Natural Honey as Eco-friendly Corrosion Inhibitor for Metals and Alloys - A Review

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Abstract
NH has the ability to control corrosion of various metals and alloys such as Al, CS, Cu, Cu Sn, MS, Sn and SS. It behaves as a CI in acidic, basic or neutral media. In the several studies herein reviewed, various techniques like WL, and electrochemical methods such as PDP and EIS, have been used to evaluate NH corrosion IE (%). Protective films have been analyzed by FT-IR, UV-vis spectroscopy, GC-MS, SEM and EDX methods. NH adsorption onto the metals and alloys surface has obeyed Langmuir's isotherm. PDP studies have revealed that NH may function as an anodic, cathodic or mixed type of CI, depending on the metal nature and on the corrosive environment.

Keywords: CI; EDX; EIS; FT-IR; GC-MS; NH; PDP; SEM; WL.

Introduction•

Corrosion is basically defined as the deterioration of any metallic material, due to a chemical or electrochemical attack by its corrosive environment. The loss caused by metals corrosion is huge for individuals, organizations and countries. These damages range from components or equipment breakdown, plant shutdown, loss of life and properties [1-3]. Al and its alloys are important materials, due to their high technological value and wide range of industrial applications, especially in aerospace, household and marine sectors. CS is frequently used for manufacturing pipe lines in petroleum industries. Cu is considered to be one of the most important metals, which is frequently used in different industrial applications. MS and SS corrosion studies have received considerable attention, because both are among the most commonly used metallic materials, particularly in automobile, food processing, chemical, construction and pharmaceutical industries [4, 5].

One of the most effective alternatives for the protection of metallic surfaces is the use of CI. A CI is generally referred to as a chemical substance that, when applied in small quantities to a corrosive medium, reduces the CR of a metal or alloy [6]. CI retard metal corrosion by adsorbing onto a metallic surface. The process is influenced by some factors, which include the CI's molecular size and Ct, nature of substituents, solution T and nature [7, 8]. Problems associated with traditional organic and inorganic CI, which limit their usage nowadays, include biotoxicity, environmentally unfriendly properties, high cost and non-availability on demand. Therefore, in line with environmental protection regulations, the new trend in

•The abbreviations list is in pages 371-372.

industry is nowadays orientated towards finding new ecologically harmless, green CI with low risk of pollution. These CI molecules consist of heterocyclic compounds with polar functional groups (e.g. N, S, O and P) and conjugated double bonds with different aromatic systems. Basically, these substances adsorb onto the metal surface, blocking the dissolution reaction in aggressive media. They are both physically and chemically active adsorbate type substances [9, 10].
Honey is a natural, golden brown beehive substance [11]. It is produced by honeybees (*Apis mellifera*) from flowers' nectar, which is a sweet, flavorful and viscose liquid [12, 13]. Honey has been used for centuries as a sweetener, flavoring agent and medicine [14]. It is a rich source of carbohydrates, and usually contains a rich diversity of minor constituents (minerals, proteins, vitamins and others), adding nutritional variety to human diets. It is used as antibacterial, antioxidant and antimicrobial agent.
Honey is also relatively cheap and readily available, so it fulfils all requirements for nontoxic CI. NH compounds offer interesting possibilities for CI, due to their safe use and high solubility in water [15]. NH has the ability to control the corrosion of a wide variety of metals and alloys such as Al, CS, Cu, CuSn, MS, Sn and SS. It behaves as CI in acidic, basic and neutral media.

Factors influencing metal corrosion

T and immersion time

T has an important influence on the corrosion phenomenon in metal surfaces. Immersion time is another factor that could modify IE (%). IE (%) of NH is calculated by using WL and electrochemical tests, such as PDP and EIS measurement. Polarization tests, such as PDP, are based on the evaluation and analysis of the current produced by a variable potential in a working electrode [16]. EIS provides more information such as mechanisms and different resistance values of the system. Various techniques like EDS, EDX, FT-IR and SEM have been used to analyze the nature of the protective film formed on the metal surface. SEM provides a clear comparison between metal surfaces with and without CI, as well as other morphological data.
The present work aimed to review the results regarding CI action of NH on some metals and alloys, such as Al, CS, Cu, CuSn, MS, Sn and SS, in various acidic, alkaline and neutral corrosive media. Different methods have been employed to evaluate CI process. The protective film has been analyzed by various surface analysis techniques.

Metals

Various metals and alloys have been used in CI studies such as Al [17-21], CuSn [22, 23], Cu [24], CS [25, 26], MS [27, 28], 304 austanic SS [29], steel [30] Q-235 steel [31], AISI-304 SS [32] and Sn [33].

Media

Usually, metals corrosion has been studied in various environments. NH has been used as CI in acidic [20, 27-30, 32] and neutral media [23, 31], SW [17, 19, 21] and NaCl [18, 22, 24-26, 33].

Techniques

EIS [17, 18, 21, 23, 30], PDP [17-26, 28-30, 33], SE [28, 30], WL [19, 21, 24, 25, 27, 29, 33], WL with T [30, 31] and with time [32] were used to assess NH as CI.

Surface film analysis

Films formed on metal surfaces were studied by various techniques, like EDS [17], EDX [22, 26, 30], FT-IR [17, 26, 32], GC-MS [28] and SEM [17, 22, 26, 29, 30].

Adsorption isotherms

Langmuir's adsorption isotherm [17-19, 24-26, 28-30, 33] was suggested. NH uses on various metals and alloys in different solutions are presented in Table 1.

Table 1: CI by NH.

Metal/alloy	Medium + additive	Used techniques	Type of inhibitor	IE(%)	Ref. no.
Al -Mg-Si	SW	PDP, EIS, LPR, FT-IR, SEM and EDS	Mixed	91.85 WL and 91.58 PDP	[17]
AA 5052	0.5 M NaCl	PDP, LPR and EIS	Mixed	81.7 PDP	[18]
AA 6061	Tropical SW	WL and PDP	Mixed	80.60 WL and 91.58 PDP	[19]
A 4%- Si	0.5 M H_2SO_4	PDP	--	88.79 PDP	[20]
AA 7631	Tropical SW	EIS, PDP and WL	Mixed	75.76 WL, 72.00 PDP and 91.72 EIS	[21]
CuSn	0.5 M NaCl	PDP, SEM and EDX	Mixed	--	[22]
CuSn	0.2 g/L Na_2 SO_4 + 0.2 g/L $NaHCO_3$	PDP and EIS	Mixed	89.58 PDP and 93.19 EIS	[23]
Cu	0.5 M NaCl	WL and PDP	Cathodic	89.0 WL and 97.6 PDP	[24]
A-106 CS	0.5 M NaCl	WL, PDP	Anodic	91.26 PDP	[25]
CS	0.5 M NaCl	PDP, SEM, EDX and FT-IR	Mixed	77.68 PDP	[26]
MS	1 N H_2SO_4 +1 N HCl	WL	--	54.0 WL in HCl	[27]
MS	1 M H_2SO_4	PDP, GC-MS and SE	Mixed	87.5 PDP	[28]
A315 MS + 304 austanic SS	0.5 M H_2SO_4	WL, PDP and SEM	Mixed	89.0 and 94.0 WL in MS and SS	[29]
Steel	1.0 M HCl + 0.1 M KI	WL with T, PDP, EIS, SEM, EDX and SE	--	--	[30]
Q-235 steel	Simulated brine solution	WL with T	--	93.0 WL	[31]
AISI-304 SS	17% HCl	WL with time and FT-IR	--	90.73 WL	[32]
Sn	3% NaCl	WL and PDP	--	--	[33]

GC-MS spectral study

[28] have studied NH as CI of MS in a 1 M H_2SO_4 solution. They have carried out GC-MS spectra of NH, and found 11 peaks corresponding to the following CI compounds: acetic acid, acetophenone, batyrolactone, benzeneaceta-aidehyde, butanoic acid 3-methyl and toluene, in high amounts; and benzaldehyde, benzene ethanol, benzene methanol, furtural and nonanal, in low amounts.

ATR-FTIR absorption spectral study

ATR-FTIR absorption spectra study of a film formed on the SS AISI 304 surface in a 17.0% HCl solution with 2.0% NH reveals 'O' atoms in functional groups (O–H, C–O, C=O), which are shown in Fig. 2 [32].

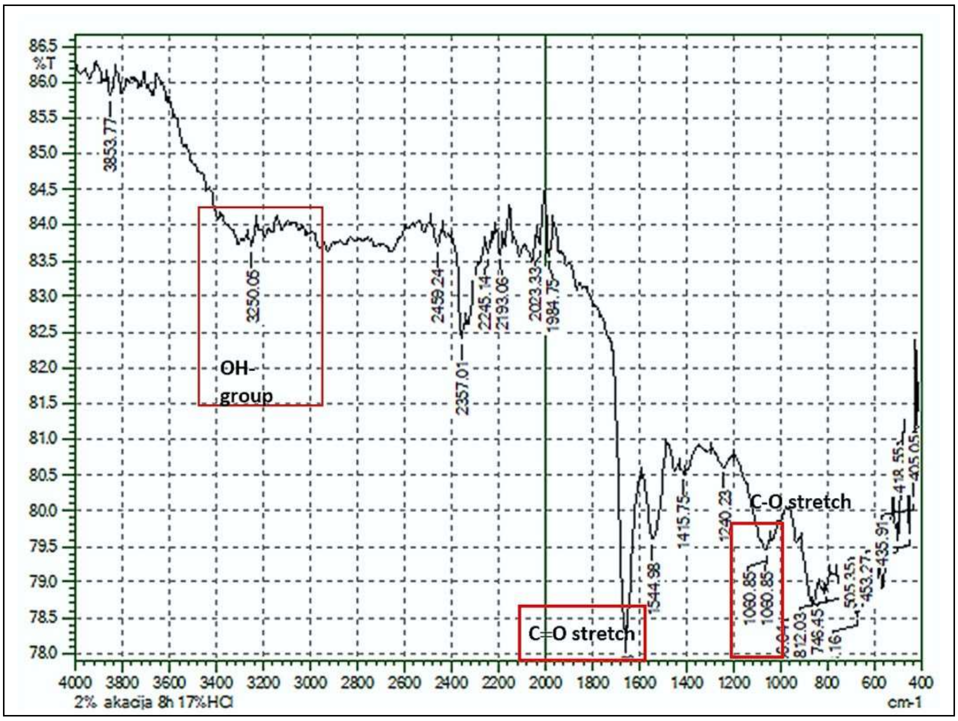


Figure 2: ATR-FTIR absorption spectra of a film obtained from SS AISI 304 surface, after 8h exposure to a 17.0 wt.% HCl solution with 2.0 wt.% Acacia honey [32].

PDP study

CuSn surface exhibited better CI with NH, as shown by PDP results from Tafel curves in $Na_2SO_4/NaHCO_3$ (Fig. 3). Tafel curves shown in Fig. 3 indicate that NH acts mainly as a mixed-type of inhibitor with predominant control of the cathodic reaction [23].

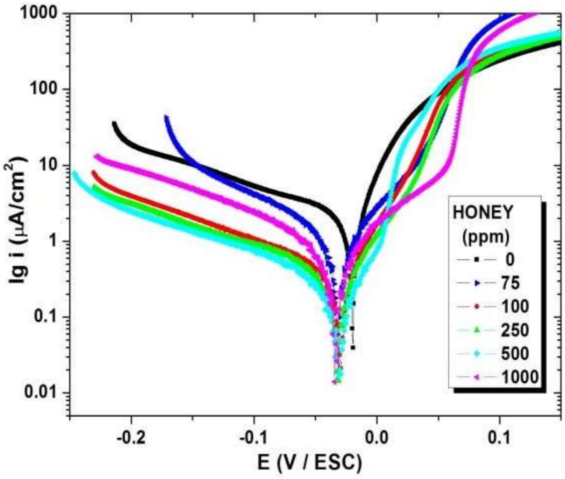


Figure 3: Polarization curves (Tafel curves) for CuSn in a $Na_2SO_4/NaHCO_3$ (pH 5) solution without and with NH in various Ct [23].

EIS study

EIS diagram of CuSn immersed in a $Na_2SO_4/NaHCO_3$ (pH-5) solution without and with NH in various Ct is depicted in Fig. 4 [23].

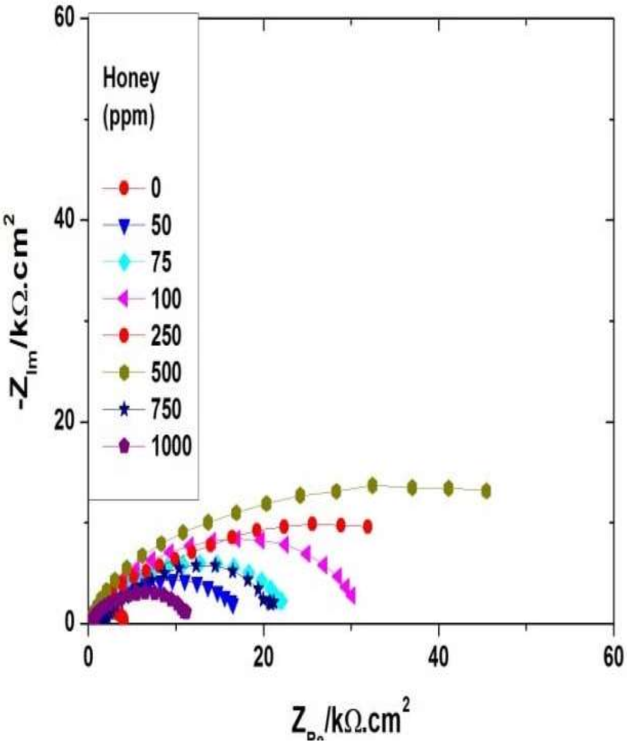


Figure 4: EIS diagram of CuSn in a $Na_2SO_4/NaHCO_3$ solution without and with NH in various Ct [23].

Chemical composition of NH

NH composition basically varies with the floral source, but seasonal, environmental and processing conditions are also important [34-36]. Until now, about 600 compounds have been characterized in different NH [37]. Phenolic acids and polyphenols are plant-derived secondary metabolites.

Carbohydrates

Honey comprises three kinds of sugar: fructose, glucose and sucrose, with Ct of 41, 34, and from 1 to 2%, respectively [38]. Sugars' structure is shown in Fig. 1.

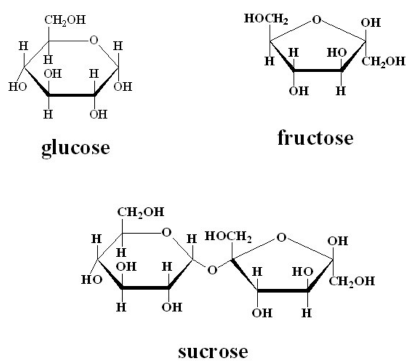


Figure 1: Structure of glucose, fructose and sucrose.

Amino acids and proteins

Proteins in honey may have a very complex structure or simple compounds, i.e., amino acids [39] and proteins, of which content is relatively low, at the most, 0.7%.

HMF (Hydroxy methyl furfuraldehyde)

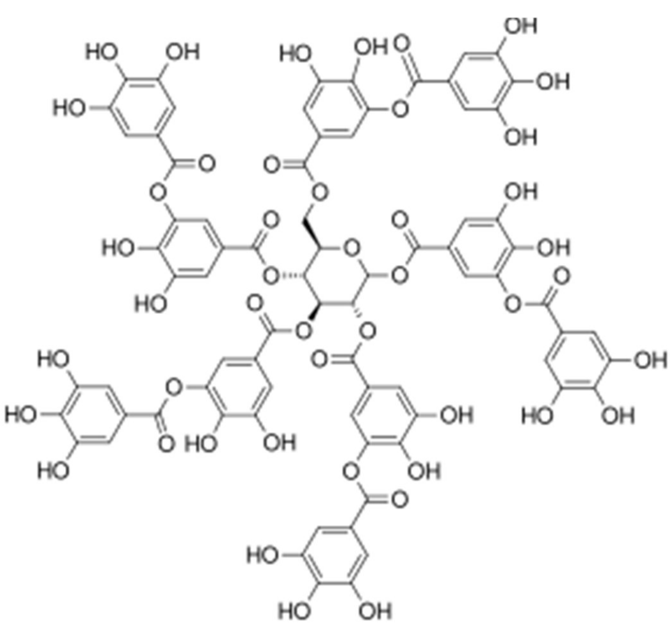
HMF is a six-C heterocyclic organic compound containing both aldehyde and alcohol (hydroxy methyl) functional groups [40].

Minerals and trace elements

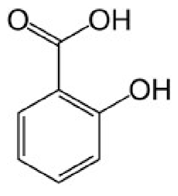
NH is a relatively healthy and easily digestible foodstuff containing a range of nutritiously important complementary elements: saccharides, organic acids, amino acids, polyphenols, mineral matter, colors, aromatic substances, trace amounts of fat and some valuable but unstable compounds, such as enzymes, hormones, some vitamins and a few minor compounds [18, 20, 23, 33, 41, 42]. NH contains most minerals: Ca, Cl, Cu, Fe, K, Mg, Mn, Na, P, S and Si [43]. It is is non-toxic, and it contains phenolic compounds that make it a good source of antioxidants which prevent oxidation reaction of the corrosion process [33].

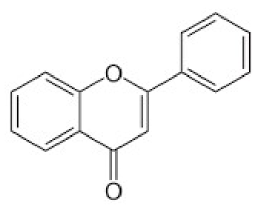
Mechanism of CI by NH

The major chemical constituents of NH responsible for CI are polyphenol, phenolic acid, tannins, saponins and flavonoids [18, 24, 29]. The structure of NH's main CI constituents, which protect the passive film on the metal surface, is shown in Fig. 5 [29].



(a) Polyphenol.





(b) Phenolic acid. **(c)** Flavonoid (tannin).

Figure 5: Structure of main CI constituents in NH- **(a)** polyphenol; **(b)** phenolic acid; **(c)** flavonoid (tannin) [29].

This film serves as a barrier between the sample and the corrosive environment interface, thus preventing and/or stifling corrosion reactions of anodic (oxidation/dissolution) and cathodic (reduction) processes [44]. It is hard to decide which of these components is responsible for CI. It may be a single component, more than one or even all of them acting in synergy. A FT-IR study [17, 26, 32] indicated that NH is an organic compound containing polar groups such as N, S and O, as well as heterocyclic containing conjugated double bonds that have made it suitable as good CI, due to the presence of C=C group, N, S and O atoms in its molecules. These results are supported by other researchers [24, 25, 28, 33, 42]. It was reported by [45] that the constituent molecules of NH contain O atoms in functional groups (O–H, C–H, C–O, C=O) which meet general features of typical CI.

Conclusions

This review paper summarized the research works carried out by various researchers on the CI of various metals and alloys in different acidic, neutral and alkaline media by NH. IE(%) of NH was calculated using EIS, PDP and WL methods. Other techniques like EDX, FT-IR, GC-MS and SEM were also used to study the nature of the surface film produced on metals. Langmuir's adsorption isotherm was found to be the most common. NH behaved as anodic, cathodic and mixed-type CI, and its molecules are adsorbed on both anodic and cathodic sites at the metal surface. NH obtained corrosion IE(%) values above 54.0%, most of them from 72.0 to 97.6%. Results given by WL data were in good agreement with data obtained from PDP and EIS methods.

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Author's contribution

R. T. Vashi: conception and design of the analysis; collected and analyzed data; wrote the paper as single author.

Abbreviations

AFM: atomic force microscope
Al: aluminum
ATR: attenuated total reflectance
C: carbon
Ca: calcium
CI: corrosion inhibition/inhibitor
Cl: chloride
CR: corrosion rate
CS: carbon steel
Ct: concentration
Cu: copper
CuSn: bronze
EDS: energy dispersive spectroscopy
EDX: energy-dispersive X-ray spectroscopy

EFM: electrochemical frequency modulation
EIS: electrochemical impedance spectroscope
Fe: iron
FT-IR: Fourier-transform infrared spectroscope
GC-MS: gas chromatography mass spectrometry
H: hydrogen
H_2SO_4: sulfuric acid
HCl: hydrochloric acid
IE(%): inhibition efficiency
K: potassium
LPR: linear polarization resistance
MS: mild steel
Mg: magnesium
Mn: manganese
N: nitrogen
Na: sodium
NaCl: sodium chloride
NaOH: sodium hydroxide
NH: natural honey
O: oxygen
OCP: open circuit potential
P: phosphorous
PDP: potentiodynamic polarization
S: sulphur
SE: synergistic effect
SEM: scanning electron microscopy
Si: silicon
Sn: tin
SS: stainless steel
SW: sea-water
T: temperature
UV-vis: ultraviolet-visible spectroscope/spectrophotometry
WL: weight loss

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