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Strengths and Weaknesses of Three Different Popular Acid Base Theories: A Comparation Study

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Abstract

Arrhenius, Bronsted-Lowry, and Lewis acid–bases theories are the most popular ones in Chemistry and studied by undergraduated students in Element Chemistry. A comparison study is needed to find each strength and weakness to stimulate analysis method and application developments. Reference searching method was conducted to obtain the informations. Result of study shows that Bronsted-Lowry and Lewis are focused on reaction by donor/acceptor process, independent of solvent and phase. Arrhenius is more concerned on dissolved product and applicable in water solvent only. Bronsted acid/base strength is determined as K_a/K_b mathematically and experimentally but Lewis acidity is a sequence based on K_f, K_{BA}, and ΔH. All Bronsted acids contain Lewis acid and base, all Bronsted bases are Lewis ones, but not vice versa. Bronsted-Lowry reactions are the broken bonds, but Lewis ones can be with or without the breaking. Lewis and Bronsted-Lowry reactions support cementation, metal complexe and organometal synthesis, Arrhenius and Bronsted-Lowry support rock mineral activations, and those all three theories are suitable for metal ion adsorption by carboneous materials and metabolism reactions in erythrocyte. Based on the study, Lewis indicated the most superiority in both concept and application.

Keywords: Arrhenius; Bronsted - Lowry; Lewis; concept, application.

1.Introduction

Acid-base theory is one of topics which are studied by undergraduated students in Inorganic Chemistry field. the The acid-base theory is important to learn because many life aspects involve acid and base, including acid/base substances, acid-base adducts, and acid-base reactions. For example, acid compounds such as ascorbic acid, folic acid, citric acid, tartaric acid in the fruits [1], complexe Fe(II) ion (hemoglobin) in the blood [2], anthocyanine (acid/base indicator) in the flowers [3]. acid and basic drugs in medicine [4], formation of complexe metal ions in metal spectrophotometric analysis [5], $CaCO_3$ deposition as stalagtite and stalagmite in the cave [6], cation exchange reactions of metal cations and proton on the acid soil [7], etc.

Ten acid base theory types have been made from 1776 until 1960, sequently including Liebig, Arrhenius, Bronsted-Lowry, Lewis, Ingold–Robinson, Lux–Flood, Usanovich, Solvent system, and Frontier orbitals. The six ones of all those concepts are related to donor and acceptor terms. Among those six concepts, the only Bronsted-Lowry concept defined acid as donor and base as acceptor, while the other five ones (including Lewis) have the opposite terms [8,9]. Although there are 10 acid base concepts, the only 3 ones are studied popularly including Arrhenius, Bronsted-Lowry, and Lewis ones. Among those theories, Arrhenius is not connected to donor/acceptor.

Some journals have discussed those three acid base topics individually, including about Bronsted-Lowry concept [10-12], Lewis concept [13-15], Lewis acidity analysis method [16-20], Bronsted-Lowry acidity analysis method [21-22], Bronsted-Lowry applications [23-24], and Lewis applications [24-27]. Bronsted-Lowry and Lewis theories have been studied together especially their roles in the same organic synthesis [28-29]. A comparison study of those three concepts is needed to understand each shortage and strength. Their strengths will inspire more creations and modifications for synthesis of functional inorganic materials based on the acid base reactions. Their shortages will be a consideration to create additional theory supported by new analysis methods.

In this paper we compared those Arrhenius, Bronsted-Lowry, and Lewis acid-base including their concepts and some applications. For the concept, advantages and disadvantages of those concepts are studied. For the application, their roles are investigated especially related to inorganic synthesis, organometal synthesis, and human physiology. Result of this comparison study will be one of references in Element Chemistry course for undergraduate students in Inorganic Chemistry

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field. Purpose of this study is to improve understanding of the undergraduate students in choosing the right concept to explain the acid base reactions.

2. Comparisons as the concept

Swante Arrhenius (1884) defined that acid is a substance which produces hydrogen ions in water [30] or yields proton in aqueous solution [8] or adds concentration of H^+ or H_3O^+ ions in water [31] or forms hydrogen ion or hydronium in aqueous solution [9]. Arrhenius base is a compound which yields hydroxide ions in aqueous solution [8], or adds concentration of OH^- ions in water [31] or forms hydroxide ion in aqueous solution [9]. It means that Arrhenius acid-base theory focus on product of the dissolved proton or hydroxide in the water.

Johannes Brønsted and Thomas Lowry (1923) proposed the acid–base reaction as ion hydrogen transfer [30]. Bronsted-Lowry defined acid as a species which has a tendency to lose a hydrogen ion and a base as a species which a trend to gain a hydrogen ion [8]. In other word, acid is a proton (H^+) donor and base is a proton (H^+) acceptor. Each can be called briefly as Bronsted acid and Bronsted base, respectively [30]. Both Bronsted acid and base can be molecule or ion [31].

Based on those definitions, Bronsted-Lowry focus on the reaction process, whereas Arrhenius focus on the product. Bronsted-Lowry concept has a superiority over Arrhenius concept because it doesn't depend on solvent type or solvent presence and applicable in gas, liquid, or solid phases, about the dissolved ones or the precipited ones. In other side, the Arrhenius concept is only useful for aquatic solution. For example, in Arrhenius concept, HCl is acid due to proton production in the water; NaOH is a base due to hydroxide ion formation in the water, and neutralization reaction of both dissolved proton and dissolved hydroxide ions in aqueous solution [8]. In other side, Bronsted-Lowry concept states HCl as an acid due to the proton donor, NH_3 is a base due to the proton acceptor, and reaction of HCl and NH_3 can occur NH_3 liquid to form NH_4Cl [32].The chemical can be written as follows:

Arrhenius : $\underset{\text{Arrhenius acid}}{HCl\ (aq)} + \underset{\text{Arrhenius base}}{NaOH\ (aq)} \longrightarrow \underset{\text{Salt}}{NaCl\ (aq)} + \underset{\text{Water}}{H_2O\ (l)}$

Bronsted – Lowry : $\underset{\text{Bronsted acid}}{HCl\ (\text{dissolved in } NH_3)} + \underset{\text{Bronsted base}}{NH_3\ (l)} \longrightarrow NH_4Cl\ (\text{dissolved in } NH_3)$

Another superiority of Bronsted-Lowry concept over Arrhenius is that it can explain a substance (including a solvent compound) can has a property as acid or base (amphoteric) depend on the other reactant properties. Even, the same substances can react each other as base and acid. These properties are impossibly explained by Arrhenius concept. For example, the H_2O molecule is acid (proton donor) toward NH_3, but a base (proton acceptor) toward HF. H_2O molecules can react each other as acid and base to form H_3O^+ and OH^- as presented in Figure 1 [30]. The same explanations are applicable for amphoteric substances such as NH_3, H_2SO_4, HF, CH_3OH, CH_3CN, and CH_3COOH. The acid base reaction among themselves are stimulated by dipole attraction force due to their polar properties. Acetic acid (CH_3COOH) can also react as base in the acetic acid glacial (100% acetic acid) with other substances which are categorized as the stong acids in the water, such as H_2SO_4, HNO_3, HCl, $HClO_4$ (Figure 2). Sequence of their acid strength in glacial acetic acid is $HClO_4$ > HCl > H_2SO_4 > HNO_3 [8].

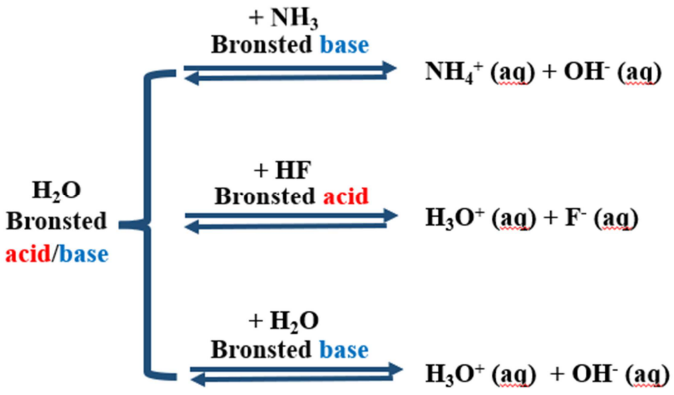

Fig. 1. Amphoteric property of H_2O in Bronsted-Lowry concept [30].

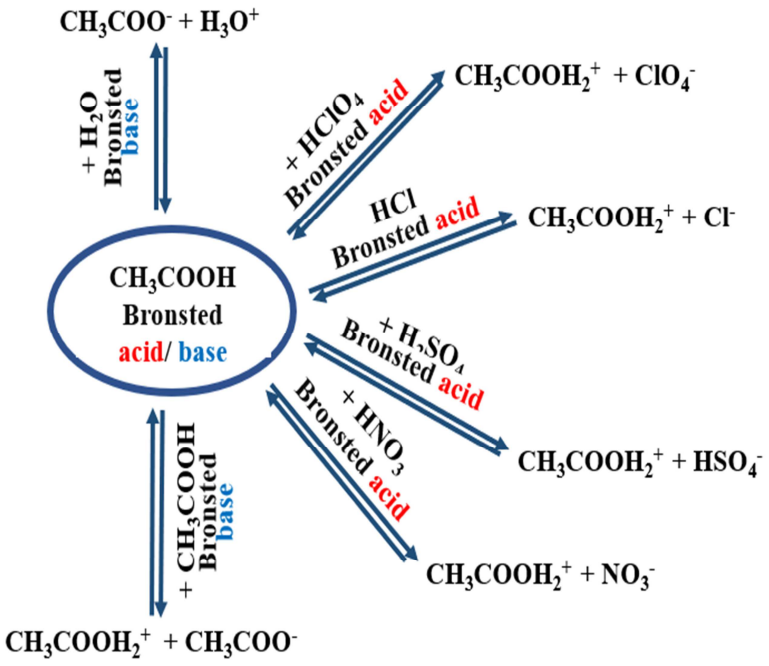

Fig. 2. Amphoteric properties of acetic acid in Bronted-Lowry concept [8].

Arrhenius modern concept used terms of "form, yield, and produce" in acid and base definitions which indicates applicable for substances which have no hydrogen for the acids or hydroxide for the bases because the consideration is not the reaction process but more about the product. For example, the acid oxides (non metal oxides) such as SO_2 and CO_2 are Arrhenius acids due to production of proton or hydronium in the water [30]. However, those oxides are not Bronsted acids due to unable to transfer proton. The Bronsted acids must contain H atoms because they are proton donors, therefore Bronsted Lowry concept more considers the process than the products. The reaction examples can be written as follows:

$$CO_2\,(g) + H_2O\,(l) \longrightarrow H_2CO_3\,(aq)$$

$$\underset{\text{Bronsted acid}}{H_2CO_3\,(aq)} + \underset{\text{Bronsted base}}{H_2O\,(l)} \rightleftharpoons H_3O^+\,(aq) + HCO_3^-\,(aq)$$

$$\underset{\text{Arrhenius acid}}{CO_2\,(g) + H_2O\,(l)} \longrightarrow \underset{\text{Dissolved hydronium}}{H_3O^+\,(aq)} + HCO_3^-\,(aq)$$

In other side, the base oxides (metal oxides) such as Na_2O, CaO, etc are both Arrhenius and Bronsted bases. In Arrhenius concept, they produce the hydroxide ions in the water and in Bronsted Lowry concept, they are the donor proton toward H_2O solvent. The reactions are as follows:

$$\underset{\text{Arrhenius/ Bronsted base}}{Na_2O\,(s) + H_2O\,(l)} \longrightarrow \underset{\text{Dissolved hydroxide}}{Na^+\,(aq) + OH^-\,(aq)}$$

Al_2O_3 is reactive toward water and produces the undissolved hydroxide, therefore Al_2O_3 is not Arrhenius base. In other side, Bronsted–Lowry keeps succesfully explaining the Al_2O_3 as Bronsted base due to proton donor toward H_2O and toward proton in acid solution. Al_2O_3 is called amphoteric oxide due to its reactivity to acid and base. In this case, Bronsted Lowry concept can show its superiority over Arrhenius that principally in the base solution Al_2O_3 acts as Bronsted base toward H_2O as the Bronsted acid and the hydroxide ions as the ligands to form the dissolved metal complexe anions (Figure 3).

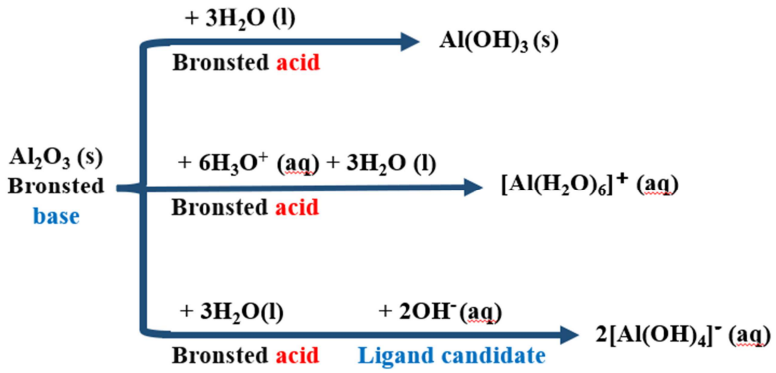

Fig. 3. Amphoteric property of alumina in Bronsted-Lowry concept [30].

G.N. Lewis (1930) defined a base as an electron-pair donor and an acid as an electron-pair acceptor [8,9, 33]. The Lewis acid includes metal ions and the main group compounds [1]. A proton (H^+) is also a Lewis acid because it can attack an electron pair, such as the pair in NH_3 to form NH_4^+. It means that every HA Bronsted acid always contains Lewis acid (H^+) and Lewis base (A^-). Therefore, the HA Bronsted acid is exactly not Lewis acid but always exhibits Lewis acidity. However, all BL bases are Lewis bases because all proton acceptors are also the electron pair donors [30]. For example, in the reaction of the Bronsted acid CH_3COOH and Bronsted base NH_3, we can see that NH_3 is also Lewis base because it use its lone pair to make reaction with H atom of the acetic acid which releases it as the proton to NH_3 to form NH_4^+ (Figure 4).

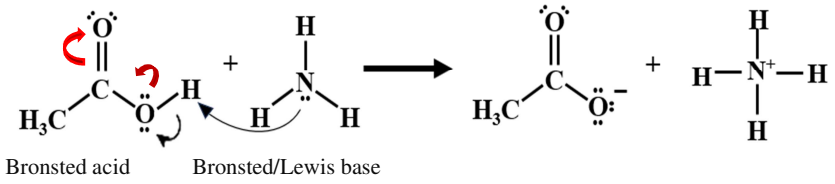

Fig. 4. Reaction mechanism of Bronsted acid (CH_3COOH) and Bronsted/Lewis base (NH_3) [34].

In Figure 4, O atom in C=O of acetic acid attract pi bonding to become lone pair which creates positive charge on centre C atom. This charge attracts lone pair of O atom on hydroxide to create new π bonding of C=O. This new bonding stimulates changing from bonding pair of O-H to lone pair of O atom which releases proton. Attraction force of positive dipole of H on hydroxide and negative dipole of N atom on NH_3 also supports deprotonation of acetic acid.

Superiority of Lewis concept over Bronsted-Lowry concept is proved by presence of the compounds as Lewis acid which do not contain hydrogen such as BF_3 (Figure 5) or contain hydrogen but not the proton donor such as BH_3 (Figure 6).

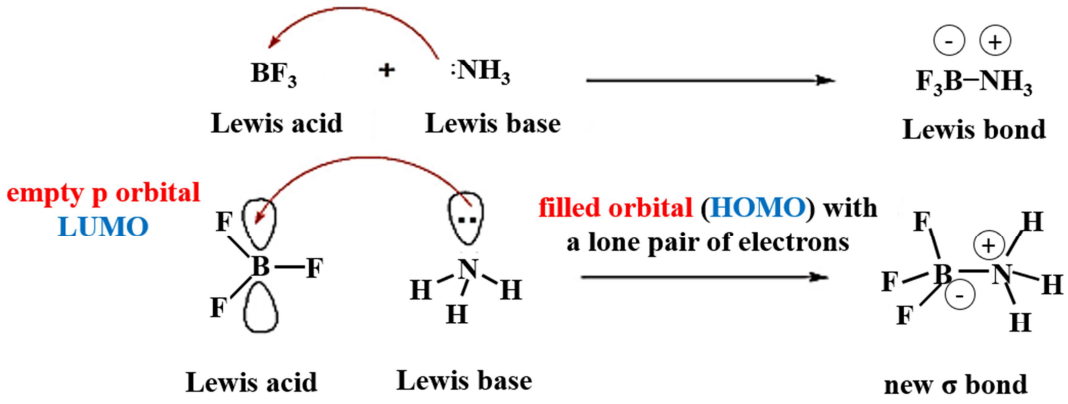

Fig. 5. Reaction of Lewis acid (BF_3) and Lewis base (NH_3) [35].

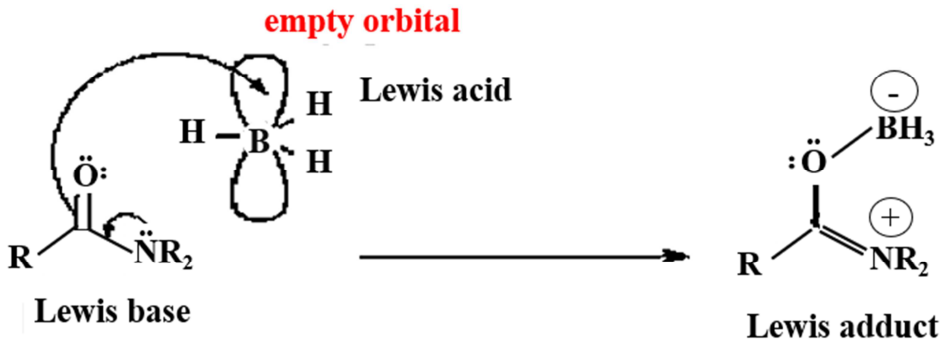

Fig. 6. Reaction of Lewis acid (BH_3) and Lewis base ($RONR_2$) [35].

In Figure 5 and 6, B atom is Lewis acid because it has potency to create one more covalent bond to achieve octet rule. This potency is caused by the provided orbitals (3s, $3p_x$, $3p_y$, and $3p_z$) in its valence shell to form a hybrid orbital of sp^3. This Lewis acid characteristics attracts lone pair of N atom on NH_3 (Figure 5) and π bonding of C=O on $RCONR_2$ (Figure 6) to form new covalent bonds of B-N (Figure 5) and B-O (Figure 6). Both reactions in Figure 5 and Figure 6 are not able to be explained using both Arrhenius and Bronsted-Lowry due to lone pair transfer without water.

Existence of the metal ions as the Lewis acids indicates superiority of Lewis concept over the Bronsted-Lowry one due to no proton. However, Bronsted-Lowry is better to explain hydrolysis reaction in the aqueous solution of the salts why their solutions are acid. If a salt (example $AlCl_3$) is solved in water, the Al^{3+} ions (Lewis acid) and the H_2O molecules (Lewis base) make coordination bonding to form the $Al(H_2O)_6^{3+}$ complexe cation. This complexe is a Bronsted-Lowry acid due to some factors including: 1). Hydrogen bonding of the H_2O ligands and the H_2O solvents, 2). Attraction of electron density from O atoms of H_2O ligands by Al^{3+}, 3). Repulsion of Al^{3+} and positive dipoles of H atoms in H_2O ligands. Those three factors make the bonds of O-H in the ligands weaker and the complexe can release H^+ toward H_2O solvents to form H_3O^+ in the Bronsted-Lowry acid base reactions named hydrolysis reactions [36] as follows:

$$AlCl_3\ (s) + nH_2O\ (l) \longrightarrow Al^{3+}\ (aq) + 3Cl^-\ (aq)$$

$$\underset{\text{Lewis acid}}{Al^{3+}\ (aq)} + \underset{\text{Lewis base}}{6H_2O\ (l)} \rightleftharpoons \underset{\text{Lewis adduct}}{[Al(H_2O)6]^{3+}\ (aq)}$$

$$\underset{\text{Bronsted acid}}{Al(H_2O)_6^{3+}(aq)} + \underset{\text{Bronsted base}}{H_2O\ (l)} \rightleftharpoons [Al(H_2O)_5OH]^{2+}\ (aq) + H_3O^+\ (aq)$$

The hydrolysis reaction between the $Al(H_2O)_6^{3+}$ cations and H_2O solvent molecules are the Bronsted-Lowry acid base reaction. However, it principally also involves H_2O solvent as Lewis base which makes reaction with Lewis acid H atom positive dipole of H_2O ligand of the metal complexe ion. The reaction mechanism shown in Figure 7.

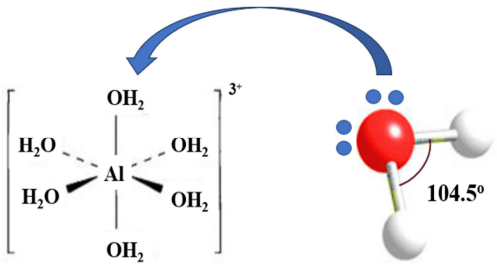

Fig. 7. Lone pair transfer from Lewis base H_2O to Lewis acid $[Al(H_2O)_6]^{3+}$[30, 36].

In Figure 7, There is bond hydrogen between dipoles of H_2O solvent and ligands. This molecular attraction force weakens covalent bond in H_2O ligand molecule. This weakening of bond is also supported by the coordination bond between Al^{3+} and O atom in H_2O ligand. Thus, the metal complex releases proton to the solvent to form deprotonated complex and H_3O^+.

Although same about transfer between donor and acceptor, concentration of the transferred proton or hydroxide in Bronsted-Lowry concept can be calculated mathematically based on the acid base equilibrium reactions and measured directly through analysis method, whereas impossible to determine the transferred electron pair concentration in Lewis concept. The examples of mathematics calculation for determination of the transferred proton and hydroxide [37, 30] are given as follows:

$$\underset{\text{Bronsted acid}}{HA\ (aq)} + \underset{\text{Bronsted base}}{H_2O\ (l)} \rightleftharpoons H_3O^+\ (aq) + A^-\ (aq) \qquad K_a = \frac{[H_3O^+][A^-]}{[HA]\,[H_2O]} = \frac{[H^+][A^-]}{[HA]}$$

$$\underset{\text{Bronsted base}}{B\ (aq)} + \underset{\text{Bronsted acid}}{H_2O\ (l)} \rightleftharpoons BH^+\ (aq) + OH^-\ (aq) \qquad K_b = \frac{[BH^+][OH^-]}{[B]\,[H_2O]} = \frac{[BH^+][OH^-]}{[B]}$$

Based on those formulas, the transferred H^+ or OH^- concentration can be calculated mathematically by using data of K_a or K_b, the resulted A^- or BH^+, and the remain HA or B, respectively. Each proton and hydroxide concentration can be also measured directly using pH meter or acid base titration analysis method.

Strength of Bronsted acid and Bronsted base can be also measured directly from its acidity constant (K_a) and its basicity constant (K_b), respectively, whereas it can't be done for Lewis acid and Lewis base. A substance which has Ka > 1 (pK_a < 1) is a strong acid because it is regarded as fully deprotonated in its solution so that the acid concentration can be negligible. In other side, a substance with K_a < 1 (pK_a >1) is a weak acid due to hard deprotonation reaction so that the acid reactant is more favour [30]. In Bronsted-Lowry concept, acidity strength is applicable for molecules and ions as listed in Table 1 and Table 2.

Table 1: Acidity constants of some Bronsted acid molecules and ions in their aqueous solution at 25°C

No.	Bronsted acid	K_a	Strength
1.	H_2SO_4	~10^2	Strong acid
	HSO_4^-	1.20 X10^{-2}	Weak acid
2.	H_3PO_4	7.52 X 10^{-3}	Weak acid
	$H_2PO_4^-$	6.23 X 10^{-8}	Weak acid
	HPO_4^{2-}	2.20 X 10^{-13}	Weak acid

Source: **[32]**

Table 1 shows that both H_2SO_4 and H_3PO_4 experience decreasing of Bronsted acidity after deprotonation. Both acid substances have oxy (S=O, P=O) and hydroxide(S-OH, P-OH) functional groups. The deprotonated hydroxide groups stimulate resonance structure. This resonance decreases formation of positive charge on S atom which weakens attraction to bonding electron pair of S-O which finally strengthen bonding of O-H and more difficult to release proton. The resonace structures of HSO_4^- , $H_2PO_4^-$, and HPO_4^{2-} are shown from Figure 8 to Figure 10, respectively.

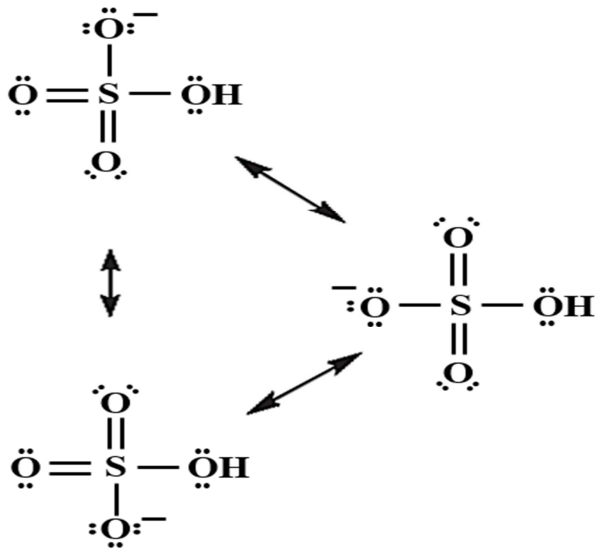

Fig. 8. Resonance structure of HSO_4^- [30].

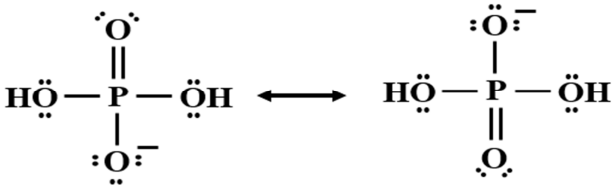

Fig. 9. Resonance structure of $H_2PO_4^-$ [38].

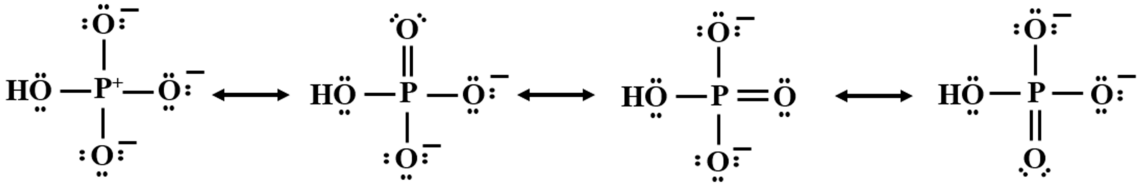

Fig. 10. Resonance structure of HPO_4^{2-} [39].

By instrument development, pKa was determined by ^{19}F NMR Spectroscopy, for example, pK_a of a fluorinated binaphthyl-derived phosphinic acid [40]. Bronsted acidity was determined by UV-Vis spectrophotometry by calculating the Hammett function (H_o) comparison of relative acidity to sulfuric acid [41]. Bronsted acid site of solid was determined by FTIR spectrometry, for example for Zr-Si oxide nanoparticles [42].

Table 2: Acidity constants for some Bronsted acids of the metal complexe ions

No.	Bronsted acid	M^{n+} radii (pm) *	pK_a	Acid Strength
1.	$[Co(H_2O)_6]^{3+}$	75	2.92	Weak acid
	$[Cr(H_2O)_6]^{3+}$	76	4.29	Weak acid
	$[Sc(H_2O)_6]^{3+}$	89	4.30	Weak acid
2.	$[Cu(H_2O)_6]^{2+}$	87	8.00	Weak acid
	$[Co(H_2O)_6]^{2+}$	89	9.65	Weak acid
	$[Mn(H_2O)_6]^{2+}$	97	10.59	Weak acid
3.	$[Mg(H_2O)_6]^{2+}$	86	11.41	Weak acid
	$[Sr(H_2O)_6]^{2+}$	132	13.18	Weak acid
	$[Ba(H_2O)_6]^{2+}$	149	13.36	Weak acid

Source: **[43], * [9]**

Table 2 presents the examples of pK_a which are only affected by charge and metal cation radii. The other examples which need discussion by involving the other influencing factors such as d^n , high spin, and low spin are not listed In Table 2 because this paper will be used for reference of Element Chemistry (semester 3), while Coordination Chemistry will be studied in semester 4. The acid strength of the complexes is affected by coordination bonding strength of M^{n+} and H_2O ligand. The stronger bonding of M-OH_2 in the complexe, the weaker bonding of OH in H_2O ligand, the easier releasing of proton from ligand. The metal cation charge of 3+ have the complexes which have lower pK_a (higher K_a) than of 2+. Among the same metal cation charge, the larger the metal cation size, the larger pK_a (smaller K_a). Larger charge and smaller size of the metal cations strengthen coordination bonding of M-OH_2. In this case, hydrogen bonding of positive dipole of H atom in H_2O ligand and negative dipole of O atom in H_2O solvent also supports the deprotonation.

In Lewis concept, no Lewis acidity constant and Lewis basicity constant are recognized like in Bronsted-Lowry concept because the transferred electron pair amount can't be determined directly. Alternatively, Lewis basicity is determined indirectly by formation reaction of Lewis adduct from L base and Lewis acid [8]:

$$\underset{\text{Lewis base}}{B} + \underset{\text{Lewis acid}}{A} \rightleftharpoons \underset{\text{Lewis adduct}}{BA} \qquad K_{BA} = \frac{[BA]}{[B]\,[A]}$$

Lewis basicity can be determined based on K_{BA} value. I_2 is the good Lewis acid to determine Lewis basicity due to soluble in different solvents. For example, the K_{BA} values in Table 3 showed the same Lewis basicity ranking in 2 different solvents by using I_2 as the Lewis acid in sequence of Lewis basicity: $(C_6H_5)_3P{=}O < (C_6H_5)_3P{=}S < (C_6H_5)_3P{=}Se$. There is decreasing

of electronegativity (O > S > Se) which causes the Lewis base softer. The soft acid I_2 makes stronger bonding with softer base.

Table 3: Data of log K_{BA} for using I_2 as Lewis acid and 2 different solvents at 25°C

No.	Lewis base	Lewis acid	Log K_{BA} in CCl_4	Log K_{BA} in $CHCl_3$
1.	$(C_6H_5)_3P=O$	I_2	1.38	0.89
2.	$(C_6H_5)_3P=S$	I_2	2.26	2.13
3.	$(C_6H_5)_3P=Se$	I_2	3.48	3.65

Source: **[8]**

Entalphy reaction is another indicator to determine Lewis basicity in formation of the Lewis adducts. Affinity of BF_3 towards various bases was measured in dichloromethane solution [Table 4]. Increasing of BF_3 affinities indicate stronger coordinate covalent bonding and increasing of Lewis basicity towards BF_3. The affinity is defined as magnitude of the enthalpy change of adduct formation in this reaction [8]:

$$BF_3 + \text{L base} \xrightarrow{CH_2Cl_2} \text{L base - } BF_3 \qquad -\Delta H^o = BF_3 \text{ affinity of Lewis basicity}$$

Table 4: Data of BF_3 affinities for different Lewis bases in CH_2Cl_2 at 25°C

No.	Lewis bases	BF_3 affinities (kJ/mol)
1.	2-trifluoromethylpyridine	82.46
2.	2-methylpyridine	123.44
3.	Pyridine	128.08
4.	3-methylpyridine	130.93
5.	4-dimethylaminopyridine	151.55

Source: **[8]**

Based on BF_3 affinities, Table 4 shows increasing of Lewis basicity from no 1 to no 5 due to different substituent type or location on pyridine structure (Figure 11). On the ortho position, substituent of trifluoromethyl reduced Lewis acidity to the lowest value. F atom has the highest electronegativity in periodic table, thus presence of $-CF_3$ as substituent will reduce electron density on pyridine structure. This condition makes N atom more difficult to donate the lone pair to the B atom of BF_3. Methyl is the electron pushing group which can increase electron density on pyridine structure, but it also reduced Lewis basicity of pyridine. It is probably caused by ortho position which is not effective to increase electron density and steric effect of methyl toward BF_3 due to near by N atom of pyridine. Methyl substituent on meta position enlarged the Lewis basicity of pyridine due to effective position to increase electron density and lack of steric effect. However, Substance of 4-dimethylaminopyridine has the highest Lewis basicity due to para position which is effective to improve electron density, low steric effect, and more methyl as the electron pusher group.

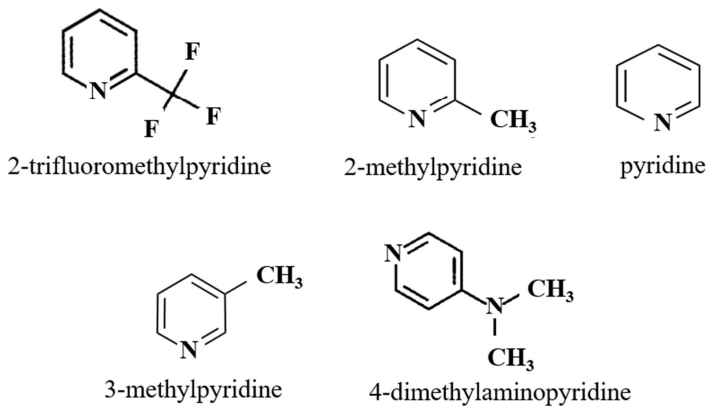

Fig. 11. Chemical structures of pyridine and its different type and location of substituents [44,-46].

In the metal complexes, Lewis acidity and Lewis basicity of the metal cations and the ligands can be predicted from stability constant or formation constant of the metal complexes for various metal cations (one metal cation type) or various ligands (one ligand type). However, this stability constant is affected by acid/base characteristics named hard acid/base and soft acid/base. Interactions of hard-hard or soft-soft species create more favourable reactions than of the hard-soft ones. Hard and soft acid/base are based on molecule or ion polarizability. Polarizability is a distortion degree of a molecule or an ion by their interactions. Electrons in the polarizable molecules will be attracted or repelled by other

molecule charges and form slightly the polar species which interact with the other molecules. Hard acids/bases are relatively small, compact, and nonpolarizable, while the soft acids/bases are larger and more polarizable. The hard acids include the metal cations which have the large positive charges (≥ 3+) or d electrons with relatively unavailable for p bonding. Soft acids are those which have d electrons or orbitals available for p bonding such as neutral atoms , 1+ cations, and heavier 2+ cations [8].

Although no acidity constant and basicity constant in Lewis concept like in Bronsted-Lowry concept, hardness and softness of acid/base are recognized in Lewis acid-base theory as Hard Soft Acid Base Concept (HSAB) and can be calculated quantitatively. Absolute hardness (η) is calculated as half of ionization energy (I) and electron affinity difference (A), both in eV. Softness (σ) is defined as the hardness inverse.

$$\eta = \frac{I-A}{2} \qquad \sigma = \frac{1}{\eta}$$

Data of stability constants for some different metal complexes are listed in Table 5 with the same Lewis hard base (NH_3).. The higher the stability constant the stronger Lewis acidity. The larger coordination number the larger metal cation size [2], thus the same coordination numbers are needed for comparison of different metals. Among the same metal cation charges of 3+ and coordination number of 6, the stability constant of the complex which were formed by the hard acid-hard base (Co^{3+} -NH_3) is much higher than by borderline acid- hard base. For among borderline metal cations and among soft metal cations, the smaller size of metal cations the larger K value. HSAB theory is good to compare the same metal, such as Cu^+ VS Cu^{2+} or Co^{2+} VS Co^{3+}, but for different metal cations, it may be not always applicable because there is other influencing factor which must be considered. For example, Ag(I) – NH_3 complex has much lower K than Cu(II) – NH_3 complex because Ag(I) is soft and Cu(II) is borderline acid while NH3 is hard. In other side, Cu(I) – NH_3 complex has much higher K than Co(II) – NH_3 complex although Cu(I) is soft and Co(II) is borderline. Thus, HSAB concept is conditional.

Table 5: Stability constants of ammonia complexes with different metal cations

No.	Lewis acid	Hardness acid (η)	M^{n+} radii (pm)**	Lewis base	Hardness base (η)	Coordination number (CN)	Lewis adduct	Stability constant (K)
1.	Ag^+	Soft	81	NH_3	hard	2	$[Ag(NH_3)_2]^+$	1.70 X 10^7
	Cu^+	Soft	60	NH_3	hard	2	$[Cu(NH_3)_2]^+$	3.80 X 10^{10}
2.	Zn^{2+}	Borderline	74	NH_3	hard	4	$[Zn(NH_3)_4]^{2+}$	3.98 X 10^9
	Cu^{2+}	Borderline*	71	NH_3	hard	4	$[Cu(NH_3)_4]^{2+}$	4.80 X 10^{12}
3.	Co^{2+}	Borderline	89	NH_3	hard	6	$[Co(NH_3)_6]^{2+}$	7.70 X 10^4
	Ni^{2+}	Borderline	83	NH_3	hard	6	$[Ni(NH_3)_6]^{2+}$	1.26 X 10^9
	Co^{3+}	Hard	69	NH_3	hard	6	$[Co(NH_3)_6]^{3+}$	5.00 X 10^{33}

Sumber: **[8,47]; *[48], **[9]**

Table 6 gives examples for the complexes with same Lewis acid but different Lewis bases. In Table 6, the stability constant of the Au(I) complexe anions increased by Lewis base sequence of $F^- < Cl^- < I^-$ which indicate sequence of Lewis basicity toward Au(I) metal cation. Au(I) is soft acid which creates strong coordination bond with soft ligand. Softness of ligand increses from F^- to I^- due to increasing of anion size which makes the anion easier to donate lone pair to the Au(I). This sequence is match with HSAB concept.

Table 6: Stability constants of complexes with same metal cations and different ligand

No.	Lewis acid	Acid hardness (η)	Lewis base	Base hardness (η)	Lewis adduct	K
1.	Au(I)	5.6 (Soft)	Cl^-	4.70 (Hard)	$[Au(Cl)_2]^-$	3.9 X 10^9
2.	Au(I)	5.6 (Soft)	Br^-	4.24 (Borderline)	$[Au(Br)_2]^-$	2.5 X 10^{12}
3.	Au(I)	5.6 (Soft)	I^-	3.70 (Soft)	$[Au(I)_2]^-$	1.0 X 10^{19}

Sumber: **[9, 49]**

Lewis basicity can be also measured as a sequence from K_{sp} (constant of solubility product) values with reaction as follows [1]:

$$AgX\ (s) + n\ H_2O\ (l) \rightleftharpoons Ag^+\ (aq) + Cl^-\ (aq) \qquad K_{sp} = [Ag^+]\ [X^-]$$

The lower K_{sp} the lower solubility of AgX , the stronger bonding of Ag(I) and X^-, the higher Lewis basicity of halogen ions toward Ag(I) cation. The soft acid Ag(I) prefer to create strong bonding with soft base with more covalent bonding characteristics. Data in Table 7 shows that K_{sp} values of AgX decreases in sequence of $F^- > Cl^- > Br^- > I^-$ due to decreasing of Lewis base hardness. This decreasing ones are caused by larger size of halogen anions which cause them easier to donate the lone pair to Lewis acid Ag(I). Beside that, although the same soft acid, Ag(I) has smaller size for AgI

and AgBr due to tetrahedral clusters in their unit cells, while larger size for Ag(I) in AgF and AgCl due to octahedral clusters (Figure 12).

Table 7: K_{sp} values of Lewis adducts with Lewis acid of Ag(I)

No.	Lewis acid	Acid hardness (η)	M^{n+} radii (pm) **	Lewis base	Base hardness (η)	X^- (pm)	Lewis adduct	K_{sp}
1.	Ag (I)	Soft	129	F^-	7.01 (Hard)	117	AgF	205
	Ag(I)	Soft	129	Cl^-	4.70 (Hard)	167	AgCl	1.8×10^{-10}
	Ag(I)	Soft	114	Br^-	4.24 (Borderline)	182	AgBr	5.2×10^{-13}
	Ag(I)	Soft	114	I^-	3.70 (Soft)	206	AgI	8.3×10^{-17}
2.	Mg^{2+}	Hard	86	F^-	7.01 (Hard)	117	*MgF_2	6.6×10^{-9}
	Ca^{2+}	Hard	114	F^-	7.01 (Hard)	117	*CaF_2	3.9×10^{-11}
	Sr^{2+}	Hard	132	F^-	7.01 (Hard)	117	*SrF_2	2.8×10^{-9}
	Ba^{2+}	Hard	149	F^-	7.01 (Hard)	117	*BaF_2	1.7×10^{-6}

Source: **[8], * [32], ** [9]**

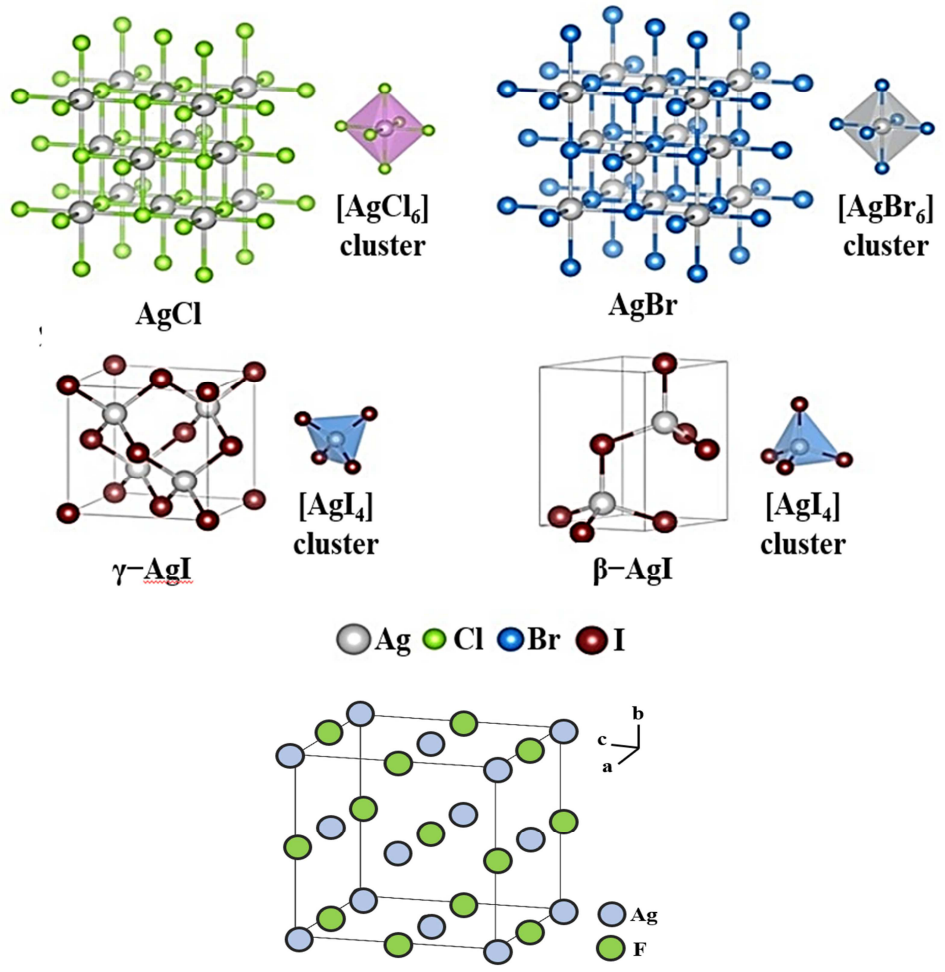

Fig. 12. Crystal structure of AgX [50,51].

Based on Table 7, HSAB concept is also applicable to explain K_{sp} of fluoride salts which are formed by some different metal cations of IIA group in periodic table. Although all metal cations and anion are same hard, their solubilities increase from CaF_2 to BaF_2; This is as consequency of larger metal cation size from Ca^{2+} to Ba^{2+} for same 8 coordination number (Figure 13) which make their bonding easier broken by H_2O solvent. However, there is anomaly about MgF_2 which has higher K_{sp} than CaF_2 although the Mg^{2+} cation size is smaller than Ca^{2+}. It is probably caused by lower coordination number of Mg^{2+} in its unit cell than Ca^{2+}. Every Mg^{2+} and Ca^{2+} cations are arrounded by $3F^-$ and $8F^-$ anions, respectively (Figure 13). Thus, H_2O

polar solvent molecules are easier to make interactions with Mg^{2+} than Ca^{2+} due to more repelled by fluoride anions to attack Ca^{2+}.

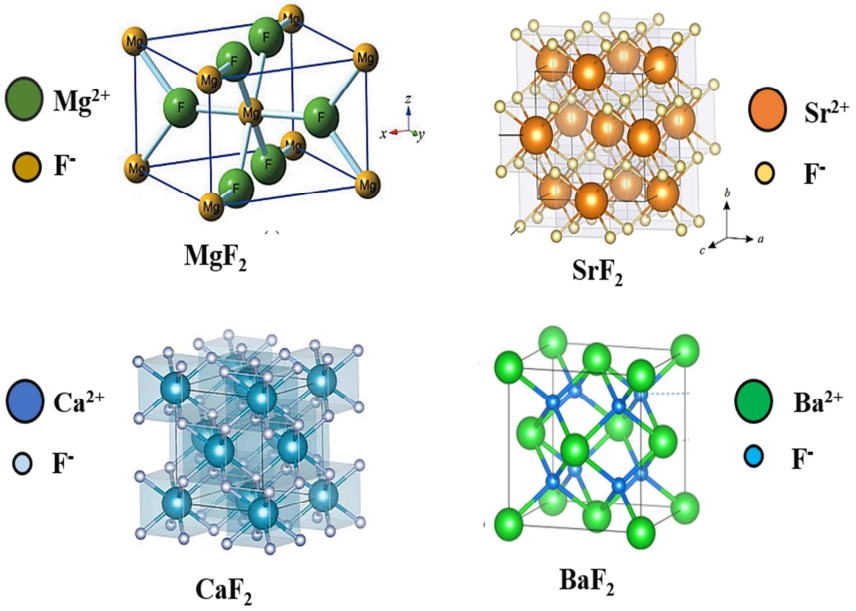

Fig.13. Crystal structures of MF_2 fluoride salts [52-54].

By development, Lewis acidity can be determined as 3 categories, including global Lewis acidity (gLA), effective Lewis acidity (eLA), and intrinsic Lewis acidity (iLA) as shown in Figure 14. Among those categories, the eLA uses spectroscopic methods to measure effect of Lewis acid on a probe molecule. The induced changes of physicochemical properties of a probe Lewis base are followed by instrumental measurements including IR/UV/Vis/ fluorescence/NMR spectroscopy [16].

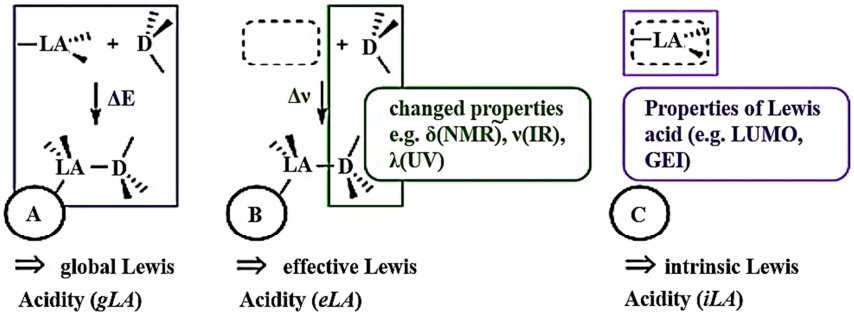

Fig. 14. Scaling methods of Lewis acidity : A) global, B) effective, C) intrinsic [16].

2. Comparisons in application

Acid-base concept can be used to explain the reactions in various applications. This section discussed some applications to understand which concepts is more applicable compared to the others.

Bronsted-Lowry concept can be applied for the reactions in rock mineral activation. For example, based on EDX analysis kaolinite mainly contains SiO_2 (53.57 %) and Al_2O_3 (43.54 %) with some chemical impurities such as Fe_2O_3 (1.08 %), K_2O (1.52%), Na_2O (0.078 %), CaO (0.085 %), MgO (0.094 %), and TiO_2 (0.073%) [55]. Sea sand consists of SiO_2 (53.16 %), Al_2O_3 (19.40 %), Fe_2O_3 (4.70 %), CaO (2.66 %), and MgO (2.08%), and Loss on ignition (18.0%) [56]. Activation with acid substances are usually needed to remove impurities of the minerals before synthesis. These impurities are the metal oxides which are not part of tectosilicate framework in sea sand or both silica and alumina layers in kaolinite. For example, the natural kaoline was activated by H_2SO_4 solution (0.1M) before grafting and intercalation treatment [57-58] or substitution/dopping with transition metal cations [58-59]. The natural sea sand was activated by HCl solution (0.1M)

before modified with sodium dodecyl benzene sulfonate (SDBS) surfactant [60]. HCl and H_2SO_4 are Arrhenius acids due to production of dissolve proton in their solutions. They are also Bronsted acids due to electron transfer reaction toward H_2O (Bronsted base) to form H_3O^+. However, the activation reactions are impossible to explain by using Arrhenius theory, it needs Bronsted-Lowry acid base reactions to remove M_2O (M = K^+, Na^+), MO (M = Mg^{2+}, Ca^{2+}), Fe_2O_3. As explained in section 2 that all Bronsted bases are Lewis base and all Bronsted acids contains Lewis acid (H^+), therefore the rock mineral activations with acids can be also explained using Lewis acid-base theory.

$$\underset{\text{Bronsted/Lewis base}}{M_2O\,(s)} + \underset{\text{Bronsted acid}}{H_3O^+(aq)} \longrightarrow 2M^+(aq) + \underset{\text{conjugate acid}}{OH^-(aq)} + \underset{\text{conjugate base}}{H_2O\,(l)}$$

$$\underset{\text{Bronsted/Lewis base}}{MO\,(s)} + \underset{\text{Bronsted acid}}{H_3O^+(aq)} \longrightarrow M^{2+}(aq) + \underset{\text{conjugate acid}}{OH^-(aq)} + \underset{\text{conjugate base}}{H_2O\,(l)}$$

$$\underset{\text{Bronsted/Lewis base}}{Fe_2O_3\,(s)} + \underset{\text{Bronsted acid}}{3H_3O^+(aq)} \longrightarrow 2Fe^{3+}(aq) + \underset{\text{conjugate acid}}{3OH^-(aq)} + \underset{\text{conjugate base}}{3H_2O\,(l)}$$

TiO_2 needs heating for dissolving process because TiO_2 is soluble in hot H_2SO_4 and HCl solution [61, 44] with this acid-base reaction:

$$\underset{\text{Bronsted/Lewis base}}{TiO_2\,(s)} + \underset{\text{Bronsted acid}}{2H_3O^+(aq)} \xrightarrow{\Delta} Ti^{4+}(aq) + \underset{\text{conjugate acid}}{2OH^-(aq)} + \underset{\text{conjugate base}}{2H_2O\,(l)}$$

In dry synthesis of $ZnFe_2O_4$/CNS, $MnFe_2O_4$ and $ZnCr_2O_4$//CNS composites, the ZnO/CNS composite was reacted with KOH and salt chlorides by calcination. The ZnO/CNS was prepared from biomass and $ZnCl_2$ hydrothermally and with microwave sequently [62] or with dry microwave and product dispersion process in water solvent mechanically [63]. The spinel dry synthesis is solid state synthesis method which involve diffusion of ions in solid phase thermally to form the product [64]. The spinel formation in the calcination processes are Lewis acid base reaction involving Lewis acids (Zn^{2+}, Mn^{2+}, Cr^{3+}, Fe^{3+}) using sources of metal chloride salts and Lewis bases (O^{2-}) from sources of KOH. The reactions are not Arrhenius or Bronsted – Lowry ones because they involved the lone pair transfer in solid phase. The chemical reactions are as follows:

$$ZnO/CNS\,(s) + 2KOH\,(s) + ZnCl_2\,(s) + 2CrCl_3.6H_2O\,(s) \longrightarrow ZnCr_2O_4/CNS\,(s) + 2KCl\,(s) + ZnO\,(s) + 6HCl\,(g) + CO_2\,(g) + 6H_2O\,(g)$$

$$ZnO/CNS\,(s) + 2KOH\,(s) + MnCl_2\,(s) + 2FeCl_3\,(s) \longrightarrow MnFe_2O_4/CNS\,(s) + 2KCl\,(s) + ZnO\,(s) + 6HCl\,(g) + CO_2\,(g)$$

Existence of $ZnCr_2O_4$ and $ZnFe_2O_4$ spinels as Lewis adducts appear clearly in their same crystal structures (Figure 15). Both spinels are the normal structure with Zn^{2+} cations occupy tetrahedral sites arrounded by O^{2-} anions and Fe(III) or Cr(III) cations in octahedral sites arrounded by six O^{2-} anions. While for $MnFe_2O_4$, Mn^{2+} and Fe^{3+} in $MnFe_2O_4$ occupy both sites [62,63]. Those transition metal cations are Lewis acids and oxygen anions are Lewis bases.

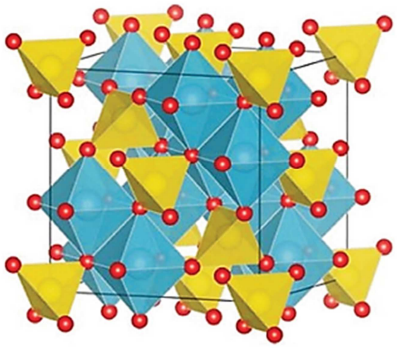

Fig.15. Crystal structures of $ZnFe_2O_4$ spinel [65].

Adsorption of metal cations by activated carbon or biochar can be explained by cation exchange reaction and complexation or ionic interactions (Figure 16). The cation exchange reactions can occur between proton or metal cation such as Na^+ with adsorbate metal cations. The surface complexation reaction can be occurred between oxy functional groups such as $-COO^-$ and $\equiv SiO^-$. The exchange reactions can be Bronsted-Lowry or Lewis acid base reactions. However, the carboneous materials which release the dissolved proton in adsorption through exchange reaction are Arrhenius acids.

$$\underset{\text{Lewis base/Bronsted acid}}{-COOH\,(s)} + \underset{\text{Lewis acid}}{M^+(aq)} + \underset{\text{Bronsted base}}{H_2O\,(l)} \longrightarrow \underset{\text{Lewis adduct}}{COO^-M^+\,(s)} + \underset{\text{conjugate acid}}{H_3O^+(aq)}$$

$$\underset{\text{Lewis base/acid}}{2\text{-}COO^{-}Na^{+}\,(s)} + \underset{\text{Lewis acid}}{Pb^{2+}\,(aq)} \longrightarrow \underset{\text{Lewis adduct}}{COO^{-}M^{+}\,(s)} + \underset{\text{Lewis adduct}}{Na^{+}\,(aq)}$$

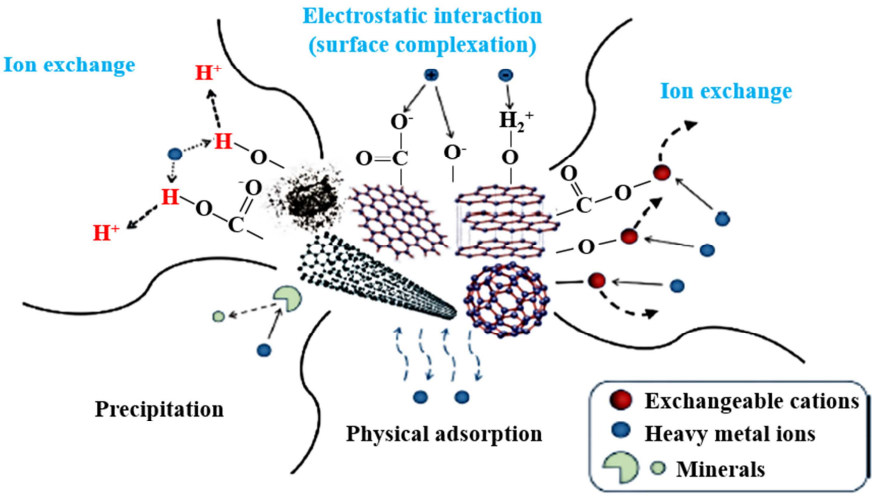

Fig. 16. Adsorption mechanism of heavy metal cations by carboneous materials [66].

In the chemical industry, cement is made by calcining the mixed ground limestone ($CaCO_3$) and aluminosilicates sources (clay, shale, sand) to1500°C in a rotary kiln [30]. In calcination process, limestone decomposes to lime (CaO) which reacts with the silicates to form molten calcium silicates such as Ca_2SiO_4, Ca_3SiO_5, and $Ca_3Al_2O_6$. In this reaction CaO is Lewis base whereas SiO_2 and Al_2O_3 are Lewis acid [67]. These reactions are impossible to explain using both Bronsted-Lowry concept and Arrhenius concept due to no proton transfer and no water solvent, respectively.

$$\underset{\text{Lewis base}}{2CaO\,(s)} + \underset{\text{Lewis acid}}{SiO_2\,(s)} \xrightarrow{\Delta} \underset{\text{Lewis adduct}}{Ca_2SiO_4\,(s)}$$

$$\underset{\text{Lewis base}}{3CaO\,(s)} + \underset{\text{Lewis acid}}{SiO_2\,(s)} \xrightarrow{\Delta} \underset{\text{Lewis adduct}}{Ca_3SiO_5\,(s)}$$

$$\underset{\text{Lewis base}}{3CaO\,(s)} + \underset{\text{Lewis acid}}{Al_2O_3\,(s)} \xrightarrow{\Delta} \underset{\text{Lewis adduct}}{Ca_3Al_2O_6(s)}$$

The ordinary Portland cement consists of four major inorganic phases, including 50−70% tricalcium silicate, $3CaO{\cdot}SiO_2$ or Ca_3SiO_5 (C_3S), 10−20% dicalcium silicate, $2CaO{\cdot}SiO_2$ or Ca_2SiO_4 (C_2S), 5−10% tricalcium aluminate, $3CaO{\cdot}Al_2O_3$ or $Ca_3Al_2O_6$ (C_3A), and 5−15% tetracalcium alumino ferrite, $4CaO{\cdot}Al_2O_3{\cdot}Fe_2O_3$($C_4AF$) [67]. The some cement components such as Lewis bases (CaO, MgO), and Lewis adducts (C_3A, C_4AF, C_3S, C_2S) in across section of the cement grain is shown in Figure 17 with crystal structures in Figure 18. Based on Figure 18, there are different chemical silicate structures of Ca_2SiO_4 and Ca_3SiO_5. The SiO_4^{2-} anions are separated as monomers for the former while $[O_3Si\text{-}O\text{-}SiO_3]^{6-}$ anions as dimers for the later and negative charges are neutralized by Ca^{2+} cations. In $Ca_3Al_2O_6$, Every Al^{3+} is arrounded by six O^{2-} anions, four of them make brigdes of Al-O-Al and two of them are neutralized by Ca^{2+} cations. In Figure 18 we can see that Lewis acids (Ca^{2+}) make the ionic bond with Lewis bases (AlO^- and SiO^- sites) of their each polyhedron structure. Both Bronsted-Lowry and Arrhenius theory are impossible to explain them due to no proton transfer nor no solvent reaction, respectively.

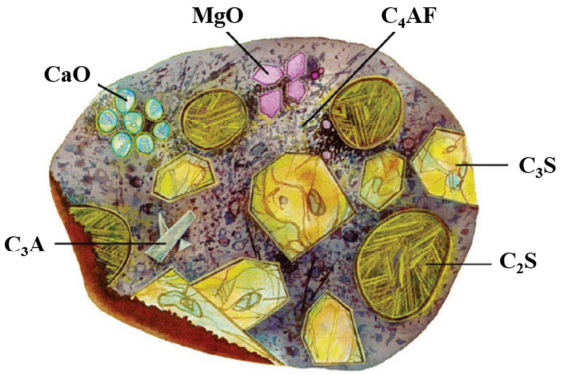

Fig. 17. Lewis bases and Lewis adducts in a cross section of a cement grain [68].

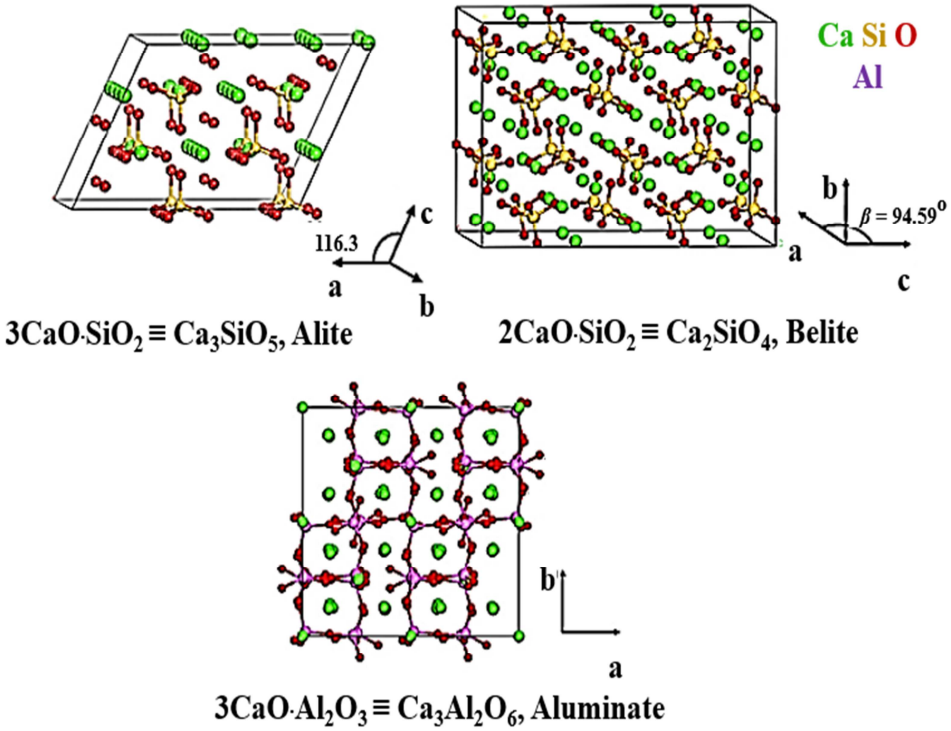

Fig. 18. Crystal structure unite cell of some cement components [67].

Another new type of acid-base cement, ferrous oxalate cement (FOC), is prepared at room temperature by chemical reactions of iron-rich copper slag (CS), oxalic acid/$H_2C_2O_4 \cdot 2H_2O$ (OA), borax/$Na_2B_4O_7 \cdot 10H_2O$ (B) and water to form paste. Borax is commonly used to retard cement acid base reaction. The cement which was resulted without borax had higher compressive strength than with borax. CS contains iron oxides and silica totally (81%). The cement formation reaction can be explained by decreasing of the dissolved oxalate and Fe(II) after cementation reactions to 24 h (Figure 19). There was increasing of pH from 1.5 to 5.2 after 24 h. At pH > 4.5 the oxalic acid species are $C_2O_4^{2-}$ (> 65%) and $HC_2O_4^-$ anions (> 35%). There were $C_2O_4^{2-}$, $HC_2O_4^-$, Fe(II), and H_2O in the paste, thus cementation reactions can be predicted as Lewis acid-base reactions as follows [24]:

$$\underset{\text{Lewis acid}}{Fe^{2+}(aq)} + \underset{\text{Lewis base}}{C_2O_4^{2-}(aq)} + \underset{\text{Lewis base}}{2H_2O(l)} \rightarrow \underset{\text{Lewis adduct}}{FeC_2O_4 \cdot 2H_2O(s)}$$

$$\underset{\text{Lewis acid}}{Fe^{2+}(aq)} + \underset{\text{Bronsted acid}}{HC_2O_4^-(aq)} + \underset{\text{Lewis/Bronsted base}}{3H_2O(l)} \rightarrow \underset{\text{Lewis adduct}}{FeC_2O_4 \cdot 2H_2O(s)} + \underset{\text{Conjugate acid}}{H_3O^+(aq)}$$

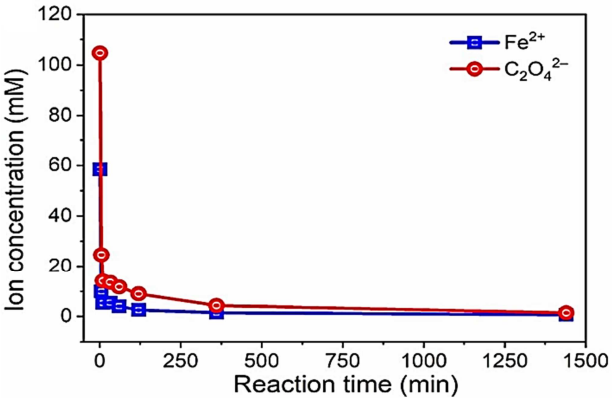

Fig. 19. Concentrations of Fe(II) and $C_2O_4^{2-}$ ions in cementation reaction times [24].

Figure 20 shows that every Lewis acid Fe(II) is arrounded by two Lewis base $C_2O_4^{2-}$ anions and two Lewis base H_2O molecules which form octahedral polyhedrons with Fe(II) centres. Thus, they are match with Lewis acid/base in the chemical equations. Although H_2O contains H atoms, the O atom of H_2O molecules which has the role as donor atom of Lewis base to make coordination bond with Fe(II) metal cations. Therefore, Bronsted-Lowry is not applicable to explain both reactions and crystal structure. Arrhenius is also not applicable due to the solid state phase.

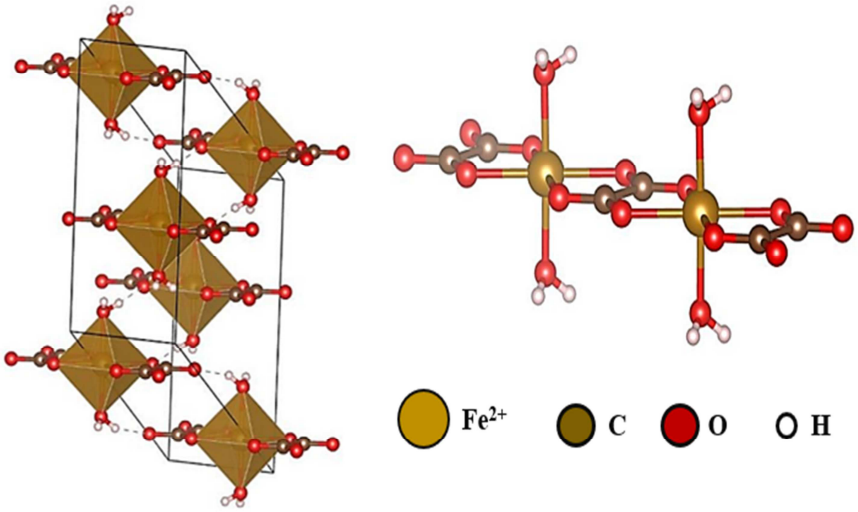

Fig. 20. Crystal structure of $FeC_2O_4.2H_2O$ [69].

Degradation of cementation materials in various acid solutions can be explained by using Bronsted-Lowry method. For example, the cement samples were immersed in four different acid solutions (acetic acid, citric acid, tartaric, oxalic acid) with same concentration of acid (0.28M) at pH 0.085 for oxalic acid but at pH 4 for acetic acid, tartaric acid, and citric acid by addition of NaOH solution. The cement contained CaO (64.87%), SiO_2 (21.19%), Al_2O_3 (3.94%), Fe_2O_3 (2.36%), MgO (2.37%) and minor components (TiO_2, Na_2O, K_2O, MnO) for each less than 0.3%. The immersed cements (for 1 year) in those each solutions showed the different mass losses (Figure 21). Sequence of their mass losses by using citric acid > tartaric acid > acetic acid > oxalic acid. Reason of mass loss was considered from solubility of CaO in acid solution and solubility of salts which were formed by Ca^{2+} with anions which were produced by deprotonation reaction of acids [70]. This is due to its highest and much higher content in the cement than some other oxides such as Al_2O_3, Fe_2O_3 , MgO which are also soluble in acid solution.

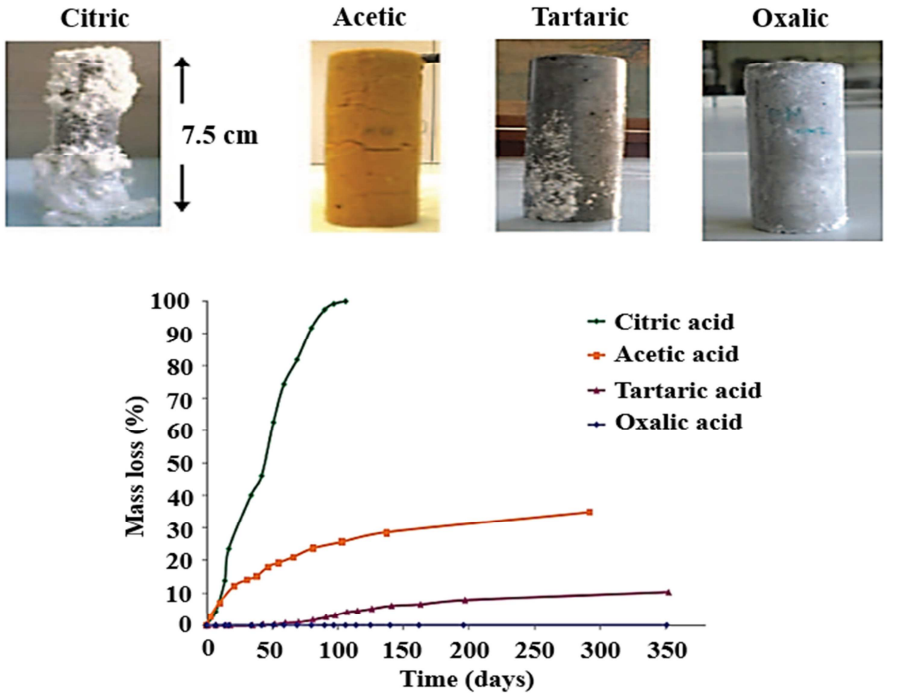

Fig. 21. Degradation of cement after the specimens immersion for 1 month in the various acids [70].

In cement, Ca^{2+} cations make ionic bonding with silicates and aluminate sites. When the cement was immersed in the acid solution, ion exchanges occurred between Ca2+ and H+ without destroy silicate structure as follows:

$$(\equiv SiO^-)_2\ Ca^{2+}\ (s) + 2H_3O^+\ (aq) \longrightarrow 2\equiv Si\text{-}OH\ (s) + Ca^{2+}\ (aq) + 2H_2O(l)$$

Aluminate structure in cement can be destroyed by H^+ due to its amphoteric characteristics. However, the discussion is focused on Ca^{2+} dissolution by acid solution due to its much higher content in the cement. Based on K_a values and calculation of anion and acid substance concentration ratios (Table 8) and Figure 22, the anions in the solutions at pH 4 are CH_3COO^-, $C_4H_5O_6^-$, $C_4H_4O_6^{2-}$, $C_6H_7O_7^-$, $C_6H_6O_7^{2-}$, and $C_6H_5O_7^{3-}$. Among those anions, both $C_4H_4O_6^{2-}$ anion of tartaric acid and $C_6H_5O_7^{3-}$ anion of citric acid can form precipitation with Ca^{2+} cations as $CaC_4H_4O_6$ and $Ca_3(C_6H_5O_7)_2$ on surface of the cement. Based on K_{sp} values (Table 9), precipitation of calcium citrate tetrahydrate is easier than calcium tartrate. However, for the same concentration of acid (0.28M), the $C_6H_5O_7^{3-}$ concentration in the citric acid solution is much lower than $C_4H_4O_6^{2-}$ in tartaric acid solution (Table 8). Thus, mass loss of cement in the citric acid solution much larger than in tartaric acid solution.

Table 8: Formula and pK_a of various Bronsted acid

No.	Bronsted acid	Formula	pK_a	K_a	pH	Bronsted bases in solution
1.	Oxalic acid	$H_2C_2O_4$	$pK_{a1} = 1.25$ $pK_{a2} = 4.27$	5.62×10^{-2} 5.37×10^{-5}	0.085	$[HA^-]/[H_2A] = 0.068$ $[A^{2-}]/[H_2A] = 4.47 \times 10^{-6}$
2.	Acetic acid	CH_3COOH	$pK_a = 4.76$	1.74×10^{-5}	4	$[A^-]/[HA] = 0.174$
3,	Tartaric acid	$C_4H_6O_6$	$pK_{a1} = 3.04$ $pK_{a2} = 4.37$	9.12×10^{-4} 4.27×10^{-5}	4	$[HA^-]/[H_2A] = 9.12$ $[A^{2-}]/[H_2A] = 3.89$
4.	Citric acid	$C_6H_8O_7$	$pK_{a1} = 3.13$ $pK_{a2} = 4.76$ $pK_{a3} = 6.4$	7.41×10^{-4} 1.74×10^{-5} 3.98×10^{-7}	4	$[H_2A^-]/[H_3A] = 7.41$ $[HA^{2-}]/[H_3A] = 1.29$ $[A^{3-}]/[H_3A] = 5.13 \times 10^{-3}$

Source: **[70] ; *[37]**

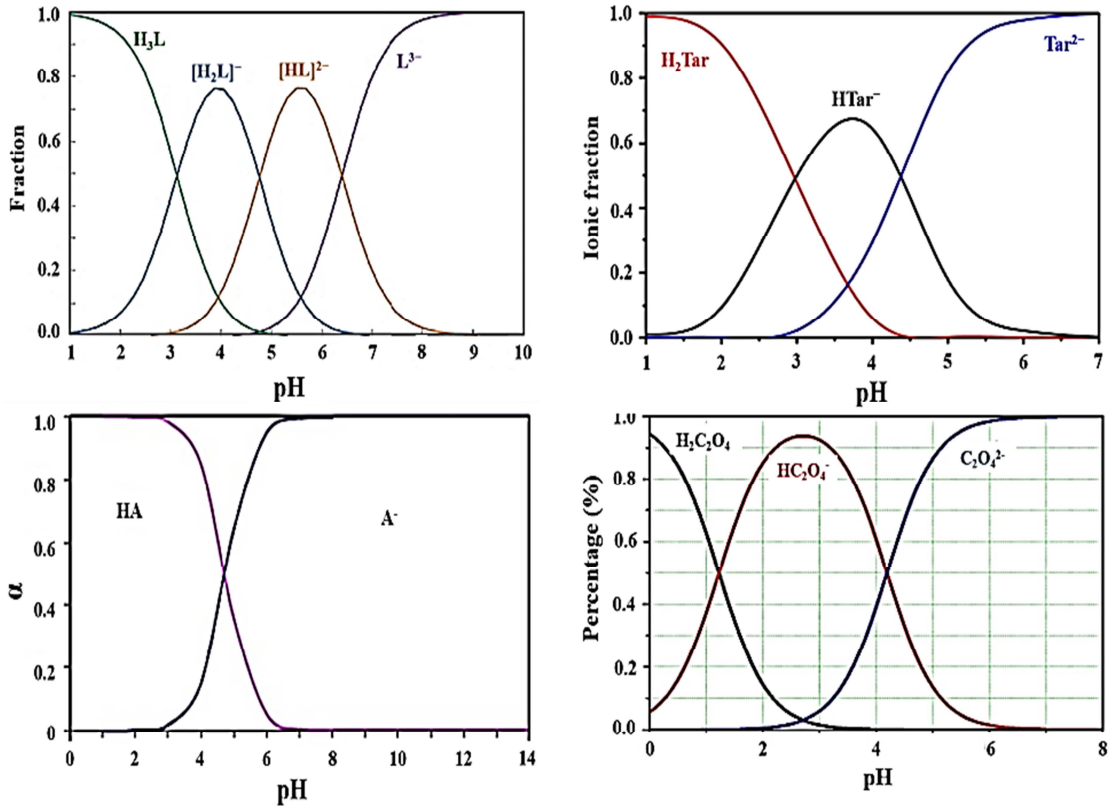

Fig. 22. Species of citric acid, tartaric acid, and acetic acid at various pH [24, 71.72, 73].

Acetic acid provided CH_3COO^- in the solution. This anion can't precipitate Ca^{2+}, while tartaric acid solution provided $C_4H_4O_6^{2-}$ anion which can precipitate Ca^{2+} on the cement surface, therefore mass loss of cement in acetic acid is larger than in tartaric acid. The CH_3COO^- anions in the acetic acid solution attracted the Ca^{2+} cations to dissolve them in the cation exchange reaction as follows:

$$(\equiv SiO^-)_2\ Ca^{2+}\ (s) + \underset{\text{Bronsted acid}}{2H_3O^+\ (aq)} + \underset{\text{Lewis base}}{4CH_3COO^-\ (aq)} \longrightarrow \underset{\text{Bronsted adduct}}{2\equiv Si\text{-}OH\ (s)} + \underset{\text{Lewis adduct}}{2Ca(CH_3COO)_2\ (aq)} + H_2O\ (l)$$

Although acetic anion can't precipitate $Ca2^+$ due to solubility of $Ca(CH_3COO)_2$ and citrate anion in the acid solution can do it (Table 9) on the cement surface, the mass loss in the citric acid was much higher than in the acetic acid. It is probably presence of $C_6H_7O_7^-$ and $C_6H_6O_7^{2-}$ anions in the solution which can't precipitate Ca^{2+} but can attract Ca^{2+} to dissolve it into the solution system as follows:

$$(\equiv SiO^-)_2\ Ca^{2+}\ (s) + \underset{\text{Bronsted acid}}{2H_3O^+\ (aq)} + \underset{\text{Lewis base}}{2C_6H_7O_7^-\ (aq)} \longrightarrow \underset{\text{Bronsted adduct}}{2\equiv Si\text{-}OH\ (s)} + \underset{\text{Lewis adduct}}{Ca(C_6H_7O_7)_2\ (aq)}$$

$$(\equiv SiO^-)_2\ Ca^{2+}\ (s) + \underset{\text{Bronsted acid}}{2H_3O^+\ (aq)} + \underset{\text{Lewis base}}{C_6H_6O_7^{2-}\ (aq)} \longrightarrow \underset{\text{Bronsted adduct}}{2\equiv Si\text{-}OH\ (s)} + \underset{\text{Lewis adduct}}{CaC_6H_6O_7\ (aq)}$$

Table 9: Data of solubility and K_{sp} of some acids

No.	Salt	Chemical formula	Solubility /K_{sp}	Reference
1.	Calcium citrate tetrahydrate	$Ca_3(C_6H_5O_7)_2 \cdot 4H_2O$	0.30807 g/100 mL (25°C) $7.6 \pm 0.5 \times 10^{-17}$	[74]
2.	Calcium acetate monohydrate	$Ca(CH_3COO)_2.H_2O$	34.7 g/100 mL (20 °C)	[70]
3.	Calcium tartrate tetrahydrate	$CaC_4H_4O_6 \cdot 4H_2O$	0.0266 g/100 mL (0 °C)	[70]
	Calcium tartrate	$CaC_4H_4O_6$	7.7×10^{-7} mol^2/L^2	[75]
4.	Calcium oxalate monohydrate	$CaC_2O_4 \cdot H_2O$	Insoluble 6.7×10^{-9} mol^2/L^2	[70] [76]

Acid base reactions occur in the body, for example in erythrocyte (Figure 23). Erythrocyte contains 68 –70% % H_2O in human [77]. The CO_2 gasses which are formed in tissue by metabolism diffuse into red blood cell (erythrocyte). About 5% of them remains as a gas and 90–95% is converted to H_2CO_3 by water enzymatically by the cytosolic enzyme carbonic anhydrase II [78]. The carbonic anhydrase enzyme (CA) can reduce this reaction time from several minutes to second [79]. H_2CO_3 (Bronsted acid) makes further reaction with H_2O (Bronsted base) to form H_3O^+ and HCO_3^-. Therefore, CO_2 in erythrocyte is Arrhenius acid because it produces H^+ by water presence and decrease the blood pH. The oxyhemoglobin (HbO_2) acts as Bronsted base and accept the released H^+ to form the protonated deoxyhemoglobin (HHb) by releasing O_2 into the tissue cell [78]. Thus, H^+ production by H_2CO_3 does not change pH. The increased bicarbonate ions in the erythrocyte migrate into the plasma [80]. The HCO_3^- anion leaves these cells towards the plasma by exchanging with chloride. Erythrocytes with the protonated deoxyhemoglobin (HHb) formed in the tissue capillaries travel to the lungs. The uptake of oxygen gas transforms the protonated deoxyhemoglobin (HHb) into oxyhemoglobin (HbO_2) by releasing proton. This proton combines again with HCO_3^- to form H_2CO_3 by carbonic anhydrase II, generating water liquid and CO_2 gas [78]. All those reactions in the blood cell such as deptotonation of H_2CO_3 and HHb and protonation of Hb and bicarbonate ions are Bronsted-Lowry acid-base reactions due to the proton transfers. Both H_2CO_3 and HHb are Arrhenius acids due to formation of the dissolved proton in the water. Lewis acid base reaction took the role in reaction of CO_2 and H_2O to form H_2CO_3.

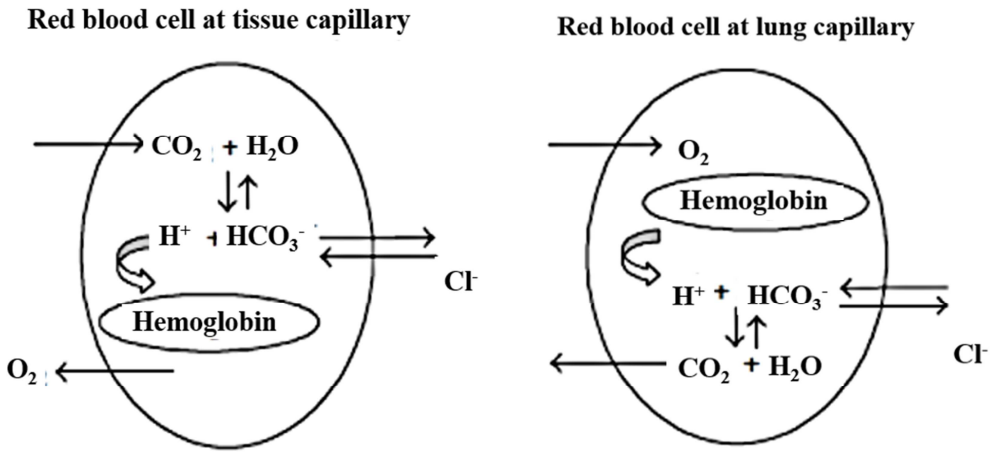

Fig. 23. Acid base reactions in erythrocyte of tissue and lung caplillaries [**78**].

Lewis acid base concept is useful to explain the reactions in organometal synthesis than Bronsted-Lowry and Arrhenius theories especially for the reactions with no proton transfer and production of dissolved proton or hydroxide, respectively. For example, in synthesis of boron substance which contains telurrium metal cation, Lewis acid-base theory can be used to explain chemical reaction in Figure 24.

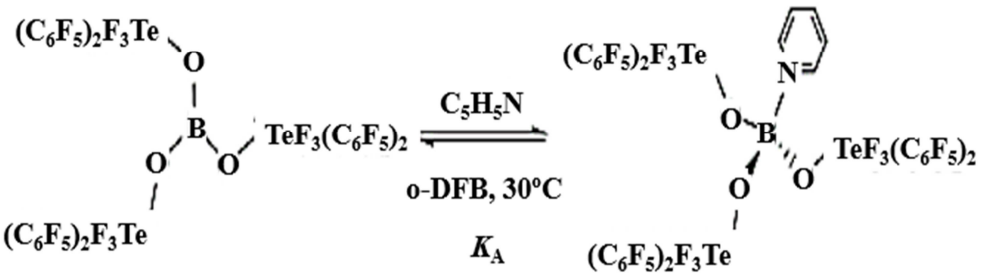

Fig. 24. Acid-base reaction of $B[OTeF_3(C_6F_5)_2]_3$ and C_5H_5N [81].

One of reasons for Lewis acid base reaction is to complete a molecule octet of valence electrons by accepting an electron pair [30]. Figure 20 shows that Lewis acid of $B[OTeF_3(C_6F_5)_2]_3$ can make reaction with Lewis base of C_5H_5N because chemically B atom can accept an electron pair from N donor atom of the C_5H_5N to complete its octet. In this reaction there is changing of hybridization from sp^2 (triangular molecular shape) to sp^3 (tetrahedral). Physically, this reaction can be performed due to its stability in tetrahedral shape toward repulsion among the substituents. Boron atom is Lewis acid and pyridine (C_5H_5N) is Lewis base. The reaction can't be explained using Bronsted-Lowry or Arrhenius due to lone pair transfer.

Another example of Lewis acid-base reaction in organotelurrium synthesis is ligand substitution reaction of Au(III) complexe compound. In this reaction, the F- ligand was substituted with $[OTeF_3(C_6F_5)_2]^-$ in Figure 25. In Figure 25 no addition of $[Au(CF_3)_4]^-$ Lewis base toward B atom Lewis acid in $B[OTeF_3(C_6F_5)_2]_3$ to complete its octet, probably due to its big size to minimize its repusion with other substituents. Alternatively, all $OTeF_3(C_6F_5)_2$ substituens are substituted by smaller F^- ion to form the smaller molecule (BF_3) by sustaining its trigonal planar. Another reason, the Au(III) hard acid makes the favourable interaction with hard base O atom of $[OTeF_3(C_6F_5)_2]^-$ ion. The organometallic reaction is impossibly explained using Bronsted – Lowry and Arrhenius theories due to no proton transfer and no water solvent, respectively.

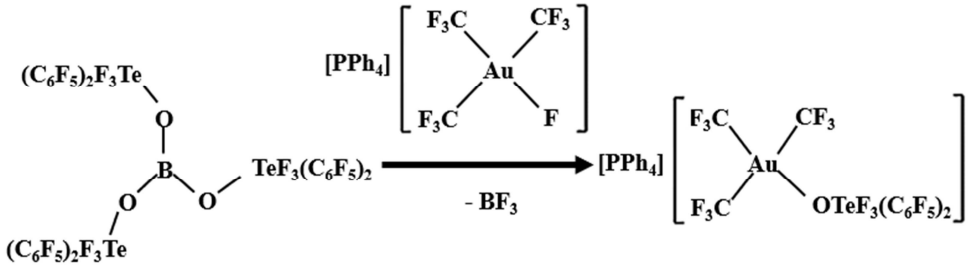

Fig. 25. Acid-base reaction of $B[OTeF_3(C_6F_5)_2]_3$ and $[PPh_4][Au(CF_3)_4]$ [81].

An advanced material such as Hb–PVP micro and nanofiber composite was prepared from haeomoglobin (Hb) and PVP (polyvinylpyrrolidone) using 2,2,2-Trifuoroethanol (TFE) 99% as the solvent. The material was synthesized for carbon monoxide capture. The codes of products include Hb-O (Hb-TFE) and Hb/PVP-X (Hb-PVP-TFE with x wt.% PVP) [19]. Characterization by FTIR spectrometry was used to identify presence of PVP based on new bands from PVP and wavenumber swift related to chemical interaction of Lewis acid (Fe^{2+} of Hb) and Lewis base (N or O atoms of PVP). Chemical structures of Hb and PVP also FTIR spectra of Hb and Hb-PVP composite are presented in Figure 26. In Figure 22 the additional C-N vibration appeared significantly for the composite which contains 16% and 32% PVP. No significant band swift for the Fe^{2+}- N vibration at about 500 cm^{-1}. It indicates that the chemical interactions of Lewis acid Fe^{2+} in Hb and Lewis bases N or O in PVP are more about molecular attraction force than about coordination bonding. This interaction is impossible to be explained using both Arrhenius and Bronsted-Lowry concepts due to solid phase and no proton transfer, respectively.

Effects of different Lewis acids (metal cations) on thermal stability of the metal complexes were studied using the synthesized pyrimidine and H_2O as ligands and Cl^- anions as the counter ions. The thermal stability was identified using TGA (Tabel 10). Table 10 shows that the metal complexe formation increased the ligand thermal decomposition temperature range from 25-600 to 30-1000°C. Beside that, two different metal cations gave 2 different thermal decomposition range especially for the first step, including 30-250°C for usage of Co^{2+} cation but 30-400°C for usage of Ni^{2+} cation. Both metal cations are Lewis intermediate acids but Ni(II) has smaller size than Co(II) so that it makes shorter M-N bonding with pyrimidine ligand and shorter M-Cl with Cl^- ligand than Co(II) ion. The metal complexe cations in this research are Lewis adduct, resulted from reaction between Lewic acid (Co^{2+} or Ni^{2+}) and Lewis bases (H_2O and pyrimidine).

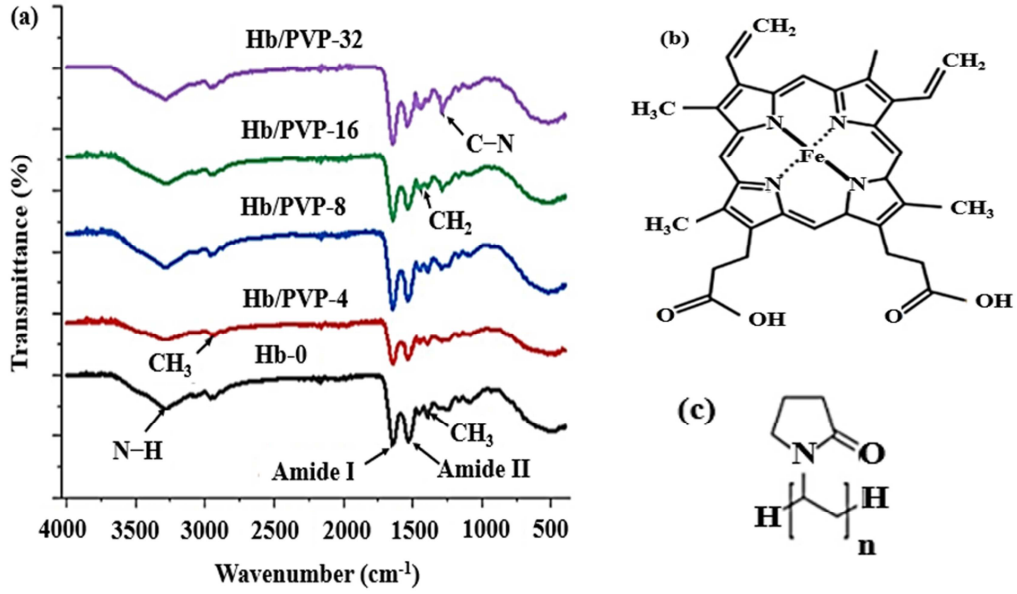

Fig. 26. a) FTIR spectra of pristine Hb-O and Hb/PVP composites at various % PVP, b) Hb structure, c) PVP structure [26].

Table 10: TGA data of pyrimidine and metal-pyrimidine complexes

Ligan and metal complexes	M^{2+} free radius (pm)*	d^n (M^{2+})*	H/I/S acid (M^{2+})*	M-O (pm)*	M-N (pm)*	M-Cl (pm)*	TGA data **	
							T (^oC)	Mass loss (%)
Pyrimidine (L)							25-250 250-600	37.73 62.25
$[Co(L)_2(H_2O)_2]Cl_2$	74	d^7	I	205	220	240	30-250 250–1000	38.68 52.17
$[Ni(L)_2(H_2O)_2]Cl_2$	69	d^8	I	205	210	235	30-400 400–1000	39.63 51.37

Source: ***[82]***; ****[83];*** Code: H = Hard; I = Intermediete, S = Soft

3. Conclusions

Study of comparison of three popular acid-base theories has been done. The comparisons include their concepts and applications. In the concept study, the superiority sequence is Lewis > Bronsted-Lowry > Arrhenius based on presence of solvent, solvent type, and protic/unprotic system, dissolve/undissolved products, and phase. Lewis reactions are not limited to the breaking of compound like Bronsted-Lowry. However, Bronsted acid or base strength can be determined quantitatively while Lewis acidity in sequence only.

The reactions in some application study can be explained by single or multi acid-base theories, including Bronsted-Lowry for cement degradation, Lewis for synthesis of cement thermally, organometal, metal complexe, and spinel/CNS, Arrhenius and Bronsted-Lowry for the acid activations of kaolinite or sea sand, Lewis and Bronsted-Lowry for room temperature cement formation, and those all three ones for metal ion adsorption by carboneous materials and metabolism reactions in erythrocyte.

4. Conflicts of interest

"There are no conflicts to declare".

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