see [374]. The aqueous phototransformation of 1*H*-benzotriazole has been studied [375].

Inhibition of copper corrosion by BTA and several 5-substituted BTAs was compared [376]. Some of them worked simply via surface blocking by adsorption, and some participated in more complex surface reactions involving Co(I)-ions. Corrosion inhibition on 410 Martensitic stainless steel by 1,3-benzothiazole (Figure 26) was found to provide inhibition up to 98% at a very low concentration [377]. This was attributed to chemisorption resulting in electrostatic attraction and covalent bonding.

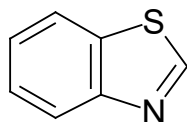


Figure 26. 1,3-benzothiazole.

Corrosion inhibition by three mono- and disubstituted furans for mild steel in aqueous HCl was studied experimentally and theoretically [378]. The 2-(*p*-toluidinylmethyl)-5-methyl furan performed best with 94% inhibition at 20 mM concentration. Apparently although not reported, the nitro and bromo substituents interfered with adsorption and inhibition via the less efficient molecules, corresponding to the slightly lower adsorption energies of these molecules. Benzylidene, actually benzylidene-5-phenyl-1,3,4-thiadiazol-2-amine (Figure 27), was prepared and studied as a corrosion inhibitor for mild steel [379]. In a report that is also elsewhere hard to comprehend, an inhibition efficiency of 92% is stated as a characteristic of this compound, apparently derived from weight loss measurements at 0.5 mM inhibitor concentration.

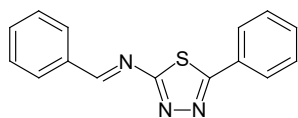


Figure 27. Benzylidene-5-phenyl-1,3,4-thiadiazol-2-amine.

A comparison of several five-membered heterocycles based on theoretical considerations was reported [380]. The claimed good correlation between some calculated data and experimental evidence reported elsewhere [381] apparently pertains only to data obtained with one particular DFT dataset. A repetition of an earlier study [380] with a CO₂-saturated NaCl solution yielded 2-aminobenzotriazole as the best inhibitor, with 93% inhibition at 20 mM inhibitor concentration [382].

DFT-based theoretical considerations regarding interactions between five-membered heterocycles and a Fe(110) surface suggesting flat adsorption have been reported [383].

1.4. Natural Compounds, Pharmaceuticals, Drugs, Dyestuffs, and Mixtures

The growing interest in biobased corrosion inhibitors has been highlighted [45,384–389]. Vanillin (Figure 28) was used as a remarkably efficient corrosion inhibitor (96% at 200 ppm inhibitor concentration) for aluminum in 5 M HCl solution [390].

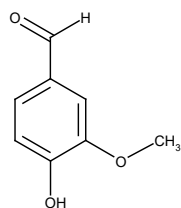


Figure 28. Vanillin.

The remarkable efficiency was attributed to strong, presumably flat, adsorption. This matches the reported observation that adsorption can be described best with a Langmuir adsorption isotherm, based on the absence of intermolecular interaction—indeed, most likely when flat adsorption prevails. Schiff bases prepared from vanillin and thus claimed to be green have been shown to exert high inhibition [391]. Corrosion inhibition by ten anthocyanins, major dyes in plants, was compared [392]. Delphinidin (Figure 29) was found to be most efficient, and this was explained by its having the lowest electron accepting power according to theoretical computations. A more simple explanation would be the relatively largest number of OH substituents supporting adsorptive interaction with the metal surface.

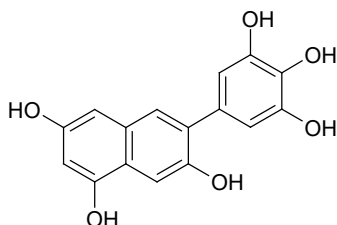


Figure 29. Delphinidin.

Tyrosine has been tested as an inhibitor with steel in hot sulfuric acid [393]. Results suggest formation of a protective layer beyond inhibitor adsorption. Adenin has been examined as a corrosion inhibitor for use in chemical mechanical polishing processes [394]. Inhibition by three amino acids reached 60% at millimolar concentration [395]; the amino acid derivate l-alpha-aminoisocaproate reached 85% inhibition at 5.63% inhibitor concentration [396]. Significantly higher inhibition up to 99% was achieved with phenylalanine and tryptophan after 240 h exposure in an environment simulating carbonated pore solution in concrete [397]. The former amino acid was assumed to be adsorbed via the carboxylate moiety with the benzene ring pointing towards the solution, resulting in a shielding effect keeping water away from the metal surface, whereas with the latter amino acid, the nitrogen of the indole system interacted with iron ions. In both cases, a passivating layer was formed, not the typical inhibition mode assumed elsewhere in this report. Corrosion inhibition by cysteine and some of its derivatives on mild steel has been studied experimentally and with computational methods [398]. The substituted compounds performed better, and the best was *N*-acetyl-S-benzyl-L-cysteine (Figure 30) with an aromatic benzyl unit. Theoretical considerations confirmed the participation of free electrons at the heteroatom and of π electrons of aromatic ring system, thus explaining the ranking of efficiencies. In a comparison of experimental and theoretical corrosion inhibition on mild steel by amino acids, the highest inhibition was predicted for those with an aromatic moiety (tryptophane > tyrosine > phenylalanine) [399]. This matched the experimental results, but despite the impressive long tables, it remains unclear how the predicted rating was actually derived from these numbers. The effects of side chains on α -amino acids on aluminum corrosion inhibition were studied in a comparison of glycine, alanine, and phenylalanine [400]. Somewhat surprisingly, the authors claimed in the conclusion that alanine was the most effective, apparently based on LPR measurements, whereas weight loss measurements suggested phenylalanine to be most effective. The actual differences

between the respective results of each method were bigger than between the amino acids, leaving the conclusion somewhat uncertain.

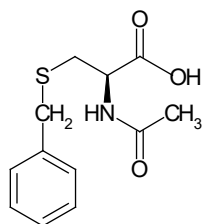


Figure 30. *N*-acetyl-*S*-benzyl-*L*-cysteine.

Additionally, 3-(toluyl)-*N*-(1, 1-dimethyl-2-hydroxyethyl)-alanine (Figure 31) has been suggested as a particularly effective temporary inhibitor [23,401].

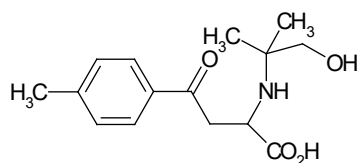


Figure 31. 3-(Toluy)-*N*-(1, 1-dimethyl-2-hydroxyethyl)-alanine.

The substance 6-benzylaminopurine (Figure 32) is a derivative of another naturally occurring compound, and its inhibiting properties reaching 96% have been reported [402].

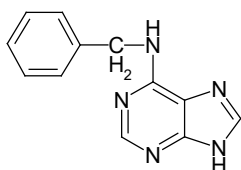


Figure 32. 6-Benzylaminopurine.

In a report on X60 carbon steel corrosion inhibition by dopamine-functionalized graphene oxide, the authors extensively discuss silver nanoparticle sensors [403]. The results of the corrosion measurements reported suggested 85% inhibition at 5 ppm inhibitor concentration.

Chitosan fragments of various sizes were treated computationally with DFT to “calculate the anticorrosion activity” [404]. Calculations suggested an optimum with the pentamer, while shorter and longer chains were theoretically less efficient. Unfortunately, the agreement with published experimental results claimed in the abstract was not supported in the remaining report. Corrosion inhibition by chitosan and further carbohydrates has been reviewed [405]. Chitosan and chemically modified derivatives have been studied theoretically and experimentally as corrosion inhibitors [406]. Inhibition of up to 93% at inhibitor concentrations around 250 ppm was found for the thio derivative.

Previously mentioned theoretical tools [404] were applied in a similar approach by the same author(s) to typical constituents of chamomile extract [407]. The strong inhibition claimed in the introduction was nowhere supported in the report. Whether quercetin is the most effective constituent in the extract as claimed in the report remains a question for further research. Actually, high inhibition by quercetin on mild steel has been reported elsewhere (97% at 1000 ppm inhibitor concentration) [408]. More theoretical considerations on quercetin and six derivates can be found [409]; somewhat surprisingly, amino nitrogens present only in two derivatives can increase the electron donating power and thus enhance adsorption. The mostly planar structure of the studied derivatives helps in stronger adsorption, according to theoretical considerations [410]. Similar considerations have been applied to carbohydrates as corrosion inhibitors for copper [411]. The claimed best inhibition by

lactose waits for experimental verification. The high corrosion inhibition by red onion peel extract (see below) was most likely due to its main ingredient: quercetin [412].

Fennel seed extracts demonstrated corrosion inhibition activity (IE = 85.8% at 150 ppm concentration) on stainless steel 304 [413]. Activity was attributed to the fenchone and aromatic anethol (1-methoxy-4-(1-propenyl)benzene) in the main constituent. A cinnamon extract was protective (80% at 250 ppm) to steel in sulfide-containing salt water [414]. An extract from *Rosmarinus officinalis* with added 5-bromovanillin provided corrosion protection for 1018 carbon steel in hydrochloric acid, and much less in sulfuric acid [415]. In a highly similar report, the author reported highly similar observations with added ZnO [416]. The author report highly similar results elsewhere [417], nothing was said about solubility or mixing issues in particular when inhibitor concentrations up to 2.5% were reached. The author used said the same extract again in a study of pitting corrosion on ferritic stainless steel [418]. Further extracts from natural raw materials have been reported [22,412,419–465]. The frequently employed theoretical tools—actually, the conclusions are frequently fairly trivial when stating that adsorption is probably operating—appear to be of limited value when studying complex mixtures of numerous compounds contained in plant extracts, as in e.g., [466], because they are either generic or non-specific or both. Additional aromatic plant extracts have been compared as corrosion inhibitors [467]. The residue of cashew nutshell liquid distillation combined with propargylic alcohol, propyl alcohol, and butylglycol provided 99% inhibition on P110 carbon steel in acidified oil well fluids [468]. Polycondensed aromatic ring systems in the residue were suggested as a main reason for efficient adsorption and high inhibition.

Although no inhibition efficiency was reported, the tannin-containing extract from *Rhizophora mucronata* was claimed to offer efficient corrosion protection [465]. Treated extract from the plant showed elsewhere only moderate protection, as reported by the same authors [469]. Corrosion protection by tannin extracted from mimosa, which is actually a mix of at least four flavonoids, has been studied theoretically [470]. Given the extended size of the molecule, the claimed perpendicular orientation is slightly surprising. Weight loss measurements described in the experimental section did not yield inhibition data, but only adsorption data. Actual inhibition by this extract remains unknown.

An aqueous plant extract from Doum (*Hyphaenethebaica* L.) containing a rich variety of organic compounds including several aromatic ones tested as a corrosion inhibitor for carbon steel showed significant inhibition efficiencies already at around 25 ppm concentration, but numbers differed substantially depending on the employed experimental method [471]. Inhibition of corrosion of ASTM/A36 steel by an extract from zoffa plants has been studied extensively [387]. At 1500 ppm concentration, 94% inhibition was found. Given the complex composition of the extract, it appears mysterious how a zoffa molecule can be responsible for the inhibition; most probably, aromatic constituents played a significant role. Inhibition of α -brass corrosion by an extract from *Chelidonium majus* plants reaching 97% inhibition efficiency at 150 ppm concentration was reported [472]. The main constituents of the extract had aromatic building blocks and many heteroatoms in five- and six-membered rings supporting adsorption on the metal surface. How fractions of transferred electrons in the gas phase can support stronger adsorption remains a question not yet answered. An extract from leaves of *Cymbopogon citratus* inhibited corrosion [473]. At a concentration of 5 g·L⁻¹, a pomelo peel extract provided 93% inhibition [474]. Presumably, aromatic units in the numerous constituents in the extract supported adsorption in addition to the heteroatom-containing functional groups invoked by the authors. In any case, a “geometric coverage” effect was claimed as the operating inhibition mechanism. An extract from sunflower seed hulls provided 98% inhibition at 400 ppm inhibitor concentration [475]. Given the wide variation of compounds identified in the extract, it is most likely that the numerous heteroatoms and aromatic compounds contributed to adsorption and inhibition. Lignin polymers extracted from *Elaeis guineensis* agricultural waste yielded a more efficient and better soluble corrosion inhibitor after chemical modification with insertion of aromatic moieties [476]. Possibly, the added aromatic units supported adsorption. Plant extracts

have also been suggested as protective corrosion inhibitors in coatings; for example, henna from *Lawsonia inermis* (see [477]). Arguments for their protective capabilities (presence of double bonds and aromatic systems) are the same as already discussed for inhibitors in the preceding text. The plant extract glucomannan with hydroxyl and epoxide groups supporting adsorption provided only 85% inhibition; after introduction of thiazole and Schiff base moieties, 99% inhibition at 500 μM inhibitor concentration was obtained [478]. The improved performance was attributed to the additional heteroatoms and aromatic as well as double-bond systems.

A limited overview of corrosion inhibition by plant extracts is available [479], and more extensive ones have been provided [480,481]. Options for simplified extraction procedures for bio-based inhibitors have been reported [388].

Several extracts from biodiesel fuel have been tested for corrosion inhibition and have been touted as green inhibitors [482]. Somewhat surprisingly, no inhibition data have been reported.

The dye new fuchsine provided 88% corrosion inhibition for stainless steel at 1 mM inhibitor concentration (Figure 33) [483]. Further dyestuffs considered as corrosion inhibitors have been reviewed [484]. A chitosan coating was impregnated with cresol red for corrosion protection of zinc [485]. When the dye was used alone as an inhibitor, its efficiency was higher because of the blocking of some functional groups during incorporation into the chitosan coating.

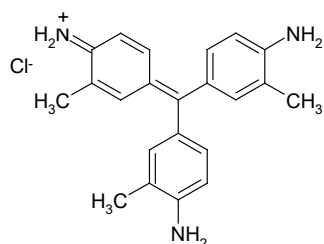


Figure 33. New fuchsine.

The use of common drugs with aromatic building blocks in their chemical structure as corrosion inhibitor has been suggested frequently, (see e.g., [486–496]) sometimes supported by QSAR considerations [497]. Missing experimental details of experimental handling of insoluble materials do not help [498]. Theoretical considerations regarding such materials are available [487,499,500]. Whether growing concerns regarding unnecessary exposure of humans to e.g., ampicillin or penicillin qualify such inhibitors as environmentally friendly seems to need closer inspection. To even use expired drugs as suggested [501,502] may be considered as a particularly strange method of waste disposal. Similar concerns may be applicable for e.g., azo dyes showing moderate efficiencies anyway [503,504]. Some imidazoline and isoxazoline compounds claimed to be “bioinspired” for reasons not obvious in the report have been synthesized and tested as corrosion inhibitors [505]. The former performed better than the latter because “imidazoline contains less electron-withdrawing nitrogen atoms in the(ir) molecular structure”, which provide superior metal–inhibitor interaction. This claim fundamentally contradicts the statement made elsewhere in this report that adsorption is fundamental for inhibition of corrosion. The arguments suggested elsewhere [326] for the designation of ionic liquids and substituted imidazoles therein as being “green” are strange at best.

1.5. The State of Things

This brief overview, which most probably misses many aromatic inhibitor molecules simply because the authors did not mention this essential property of an organic molecule in their reports, does not yield a simple recipe for a perfect inhibitor, not even for a good one. Starting points, requirements, conditions of application, and further variables differ too wildly for a general conclusion. Trial and error as well as cases of “also tested” molecules are frequently observed although not clearly stated by the authors. Only infrequently do

the authors provide some reasoning about the selection of a particular molecule or the synthesis of a new one. Frequently mentioned parameters, in particular the Gibbs energy (or free enthalpy) of adsorption for an inhibitor, provide only some support when looking for a reason for more efficient inhibition by a particular molecule in a direct comparison. The numbers are rarely available; their determination may be more tedious than running the standard corrosion studies. Thus, the obvious conclusion that a more strongly adsorbed molecule is presumably also a better inhibitor is a rather helpless one. The use of non-traditional, in particular spectroscopic and surface-analytical, tools has turned out to be less helpful than possibly expected. Microscopic methods provide only post-mortem and ex situ observations, making any correlation in terms of a “cause-and-effect” connection difficult or at least very general. Applications of in situ spectroscopies possibly providing evidence of inhibitor–surface interaction on a molecular level and thus providing support for further rational inhibitor development based on a molecular-level understanding are rarely found.

In the apparent absence of a “rational design approach” for planning new inhibitors, even the much-touted use of theoretical tools has turned out to be less than spectacularly successful; for examples, see [506–508]. Lists of calculated data having impressive length frequently do not contribute to a better understanding of the effects of a given inhibitor, even when in comparative studies, such a correlation is to be expected. As pointed out in a report on aromatic inhibitors [77], correlations between values of the HOMO-LUMO-gap (band gaps [509]) and inhibition efficiency may indicate high efficiency at a particular band gap value (illustrations in the report hardly support this), but scarcely more. At least the authors state that aromatic inhibitors are efficient inhibitors. Using a limited subgroup (substituted pyridines) with rather limited inhibition efficiencies, correlations between electronic parameters obtained by extended Hückel molecular orbital calculations and observed inhibition were reported [510]. In most cases, almost all output data were collected from DFT packages, but a correlation with experimental data—if available at all—is claimed but hard to find. Certainly no “cause-and-effect” connection is indicated, but the studies are a help when selecting or designing further inhibitor molecules. Theoretical data for just one molecule could not be demonstrated as particularly helpful in the studies reviewed above. Within a group of structurally related molecules, trends may be deduced from such calculations that would be helpful in e.g., selecting alternate substituents. Whether calculations based on examination of complexes between an inhibitor molecule and just one metal atom are helpful remains at least unclear, given the less than convincing correlations claimed [511]. The convincingly simple conclusions and suggestions related azoles and imidazoles [318] at least recommend an approach towards simple but effective inhibitors quite different from the numerous different examples of inhibitors that are extremely complex and sometimes even difficult to prepare but no more efficient at all.

Examination of inhibitors from plant extracts has repeatedly yielded quite impressive inhibition values. In some cases, an analysis of the extract and a search for both the major ingredients and the most efficient ones (for an impressive example, see [420]) have been carried out and reported, and their use may be a promising “green” approach for combating corrosion. The actual inhibitor concentrations needed even for modest inhibition range widely. Sometimes, when e.g., more than $1 \text{ g}\cdot\text{L}^{-1}$ is needed, the practical applicability appears to be at least doubtful.

2. Concluding Remarks and Perspectives

An overview of aromatic corrosion inhibitors ranging from small but nevertheless highly protective molecules to complex mixtures obtained as plant extracts from natural and thus renewable resources is provided. They act by adsorption on the surface to be protected. Depending on metal surface properties and molecular architecture, formation of self-assembled monolayers occurs; sometimes, presumably as an extension of molecular adsorption, film formation is claimed. Attention is paid to the actually observed inhibition efficiencies (which are frequently claimed, but not always reported). These are correlated

with structural features of the inhibitor molecule, which always contain heteroatoms in addition to one or more aromatic ring system. No simple reasoning, i.e., a straightforward rational approach towards the perfect inhibitor, could be derived. However, some guidelines could be identified. The sometimes apparent trend away from synthetic inhibitors with their implicit drawbacks (price, availability, toxicity, stability) towards natural inhibitors mostly obtained as plant extracts or as compounds otherwise occurring in nature (e.g., amino acids) is superimposed on the more chemical considerations. Selection of a “natural” or synthetic inhibitor should nevertheless always be based on the particular application situation.

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