Chapter 4

Ionic Liquids as Corrosion Inhibitors

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Due to economic importance of corrosion prevention of materials, the design of new and effective corrosion inhibitors is one of the important topics of the science. Especially, the development of green and sustainable methods in corrosion science is among the topics of interest of material scientists. It is widely reported that ionic liquids are effective corrosion inhibitors due to their low volatility, non-inflammability, non-toxic nature and high adsorption ability on metal surfaces. Many ionic liquids are used in the prevention of the corrosion metal surfaces. In this chapter, applications and remarkable properties as corrosion inhibitor of ionic liquids are presented in detailed. Additionally, corrosion inhibition applications of some electronic structure principles and electronic structure analyses of the ionic liquids are highlighted. Some parts about the chemical reactivity of ionic liquids are presented. The usefulness of some new parameters proposed as an indicator about the corrosion inhibition performances of molecules will be mentioned in this chapter and will be proven in the next paper projects.

An Introduction about the Corrosion

For the prevention of the corrosion known as spontaneous deterioration process of any metal and alloy as a result of chemical and electrochemical reactions with the environment, countries use large amounts of budgets each year. Although several non-metallic substances may undergo deterioration, the corrosion term covers the situations involving metals. Corrosion of metals cause serious economic losses around the world. For that reason, the studies including the prevention of the corrosion of the metals become the center of attention of the academy and industry (*1*).

Many methodologies have been developed for the prevention of the corrosion of metals. One of the most effective methods among them is the synthesis of new and effective materials having

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cost effectiveness and ease of application in industry. Corrosion protection methods reported in the literature are given as detailed in the Figure1 by taken from the review paper penned by Quraishi and coworkers (*2*).

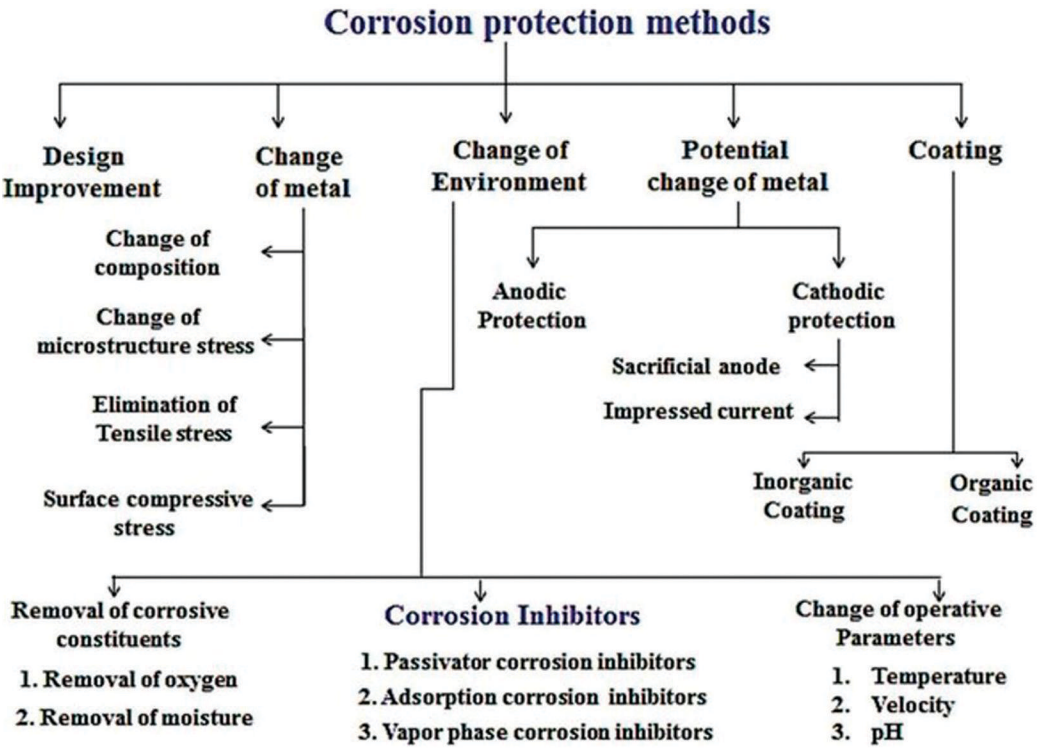

Figure 1. Reported methods in the literature for corrosion protection of metals.

The main task of an effective corrosion inhibitor is to prevent or to minimize the corrosion of the metals in a corrosive medium. There is a powerful relation between electronic/molecular structure of inhibitor molecule and its adsorption ability and characteristics. Many published papers regarding to great adsorption capacities of organic compounds are seen in the current literature (*3*, *4*). Especially, the molecules having some heteroatoms such as nitrogen, oxygen and sulfur in their molecular structure are considered effective corrosion inhibitor. In addition, aromatic rings and the existence of the multiple bonds in the structure also are among the parameters effecting the corrosion inhibition ability of molecules. In applications regarding to the new synthesized molecules, their corrosion inhibition efficiencies as well as toxicities also should be taken into consideration. For that reason, some groups or synthetic organic corrosion inhibitors and some traditional inorganic corrosion inhibitors like chromates and lead have limited applications in the corrosion science because of their hazardous effects. With the using of different substituents in the structure, corrosion inhibition performances of molecules can be easily changed. Organic compounds known as filming inhibitors also prevent the corrosion of metal surfaces by forming a protective film in corrosive medium in general. A vast majority of organic corrosion inhibitors includes polar functional groups like $–NO_2$, -OH, $-OCH_3$, CH_3, $-NH_2$, $-NO_2$, -COOH, $-CONH_2$ (*5*). With the help of these functional groups and multiple bond in the structure, inhibitor molecules interact with metal surfaces and adsorb on them. The adsorption capacities of inhibitor molecules are affected from some factors also such

as electronic structure properties of the inhibitor, temperature, time, the nature of the corrosive medium (*6*).

Ionic liquids have many applications in the many fields of the science. It is important to say that the number of the publications about the applications of ionic liquids is increasing day by day. It can be understood from here that this remarkable topic attracted the attention of many researchers working on material science, chemical engineering, synthesis of new structures, physical chemistry, corrosion science and environmental chemistry. Insights about the nature of ionic liquids are reported in the literature as detailed. In the past studies, it was widely reported that ionic liquids exhibit as "nonvolatile, non-flammable, and air and water stable". On the other hand, nowadays, we often come across to volatile, reactive and flammable ionic liquid types. Corrosion inhibition studies explaining that ionic liquids have high corrosion inhibition performance support the idea of which ionic liquids are generally reactive. It is well-known that reactive molecules can easily give the electrons to metal surfaces and act as effective corrosion inhibitor. The reactivity or stability of ionic liquids are closely related to the chemical behavior of anions and cations forming the ionic liquids. These compounds with increasing usage areas can be grouped into many types as room-temperature ionic liquids, task-specific ionic liquids, poly-ionic liquids and supported ionic liquids membranes. Because ionic liquids have both organic and ionic nature, the studies about the highlighting of their intermolecular interactions are both impressive and challenging. In the following parts, we present some details about the theoretical prediction of corrosion inhibition process in the light of important reactivity indices and electronic structure principles. Additionally, we will present some suggestions to corrosion scientists to use in the theoretical parts of their future studies about corrosion science.

The Link between Electronic Structure and Inhibition Efficiency in Corrosion Science

As parallel to the developments in the Conceptual Density Functional Theory (CDFT) (*7*), many useful parameters reflecting the stability, reactivity, electron donating ability and electron accepting ability of chemicals systems started to be used to explain the corrosion inhibition performances of molecules as theoretical in the corrosion science. For that reason, firstly let us mention from the mathematical definitions of Conceptual Density Functional Parameters. In the conceptual Density Functional Theory chemical potential (μ), electronegativity (χ), hardness (η) and softness (σ) are defined via the following equations in terms of the total electronic energy (E), the number of electrons (N), ground ionization energy (I) and ground state electron affinity (A) (*8*).

$$\mu = -\chi = \left(\frac{\partial E}{\partial N}\right)_{v(r)} = -\left(\frac{I+A}{2}\right) \tag{1}$$

$$\eta = \frac{1}{2}\left(\frac{\partial^2 E}{\partial N^2}\right)_{v(r)} = \left(\frac{\partial \mu}{\partial N}\right)_{v(r)} = \frac{I-A}{2} \tag{2}$$

$$\sigma = 1/\eta \tag{3}$$

In the derivation of a widely used electrophilicity index (we called as first electrophilicity index in this chapter) given via the equation $\omega_1 = \chi^2/2\eta = \mu^2/2\eta$, Parr, Szentpaly and Liu (*9*) assumed an electron transfer process observing until the chemical potentials of an electrophile and an idealized free electron sea with zero chemical potential become equal to each other. In addition to first

electrophilicity index, second electrophilicity index (ω_2) also, which its usefulness is emphasized by especially Szentpaly and Kaya (*10*) can be used in the corrosion inhibition studies. Second electrophilicity index is defined as:

$$\omega_2 = I.A/(I-A) \tag{4}$$

Gazquez and coworkers (*11*) introduced two new parameters, namely electrodonating power (ω^-) and electroaccepting power (ω^+), to predict the electron donating and electron accepting abilities of molecules based on ground state ionization energy and electron affinity values. These quantities are mathematically described via the following equations and lately they are widely taken into consideration in the prediction of corrosion inhibition performances of molecules.

$$\omega^+ = (I+3A)^2/(16(I-A)) \tag{5}$$

$$\omega^- = (3I+A)^2/(16(I-A)) \tag{6}$$

Simple electron transfer processes can be explained in the light of Taylor series expansion of electronic energy as a function of the number of electrons and external potential considering the equation $\Delta E = \mu\Delta N + (1/2)\eta(\Delta N)^2$. For possible donation and back-donation processes differentiating through the use of different values of the chemical potential and fixing the chemical hardness to the value of $\eta = (\mu^+ - \mu^-)$, in the case of which the molecule receives a certain amount of charge (ΔN^+) and in the case of which the molecule back donates a certain amount of charge (ΔN^-), the following formulas can be written, respectively.

$$\Delta E^+ = \mu^+\Delta N^+ + (1/2)\eta(\Delta N^+)^2 \tag{7}$$

$$\Delta E^- = \mu^-\Delta N^- + (1/2)\eta(\Delta N^-)^2 \tag{8}$$

Total energy change can be approximately calculated taking the sum of the processes mathematically defined via Eq.17 and Eq.18. On the other hand, one can write the relation $\Delta N^- = -\Delta N^+$ because received and back donated charges are equal to each other. If so, the following equation can be presented.

$$\Delta E_T = \Delta E^+ + \Delta E^- = (\mu^+ - \mu^-)\Delta N^+ + \eta(\Delta N^+)^2 \tag{9}$$

The most favorable situation is that the energy change becomes a minimum with respect to ΔN^+ and here ΔN^+ can be given as $\Delta N^+ = -(\mu^+ - \mu^-)/2\eta$. Finally, to calculate the back-donation energy, the following equation has been obtained by Gomez and coworkers (*12*) considered the approaches and equations 7-9.

$$\Delta E_{back-donation} = -\eta/4 \tag{10}$$

Although original form of the Koopmans Theorem provides a rough calculation technique for the prediction of ionization energy and electron affinities of molecules from frontier orbital energies, $I = -E_{HOMO}$ *and* $A = -E_{LUMO}$, one of the most reliable methodologies used for this aim is to use

of the finite differences approach based on the energies of the systems with different number of electrons, $I = E(N-1) - E(N)$ and $A = E(N) - E(N+1)$. Additionally, to obtain more reasonable and reliable results in corrosion inhibition studies, the extended forms of Koopmans Theorem can be preferred.

Local reactivity analyses of molecules are widely done with the help of Fukui indices. The Fukui function at a point, r, in the space around the molecule are mathematically defined as the first derivative with respect to number of electrons of electron density at a constant external potential.

$$f(r) = \left(\frac{\partial \rho(r)}{\partial N}\right)_{\nu} = \left(\frac{\partial \mu}{\partial \nu(r)}\right)_{N} \tag{11}$$

Fukui indices for nucleophilic (+) and electrophilic (-) attacks are defined via the following equations.

$$f^{+}(r) = \left(\frac{\partial \rho(r)}{\partial N}\right)^{+}_{\nu} \tag{12}$$

$$f^{-}(r) = \left(\frac{\partial \rho(r)}{\partial N}\right)^{-}_{\nu} \tag{13}$$

In the light of finite differences approach, Fukui functions can be calculated with the help of the following equations. In the given equations, qk is the charge on atom k (*13*).

$$f_k^{+} = q_k(N+1) - q_k(N) \quad \text{(for nucleophilic attack)} \tag{14}$$

$$f_k^{-} = q_k(N) - q_k(N-1) \quad \text{(for electrophilic attack)} \tag{15}$$

$$f_k^{0} = \left[q_k(N+1) - q_k(N-1)\right]/2 \quad \text{(for radical attack)} \tag{16}$$

Another parameter related to the chemical reactivity and never got the appreciation it deserved until today except for a few attempts of Cardenas (*14*), Ayers (*15*), Chattaraj (*16*), Parr and Roy (*17*) is Fukui potential, ν_f(r). Although some researchers noted that the hardness can be reported as the Fukui potential value at covalent radius, Fukui potential is generally reported as the electrostatic potential due to a distribution of charge equal to the Fukui function, *f*(r) and Fukui potentials are given as:

$$\upsilon_f^{+}(r) = \int \frac{f^{+}(r')}{|r - r'|} dr' \tag{17}$$

$$\upsilon_f^{-}(r) = \int \frac{f^{-}(r')}{|r - r'|} dr' \tag{18}$$

Cardenas and coworkers highlighted the powerful correlations between Fukui potential, the capacity of charge and global hardness and noted that Fukui potential can be successfully used to describe the chemical hardness of the atoms (*14*). Local reactivity analysis also is important such as global reactivity analysis in corrosion inhibition studies. For the prediction of local hardness, the following equation is used (*18*).

$$\eta(\mathrm{r}) = \int \eta(\mathrm{r},\mathrm{r}')\omega(\mathrm{r}')dr' \tag{19}$$

As different electrophilicity index, here, ω(r) stands for any continuous charge density. η (r, r′) is the hardness kernel and given via the following equation based on Hohenberg-Kohn-Sham universal functional of DFT, F[ρ]

$$\eta(r,r') = \frac{\delta^2 F[\rho]}{\delta\rho(r)\delta\rho(r')} \tag{20}$$

The correlation between Fukui potential and local hardness is given as detailed in some important published papers by theoretical chemists and physicists.

Important statements about the electronic structure principles and guiding rules can be effectively considered in the corrosion inhibition studies to predict the nature of the interactions between corrosion inhibitors and metal surfaces. Actually, the analysis of the corrosion inhibition efficiencies of molecules requires to do chemical reactivity prediction about them. For that reason, parameters based on solid foundations and the electronic structure principles and rules introduced for them are quite essential in the corrosion science.

Hard and Soft Acid-Base Principle

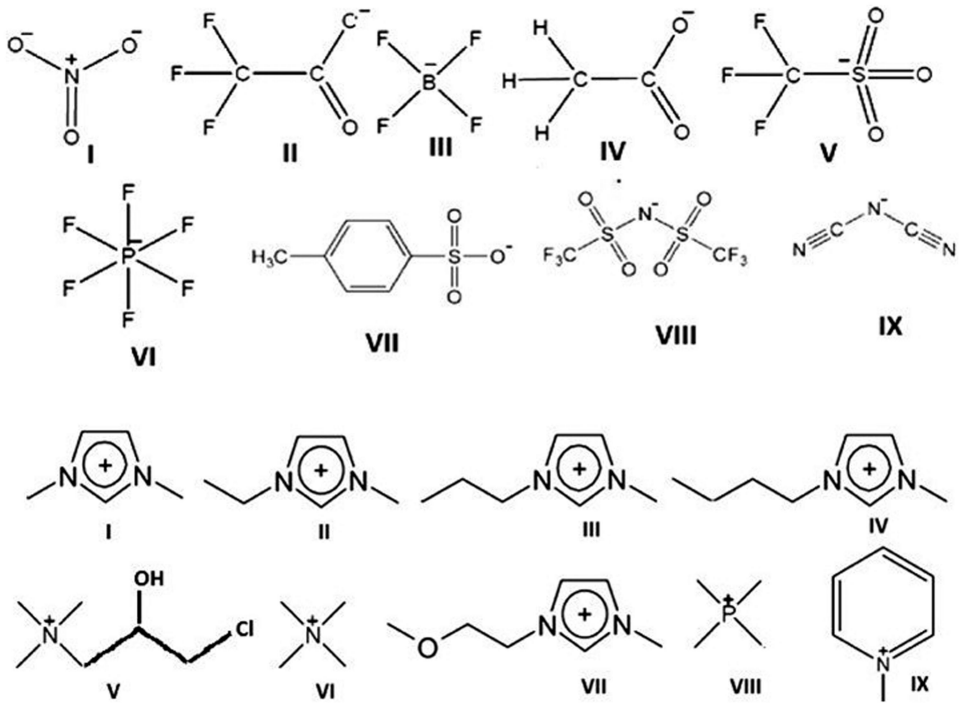

Figure 2. Widely used anions and cations in the designing of ionic liquids. Reproduced with permission from Reference (20). Copyright 2016, Elsevier.

"Hard acids prefer to coordinate to hard bases and soft acids prefer to coordinate to soft bases." Hard concept defines the chemical systems with low polarizability and high HOMO-LUMO energy gap while soft concepts defines the chemicals systems with high electron cloud polarization. Soft molecules act as effective corrosion inhibitors. According to R.G. Pearson (*19*), the products of

Lewis acid-base reactions can be predicted with the help of chemical hardness concept. The interactions between acids and bases from same category are quite powerful. It should be noted that hard-hard interactions are mainly ionic while soft-soft interactions are mainly covalent. It is well-known that both interactions are powerful chemical interactions. Some researchers presented theoretical investigations about structural properties of ionic liquids considering Hard and Soft Acid-Base Principle also.

In Figure 2, some common anions and cations using in the designing of new ionic liquids are presented. In the paper where Figure2 was taken, Benguerba and coworkers (*20*) investigated all possible ionic liquids, which can be designed via the anions and cations as theoretical. According to the calculations made by the mentioned author, Hard and Soft Acid-Base Principle should widely have considered in the designing of new ionic liquids.

Maximum Hardness Principle

"There seems to be a rule of nature that molecules arrange themselves so as to be as hard as possible." From this information, it can be emphasized that chemical hardness is used to explain the stability or reactivity of molecules and hard molecules are more stable compared to soft ones (*21*). It is required to note that an effective corrosion inhibitor should be reactive to give easily electrons to metal surfaces. The relation with hardness of chemical stability is illuminated in relation to many issues. For example, there is a remarkable correlation between stability and aromaticity. It is well-known that aromaticity provides extra stability to a chemical system. Zhou and Parr introduced the absolute hardness as a measure of the aromaticity. This is an expected situation within the framework of Maximum Hardness Principle. According to the information, it is not difficult to predict that aromatic molecules will not be effective against the corrosion of metal surfaces.

Minimum Polarizability Principle

"The natural evolution of any system is toward a state of minimum polarizability." This principle (*22*) has been proposed by Chattaraj and Sengupta in the light of Maximum Hardness Principle of Pearson. There is a remarkable correlation between softness and polarizability. For chemical systems, softness is proportional to the cube root of the polarizability as noted by Ghanty and Ghosh (*23*).

Minimum Electrophilicity Principle

According to Minimum Electrophilicity Principle (MEP) (*24*), Minimum Electrophilicity Principle, which can be considered as an extension of Maximum Hardness Principle, "in a chemical reaction, the sum of the electrophilicity indexes of the reaction products should be smaller than that of the reactants." Additionally, it can be written that "molecules tend to decrease their electrophilicity power during a chemical process, becoming this way less reactive." In corrosion inhibition studies, it is known and noted that the molecules having higher values of the electrophilicity index exhibits good electron accepting capability. Actually, in the light of MEP, it can be said that effective corrosion inhibitors should have high electrophilicity index values. This situation implies that MEP is not useful to explain the corrosion inhibition efficiencies of molecules. Already, Szentpaly and Kaya (*10*) explained as detailed the limitations and required conditions regarding to the validity of this principle in a recent paper.

Maximum Composite Hardness Rule (MCHR)

Maximum Composite Hardness Rule introduced by Szentpaly, Kaya and Karakuş (*10*) supports the using of Kaya's composite descriptor (*25*), $\eta_M/V_m^{1/3}$ (η_M: molecular hardness, V_m: molar volume) instead of single descriptors to explain the exothermic and endothermic nature of reactions and the stability/reactivity of the chemical systems. It can be said, the chemical systems with high composite descriptor value are more stable compared to others and cannot act effective corrosion inhibitors. Some analyses made by the authors of Kaya and Szentpaly showed that the composite descriptor is more useful compared to single descriptors like hardness and electrophilicity in the prediction of exothermic and endothermic nature of chemical reactions.

Minimum Magnetizability Principle

The relation with chemical reactivity of the magnetizability is explained with the help of Minimum Magnetizability Principle (*26*) proposed by Chattaraj and Pal. This principle states that "A stable configuration/conformation of a molecule or a favorable chemical process is associated with a minimum value of the magnetizability." In the light of this information, it is not difficult to predict that the molecules with high magnetizability can exhibit remarkable corrosion inhibition performances.

Maximum Entropy Principle

Entropy is one of the useful parameters related to the chemical reactivity of the chemical systems. Chattaraj and Sengupta proved that chemical systems exhibit high tendency to reach to maximum hardness, maximum entropy and minimum polarizability. Maximum Entropy Principle (*27*) states that "the most probable distribution is associated with the maximum value of the Shannon entropy." Some researchers noted that the alloys with high entropy exhibit excellent corrosion resistance as predicted via Maximum Entropy Principle.

In a recent paper, Kaya and Chattaraj (*28*) caught an important link between solid state chemistry and Conceptual Density Functional Theory investigating the relation with magnetic susceptibility of standard absolute entropy in inorganic solids. The authors saying that Maximum Entropy Principle implies the Minimum Magnetizability Principle presented the following equation for the calculation of the standard absolute entropy (S^0_{298}) based on magnetic susceptibility (χ_m).

$$S^0_{298} = \kappa(-\chi_m)^{1/3} - \tau \tag{21}$$

In this equation, κ and τ are constants depending on the stoichiometry of the inorganic ionic crystals. This important correlation between the entropy and magnetic susceptibility is a proof of which magnetic susceptibility of molecules can provide important insights about their corrosion inhibition efficiencies. In our next studies about corrosion inhibition of some new molecules, we will highlight and emphasize the usefulness as corrosion inhibition indicator of the magnetic susceptibilities.

Ionic Liquids as Corrosion Inhibitor and with Other Applications in the Science

Green chemistry (*29*) known as sustainable chemistry is an important branch investigating the design and implementations of non-toxic and eco-friendly matters and process of the chemistry and

chemical engineering. Ionic liquids can be defined as the compounds having forming completely from ions with melting point below 100 °C. The ethylammonium nitrate is the first ionic liquid discovered by Paul Walden in 1914. From that day until now, many ionic liquids having excellent properties and applications have been reported. As the compounds, which their properties were investigated in the scope of the green chemistry, ionic liquids have many applications in the important fields of the science. Many important published papers about the potentials of the ionic liquids are available in the literature. This compounds are widely preferred as green solvents in chemical synthesis studies. Ionic liquids exhibit great properties like non-toxic nature, low flammability, ease of containment and biodegradability (*30*). It is well-known that corrosion scientists prefer to use both effective and non-toxic inhibitor molecules in corrosion studies. For that reason, drugs and plant extracts also are widely considered as corrosion inhibitors because of their non-toxic nature. The process regarding to the extraction and purification of plant extracts is very tedious, laborious, expensive and time consuming. Moreover, in the mentioned process, large amounts of organic solvents, which are harmful for the environment and living organisms are used. This chapter focuses on adsorption characteristic and corrosion inhibition performances of ionic liquids. To avoid time consuming details for the applications of ionic liquids in other fields, we took and used from the review paper of Quraishi and coworkers the following figure summarizing the all applications of ionic liquids.

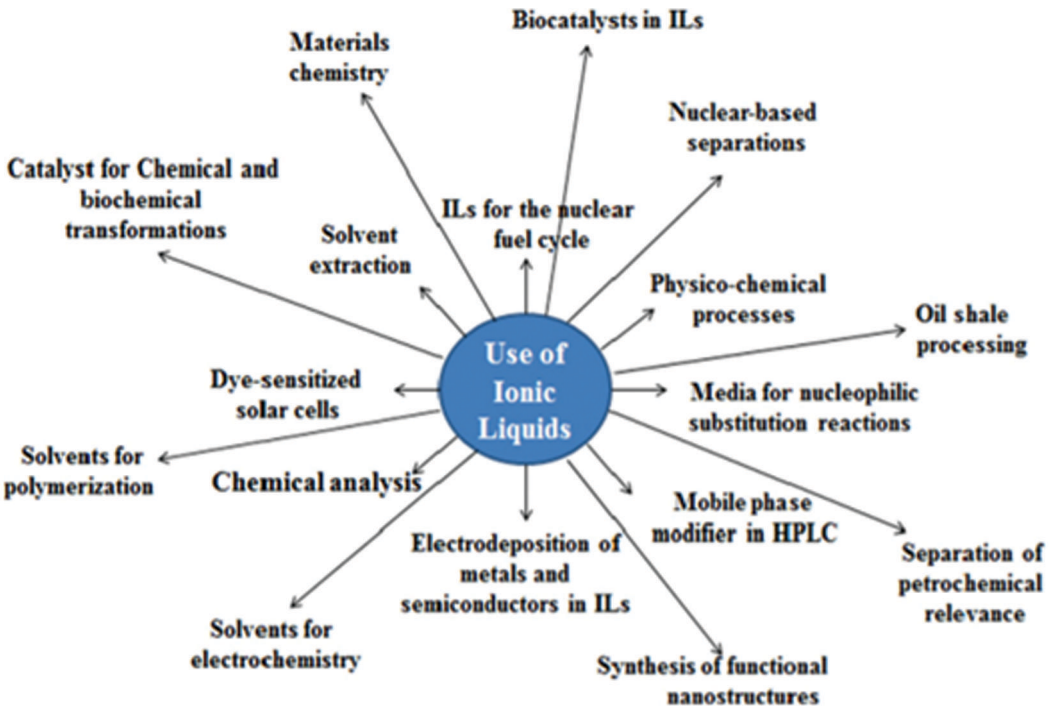

Figure 3. All applications in the science of the ionic liquids. Reproduced with permission from Reference (*2*). *Copyright 2017, Elsevier.*

All applications in the science of ionic liquids (ILs) are summarized in Figure 3. Due to remarkable properties mentioned of ILs, these compounds support the green chemistry idea of proposed by Paul Anastas and John Warner (*31*). One of the most remarkable scale regarding to the classifications of ILs has been prepared by Hajipour and Refiee (*32*). They were classified the ILs as: neutral ILs, acid ILs, basic ILs, ILs with amphoteric anions, functionalized ILs, protic ILs , chiral

ILs, supported ILs, bio-ionic liquids, poly-ionic liquids, and energetic ILs. Second classification was made by Suresh and Sandhu (*33*). These authors were classified them as only anionic and cationic ILs. In the last twenty years, many papers showing the excellent performances as corrosion inhibitors of ILs in various corrosive mediums were published. Although some traditional volatile compounds are widely used in the prevention of the corrosion of metal surfaces, their toxic effects cannot be ignored. If one considers both economic and performance situations of drugs, extracts and traditional corrosion inhibitors, it is clear that there is a need to develop new compounds and strategies for corrosion science. Owing to this need, the importance in corrosion science of ionic ILs has been understood more and more every day. It is reported that ILs have non-toxic nature, high conductivity, non-flammability stability as well as high thermal and chemical. Now, it can be easily understood why we added detailed information about electronic structure principles and parameters providing important insights for chemical reactivity analysis of the compounds in this chapter because the stable structure of ILs plays important role in terms of the superior properties they exhibit. Modifying the ions in the structure of the ILs, their corrosion inhibition performances are easily changed. It is well-known that Pearson classified the ions as hard and soft and the hardness value of an ion determines the nature and the power of the interaction with other ion or molecules of the mentioned ion. On organic molecules have extremely low solubility in the polar corrosive media while solubility of ILs is so high due to their ionic structure.

A Detailed Information about ILs as Corrosion Inhibitor for Metals Like Mild Steel, Aluminum, Copper and Zinc

Mild steel and iron is widely used in the industry. For that reason, the preventions of the corrosion of these metals is one of the important problems in the industry because these reactive materials undergo corrosive degradation in especially acidic medium. In the literature, a lot of article investigating the performances of ILs as corrosion inhibitor for mild steel are available. Olivares-Xometl and coworkers (*34*) synthesized two ILs with chemical formula 1,3-dioctadecylimidazolium bromide and N-Octadecylpyridinium bromide with the help of conventional and microwave-assisted reactions and showed that these inhibitors exhibity corrosion protection efficiency within 82–88% at 100 ppm for mild steel in 1 M sulfiric acid solution. Tuken (*35*) and coworkers investigated the inhibition performances of 1-ethyl-3-methylimidazolium dicyanamide (EMID) against the corrosion of mild steel and they proved that the obtained film from the mentioned IL protects sucessfully the metal during 120 hours exposure to 0.1 M H_2SO_4. Hua (*36*) analyzed the corrosion inhibition performance in the light of electrochemical impedance, potentiodynamic polarization and weight loss measurements of some alkylimidazolium based ILs such as 1-butyl-3-methylimidazolium chlorides (BMIC) and 1-butyl-3-methylimidazolium hydrogen sulfate ([BMIM]HSO_4) for mild steel corrosion. Author presented the efficiency order of these inhibitors as: [BMIM]HSO4 > BMIC. Zhang and coworkers (*37*) investigated The inhibition performance and mechanism of 1-octyl-3-methylimidazolium bromide ([OMIM]Br) and 1-allyl-3-octylimidazolium bromide ([AOIM]Br) for the corrosion of mild steel in 0.5 M H_2SO_4 using experimental and theoretical tools. They noted that these inhibitors act as modest cathodic inhibitors for mild steel. El Abedin and coworkers (*38*) presented a detailed study showing the anti-bacterial and anti-corrosion effects of the IL 1-butyl-1-methylpyrrolidinium trifluoromethylsulfonate. The authors explained that the mentioned inhibitors prevent the corrosion of mild steel forming a protective film. Ebenso and his team (*39*) published a comprehensive paper emhasizing the performance of some alkyl imidazolium

ILs containing tetrafluoroborate anion via DFT and Molecular Dynamics Simulations calculations and some well-known experimantal tools. In this study where quantum chemicaşl calculations are in good agreement with experimental observations and the results of Molecular Dynamic Simulations analysis, it was seen that the most hard inhibitor among namely1-ethyl-3-methylimidazolium tetrafluoroborate $[EMIM]^+[BF_4]^-$, 1-butyl-2,3-dimethylimidazolium tetrafluoro-borate $[BDMIM]^+[BF_4]^-$ and 1-decyl-3-methylimidazolium tetrafluoroborate $[C10MIM]^+[BF_4]^-$ is $[EMIM]^+[BF_4]^-$ and this compound is less effective against the corrosion of mild steel compared to others. Naderi analzyzed the corrosion behavior of mild steel in H_2SO_4 solution with 1,4-di [1′-methylene-3′-methyl imidazolium bromide]-benzene and the study confirmed the great performance as corrosion inhibitor of the mentioned compound. In a recent paper, Hajjaji and Kaya (*40*) synthesized some ILs named 1-phenethyl-3-(3-phenoxypropyl)-1H-imidazol-3-ium bromide [Imid-3PE]Br, and 1-phenethyl-3-(4-phenoxybutyl)-1H-imidazol-3-ium bromide [Imid-4PE] Br as corrosion inhibitors for mild steel. The authors proved that these molecules that their molecular structures are given in Figure3 exhibits good performance as corrosion inhibitors for mild steel.

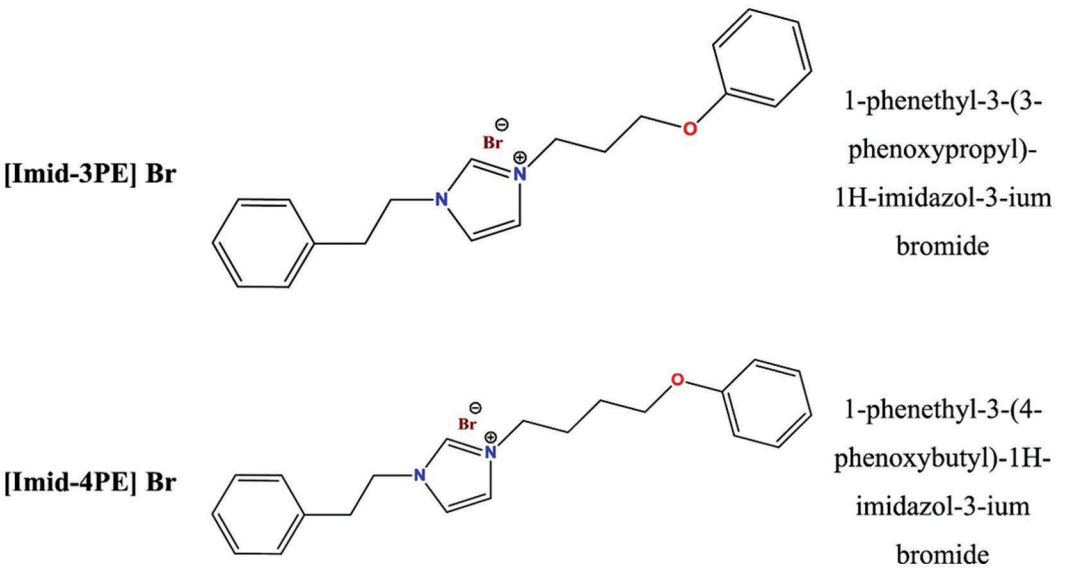

Figure 4. Molecular structures of the studied inhibitors. Reproduced with permission from Reference (40). Copyright 2021, Elsevier.

Aluminum also is among the most widely used metals in the industry because of its some important properties like low atomic mass and negligible standard electrode potential. ILs have been commonly used in the prevention of the corrosion of aluminum. This metal corrodes because of the existence of OH^- ions in especially aqueous alkaline medium. Zhang and Hua (*41*) investigated the corrosion inhibition performances of some newly synthesized ILs (1-butyl-3-methylimidazolium chlorides (BMIC), 1-hexyl-3-methylimidazolium chlorides (HMIC) and 1-octyl-3-methylimidazolium chlorides (OMIC)) against the corrosion of aluminum in 1M HCl. All experimental observations showed the corrosion inhibition efficiency ranking of these compounds as: OMIC > HMIC > BMIC. Shetty and Shetty (*42*) introduced an effective IL, namely 1,3-bis(2-oxo-2-phenylethyl)-1H-imidazol-3-ium bromide (OPEIB) for the prevention of the corrosion of 6061 Al-15alloy with the help of electrochemical impedance spectroscopy and potentiodynamic

polarization, scanning electron microscopy (SEM) and energy dispersive X-ray (EDX) spectroscopic methods. The obtained data showed that the mentioned IL acts an effective inhibitor in 0.1 M H_2SO_4 solution. Arellanes-Lozada and coworkers (*43*) noted that (poly(1-vinyl-3-dodecyl-imidazolium) ($PImC_{12}$), poly(1-vinyl-3-octylimidazolium) ($PImC_8$) and poly(1-vinyl-3-butylimidazolium) ($PImC_4$) hexafluorophosphate) compounds are effective corrosion inhibitors for aluminum corrosion in sulphiric acid. In this study, the effect also on corrosion inhibition performance of different alkylic chain lengths was investigated. Another study (*44*) was about the stability of aluminum in 1-butyl-3-methylimidazolium tetrafluoroborate IL and ethylene glycol mixtures. This research illuminated the stability of aluminum in the mentioned IL. Li and coworkers (*45*) noticed that via tetradecylpyridinium bromide, the corrosion of aluminum can be largely avoided in hydrochloric acid solution. The nature of surface interactions of some alkylimidazolium ILs with aluminum alloy Al 2011 was highlighted by Bermudez (*46*).

Although Copper and its alloys exhibit various applications such as building construction, electricity, electronics, coinages, ornamental and formation of industrial equipment due to their relatively good thermal, electrical, mechanical and corrosion resistance properties, in the presence of some ions such as chloride, sulfate and nitrate, this metal and its alloys may be easily exposed to corrosive attacks. A few important studies about the ILs against the corrosion of copper and zinc have been carried out in recent years. Qi-Bo and Yi-Xin (*47*) imparted to the literature some new ILs, namely 1-butyl-3-methylimidazolium hydrogen sulfate ($[BMIM]HSO_4$), 1-hexyl-3-methylimidazolium hydrogen sulfate ($[HMIM]HSO_4$), and 1-octyl-3-methylimidazolium hydrogen sulfate ($[OMIM]HSO_4$) and highlighted their inhibition efficiency against the corrosion of copper. The inhibition efficiency ranking of the mentioned compounds was determined as: $[OMIM]HSO_4$> $[HMIM]HSO_4$> $[BMIM]HSO_4$. In 2011, Gabler and coworkers (*48*) investigated the corrosion properties of some ammonium based ILs. The inhibition performances of two new protic (PIL) and four aprotic (APIL) namely triprotic di[(2-hydroxyethyl) ammonium] succinate (MSu) and the diprotic di[bis-(2-hydroxyethyl) ammonium] adipate (DAd) and 1-ethyl-3-methylimidazolium phosphonate ($[EMIM]EtPO_3H$); 1-ethyl-3-methylimidazolium octylsulfate ($[EMIM]C_8H_{17}SO_4$); 1-hexyl-3-methylimidazolium tetrafluoroborate ($[HMIM]BF_4$) and 1-hexyl-3-methylimidazolium hexafluorophosphate ($[HMIM]PF_6$) against the corrosion of copper were highlighted by Espinosa and coworkers (*49*). Murulana and coworkers (*50*) investigated the adsorption characteristic of 1-butyl-3-methylimidazolium tetrafluoroborate $[BMIM][BF_4^-]$ and 1-decyl-3-methylimidazolium tetrafluoroborate $[DMIM][BF_4^-]$ ILs on zinc surface using gravimetric analysis and DFT calculations based on the B3LYP functional. The authors noted that the mentioned ILs are quite effective against the corrosion of zinc surface in acidic medium. In addition to these studies, Liu and coworkers (*51*) investigated the electrodeposition of zinc films from ILs and ionic liquid/water mixtures and provided important insights regarding to the interaction with zinc films of the ILs. Acheverria (*52*) presented an important study showing that ILs increase the anti-corrosion performance of Zn-rich coatings.

Other Developments about ILs

In recent years, important papers emphasizing structure-performance relations of ILs have been published. An invite paper published in Chemical Reviews by Z. Lei and coworkers (*53*) includes quite important applications about ILs and their usage areas. A paper penned by Zhang (*54*) discussed the applications of ILs in molecular level and industrial level. Scarborough (*55*) published a comprehensive and useful paper including usage of quantum chemical methods in the prediction of energetic physical and spectroscopic properties of ILs. The mentioned paper reported that intermolecular forces are closely correlated with to physicochemical properties and they should be considered in the designing of new functional materials. Zhang (*56*) provided detailed information about nanoconfined ILs, which is one of the important classes. Song (*57*) and coworkers reported that thanks to great properties of ILs, the catalytic transformations can be performed more efficiently and with less environmental damage. Lei and coworkers (*58*) wrote a detailed review showing the usefulness of ILs in some important oxidation reactions as efficient catalysts and innovative green solvents. In separation process, the selection of suitable solvent for the study is quite important in terms of the obtaining of remarkable results. Coutinho (*59*) and his team presented a review article including the using of ILs as solvent in extraction and purification of important bioactive molecules. Important replies to the question "all ionic liquids are stable?" are given in the paper penned by Gao and coworkers (*60*). As to conclude, it can be written that ILs will find important applications in the future as well as today due to their great characteristic properties.

Conclusions

This chapter provides detailed information about the using as corrosion inhibitor of various types of the ILs. ILs with almost all the superior properties sought in a corrosion inhibitor have found important applications in many fields of the science. It is reported that these compounds, which can be easily designed and synthesized without toxic matters and processes have high inhibition performance against the corrosion of the metals like mild steel, copper, aluminum, magnesium and zinc and alloys of these metals. In addition to the scientific applications of ILs, the chapter includes remarkable information about important electronic structure principles and guiding rules used for the chemical reactivity analysis of molecules like Hard and Soft Acid-Base Maximum Hardness, Minimum Polarizability, Minimum Electrophilicity, Minimum Magnetizability, Maximum Entropy Principles and Maximum Composite Hardness Rule. Lastly, we propose the using some parameters like Fukui potential and magnetic susceptibility in the theoretical parts of the corrosion studies. The valuable proofs regarding to the performances of these parameters in corrosion science will be presented through some new paper projects of Kaya research group in the near future.

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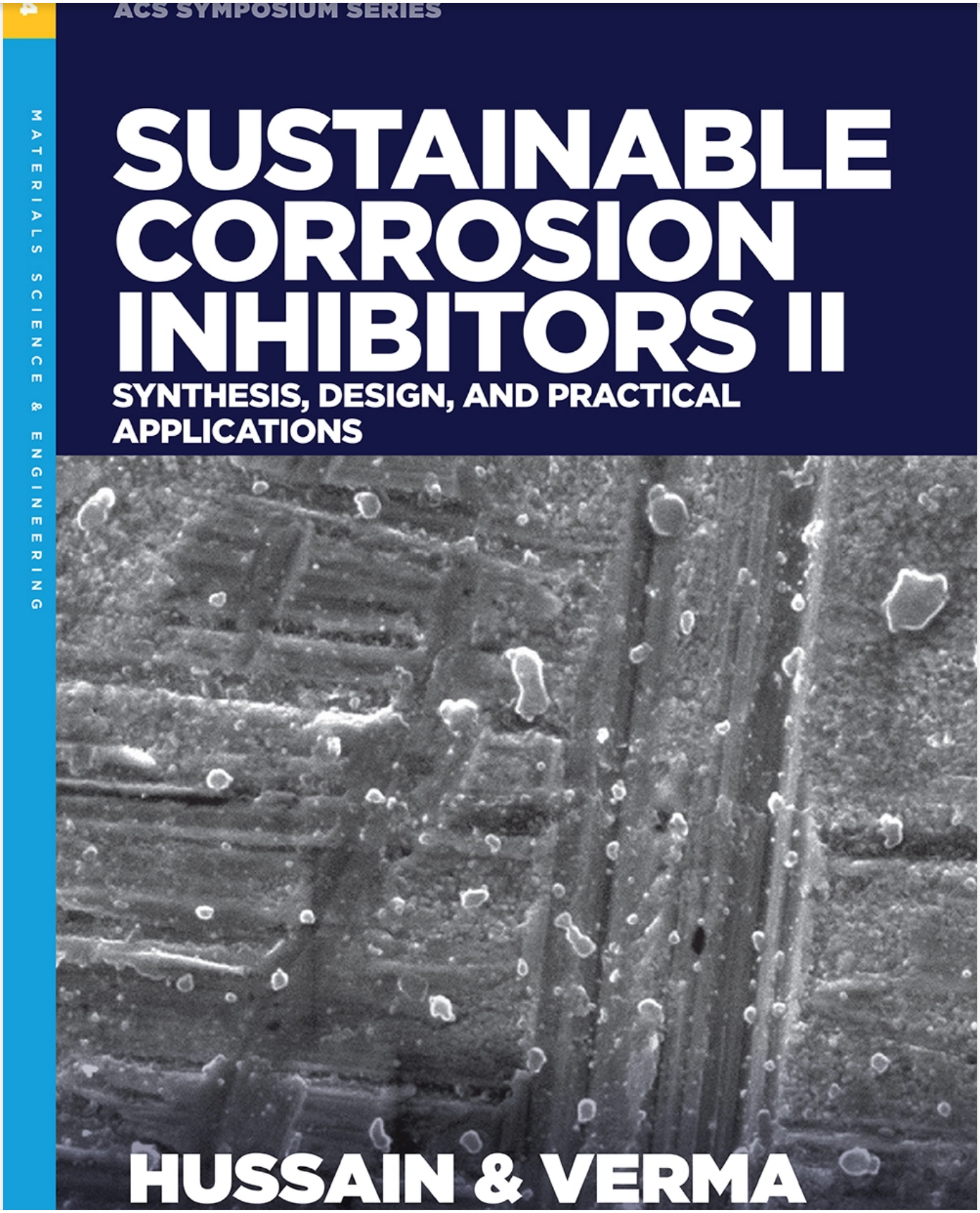

Sustainable Corrosion Inhibitors II. Synthesis, Design, and Practical Applications

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