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RESEARCH PAPER

Synthetic Study of Novel Deep Eutectic Solvent Derived from Chloride/Erythritol (Erythrine) with Iron (FeNP) Nanoparticles

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ABSTRACT

This research is aimed at developing an innovative type of deep eutectic solvent as environmentally safe alternatives to traditional acidic catalysts and ionic liquids that were marred by some environmental defects by the replacement of urea with a safer hydrogen bond donor, the erythritol was used to produce a novel deep eutectic solvent (Erythrine), which mixed with iron nanoparticles to create synergistic hybrid system with the aim of enhancing and developing catalytic efficiency in organic chemical reactions to accelerate the synthesis of Schiff bases, and then comparing the results with conventional chemical methods. Erythrine was prepared by mixing choline chloride salt with two moles erythritol and heating the mixture directly until it turns into a transparent liquid, where a significant reduction in the melting point occurs as a result of the hydrogen bonds between the two components. The iron nanoparticles (FeNP) which was prepared by precipitating iron salts using sodium hydroxide to obtain magnetic nanoparticles, these nanoparticles were mixed with Erythrine to ensure homogeneity. Two Schiff base compounds were synthesized using four different methods, including the use of a conventional method (A) of acetic acid catalyst which gave 80-81% for the two Schiff bases respectively, 88-90 when using the Erythrine catalyst in methods (B and C), and more than 90% as well as using FeNP in methods (D) respectively, in addition to the diagnosis and structural characterization of the novel Erythrine and the new Schiff bases were based on FT-IR spectroscopy, thermogravimetric and differential analysis. The morphological modification of the iron nanoparticles after the addition of Erythrine and the increase in their diameters (57.72-79.95nm) is attributed to the formation of a functional coating layer on their surfaces.

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INTRODUCTION

The first idea of deep eutectic solvents (DESs) was introduced in 2003 by Andrew P. Abbott and his team [1,2], via their groundbreaking work on the non-toxic cheap eutectic solvent (DESs) of choline chloride (ChCl), the hydrogen bond acceptor (HBA), Fig. 1. with urea as hydrogen bond doner (HBD), this mixture of low-melting liquid at room temperature, even though the individual components are solids [3].

Abbott team disclosed that a 1:2 molar ratio of choline chloride ChCl: urea "which was later commonly named Reline", formed a clear liquid with a melting point temperature range (from 17 °C - 22 °C) of "physicochemical" characters that are very close to those of ionic liquids (ILs). Therefore, DESs are suggested as sustainable alternatives to ionic liquids, retaining the advantages of their low vapor pressures, wide liquid-state temperature range, and broad electrochemical window, this melting point depression was named eutectic effect and this happened because of the strong hydrogen bonding between the ChCl salt the hydrogen bond acceptor (HBA), and urea, the hydrogen bond doner (HBD), which disrupts the crystal lattice [4].

A crystal lattice is the highly ordered, repeating three-dimensional arrangement of atoms, ions, or molecules in a crystalline solid that represents the fundamental structural framework which defines a crystal's physical and chemical properties [5,6].

Choline chloride (ChCl) is a quaternary ammonium salt with the chemical formula $[(CH_3)_3NCH_2CH_2OH]^+Cl^-$. It is crystal system: Orthorhombic with a space group: $Pna2_1$ and lattice parameters: (a ≈ 8.77 Å, b ≈ 10.66 Å and c ≈ 7.60 Å), the crystal lattice consists of choline cations $[(CH_3)_3NCH_2CH_2OH]^+$ and chloride anions (Cl^-) held together by ionic bonds between the anionic chlorine and cationic quaternary nitrogen, and hydrogen bonds (between the OH group of choline and Cl^- , The crystal lattice of choline chloride is shown in Fig. 2 as predicted by this paper, which was plotted using the Chemoffic

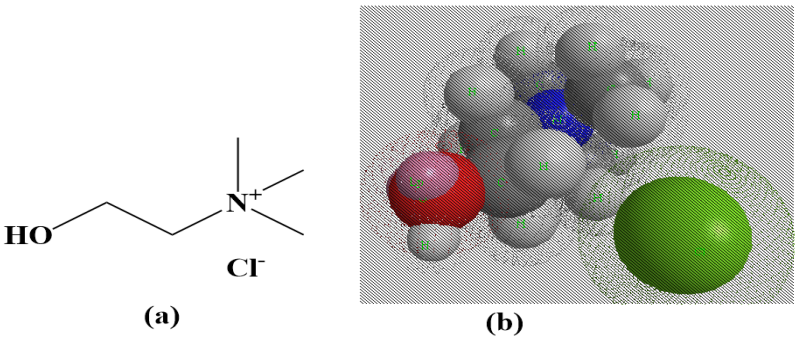


Fig. 1. Choline chloride structure.

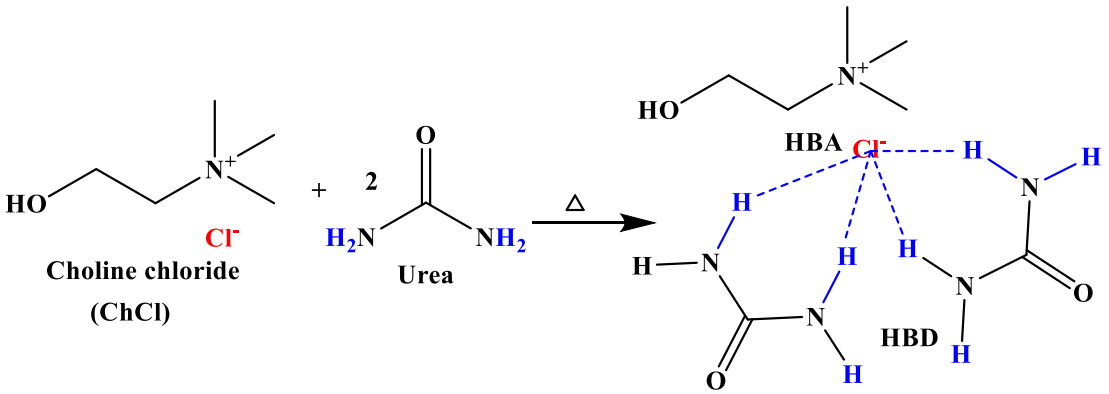


Fig. 2. Deep eutectic solvent (Reline) Synthesis.

programs: ChemDraw and chem3D .This ccrystal lattice of choline chloride (ChCl) quaternary ammonium salt with the chemical formula $[(CH_3)_3NCH_2CH_2OH]^+Cl^-$ which consists of choline cations $[(CH_3)_3NCH_2CH_2OH]^+$ and chloride anions (Cl^-) held together by ionic bonds between N^+ and Cl^- ions in addition to the hydrogen bonds between protons of choline hydroxyl group and Cl^- ions [7-10].

The formation mechanism of DESs is attributed to the disruption of conventional hydrogen bonds between individual hydroxyl hydrogens of choline chloride (as H-bond doners) and the chloride ions (as the hydrogen bond acceptor) (chloride ions), with the formation of new, stronger H-bonds between hydrogen donors (primary amine groups in urea) and H-bonds acceptors (chloride ions). These significant reduction in lattice energy leading to a lower melting point to about 12°C (liquid mixture at room temperature, i.e. deep eutectic) compared to individual components (solid choline chloride 302–305°C and solid urea133–135°C) as shown in Fig. 2. [1-3] [11,12].

The choline chloride monoclinic system (crystal system in crystallography, characterized by its unique symmetry and lattice parameters), the basic crystal structure with chemical formula of $[C_5H_{14}NO]^+Cl^-$, which consists of choline cation and chloride anion, has a unit cell parameter along the length of axis's a, b and c respectively: a ≈ 8.7 Å, b ≈ 7.8 Å, c ≈ 12.4 Å and an angle between the a and b axis's is α = 110 (angles may vary with temperature/pressure) [13].

This deep eutectic solvent (Reline) was synthesis by mixing simply choline chloride (ChCl) and urea in a 1:2 molar ratio in a test tube and directly heated over the hot plate heater with the chemical equation was shown in Fig. 2 [14].

The strong hydrogen bonding between urea and the Cl^- anion of ChCl and four hydrogen atoms of the two moles of urea and the electrostatic and van der Waals forces from the interactions between choline quaternary nitrogen cation and urea disrupts the crystal lattice of both components, leading to a liquid at room temperature with a lower melting point than either pure component [15].

Finally, in this paper, as a continuation of previous work on the preparation of Reline and its use as a catalyst in organic synthesis, an aim was performed to replace urea with a different and safe type of hydrogen bond donor, alcoholic sugar (erythritol) and to test as a catalyst in organic preparation.

Erythritol, is a sugar alcohol (polyol) used as a low-calorie artificial sweetener**.** It occurs naturally in small amounts in fruits like grapes and melons, as well as in fermented foods. However, for commercial use, it is typically produced industrially through the fermentation of glucose (often from corn or wheat starch) [16].

In recent years, deep eutectic solvents (DESs) have seen a remarkable development to their fusion with nanoparticles, as this fusion has given rise to synergistic qualities that combine the elasticity of the hydrogen bonds in DES with the unique surface properties of nanoparticles. This synergy opens up vast prospects in catalyzing chemical reactions, improving extraction, enhancing thermal stability, and increasing efficiency in environmental and medical applications. The combination of DES and nanoparticles not only improves solubility or catalytic activity, but also contributes to the design of systems [17].

This DES and its mixture with nanoparticle will be used in organic applications as catalyst with comparison with the conventional method. Also, thermal gravimetric analysis (TGA) and differential thermal analysis (DTG), in addition to IR spectra will used to characterized this novel deep eutectic solvent.

MATERIALS AND METHODS

Materials

The chemicals utilized in this study came from Fluka, BDH, and Aldrich, and used without further

Table 1. The physical and percentage yields of compounds (I and II).

No.	Molecular Formula	Molecular weight	Melting Point °C	Yield%				Elemental analysis % theory (Calculate)		
				A	B	C	D	C	H	N
I	$C_{14}H_{11}NO_2$	225.08	121-123	80	88	91	92	74.65	4.92	6.22
II	$C_{15}H_{11}NO_4$	269.07	80-82	81	90	92	92	66.91	4.12	5.20

purification.

Instruments

Melting point apparatus

Melting points (°C) were measured in open glass capillaries using the Stuart SPM30. Melting point apparatus in the Department of chemistry, college of science, University of Mosul.

Infrared apparatus (IR)

IR spectra (KBr disks) were recorded using a Bruker spectrophotometer at the University of Mosul's College of Science.

Nuclear magnetic resonance (NMR)

^{1}H-NMR and ^{13}C-NMR were recorded in Al-Basrah University/ Iraq on a Bruker (400 MHz), using DMSO as a solvent and TMS as an internal standard.

Thermal Gravimetric Analysis TGA of AC and Differential thermal analysis (DTA)

The thermal stability of this novel deep eutectic solution (Erythrine) was determined using METTLER TOLEDO device and its thermal analysis program: Stare Evaluation Software version 15.01 (2018). Horizontal balance was use in order to scanning TGA. Experiments were carried out using Mettler-toledo TGA/DCS star system in pottery (silica) crucibles at temperatures ranging from (25 to 600◦C) with heating rate of (20°C/min) under air atmosphere.

Novel deep eutectic solvent of choline chloride/ erythritol (Erythrine)

Choline chloride (ChCl) and alcoholic sugar (erythritol) in a 1:2 molar ratio, were mixed in a test tube directly heated over the hot plate heater

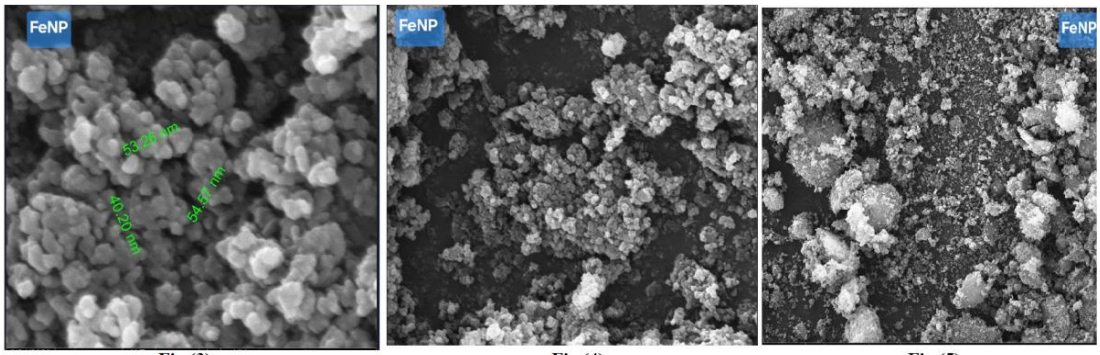


Fig (3) Fig (4) Fig (5)

Fig. 3. SEM image of FeNPs.

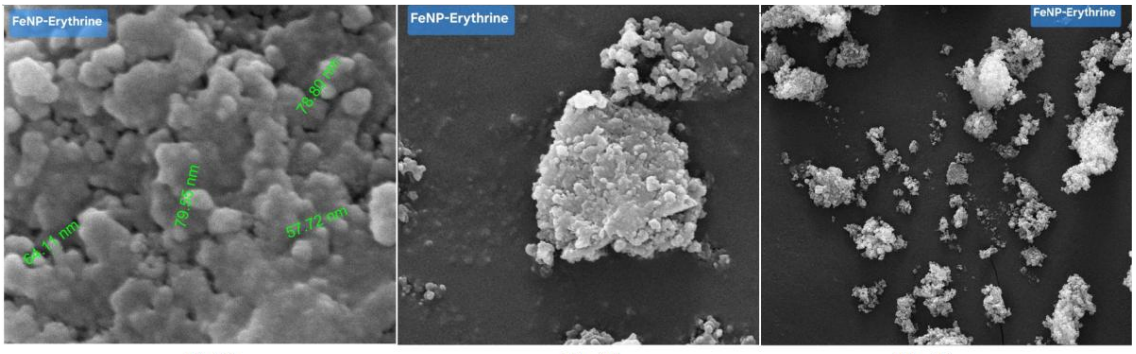


Fig (6) Fig (7) Fig (8)

Fig. 4. SEM images of FeNPs mixed with the deep eutectic solvent (Erythrine) FeNP-Erythrine (Irone nanoparticles mixed with Erythrine).

until the mixture becomes clear (transparent), then it is left to cool, and its freezing point is measured, which is approximately (38°C) [18].

Synthesis of FeNP-Erythrine (Irone nanoparticles mixed with Erythrine)

Dissolve $FeCl_3$ and $FeCl_2$ in 2:1 molar ration in deionized water. Under continuous stirring, sodium hydroxide (NaOH) solution was added dropwise under vigorous stirring until pH ≈ 10., and the stirring was continuous until black suspension of Fe_3O_4 nanoparticles will form. This freshly prepared FeNP was washed with deionized water several times to remove excess ions suspension, all the stages of these preparations were shown in Fig. 3. These FeNPs were then mixed with the deep eutectic solvent (Erythrine) and the result mixture was stirred for one hour to ensure homogeneity [19], all the stages of these FeNP-Erythrine (Irone nanoparticles mixed with Erythrine) were shown in Fig. 4.

Synthesis of (Z)-1-(benzo[d][1,3] dioxol-5-yl)-N-phenyl methanimine (I) and 1-(benzo[d][1,3] dioxol-5-yl)-N-phenyl methanimine (Z)-4-((Benzo[d][1,3] dioxol-5-ylmethylene) amino) benzoic acid (II)

Method (A): Using conventional catalyst (AcOH)

A mixture of equimolecular weight of a proper amine derivative (p-aminobenzoic acid (PABA) or aniline and piperonaldehyde in ethanol, with a few drops of acetic acid were refluxed for 2 hours. The mixture product was cooled to room temperature, washed with cold ethanol and filter, recrystallized from ethanol and calculated the melting point, percentage yield of the product, as seen in Table 1 [20].

Method (B): Using deep eutectic solvent catalyst (Erythrine)

A mixture of equimolecular weight of a proper amine derivative (*p*-aminobenzoic acid (PABA) or aniline and piperonaldehyde in ethanol, with one-gram Erythrine was refluxed for (2) hours with stirring and work up as in Method (A), as shown in Table 1 [21].

Method (C): FeNP-Erythrine

A mixture of equimolecular weight of a proper amine derivative (*p*-aminobenzoic acid (PABA) or aniline and piperonaldehyde in ethanol, with FeNP-Erythrine (0.1gm) was refluxed for (2) hours with stirring and work up as in Method (A), as shown in Table 1 [22].

RESULTS AND DISCUSSION

Novel deep eutectic solvent of choline chloride/ erythritol (Erythrine)

In this work to prepare the catalyst deep eutectic solvent (Erythrine), which was formed by mixing very simply in test tube a choline chloride (ChCl) as hydrogen bond acceptor (HBA) and urea as hydrogen bond donor (HBD) in a 1:2 molar ratio, and directly heated over the hot plate heater until the mixture becomes clear (transparent), then it is left to cool, and its freezing point is measured, which is approximately (50°C) via the following equation, Fig. 5 [23,24].

The using of this polyhydroxy alcohol(erythritol), plays a crucial role in forming the deep eutectic solvent (Erythrine) with choline chloride (ChCl) due to their multiple hydroxyl (–OH) groups, which are disrupt the choline chloride salt crystal lattice and form a liquid solvent due to their enhancement

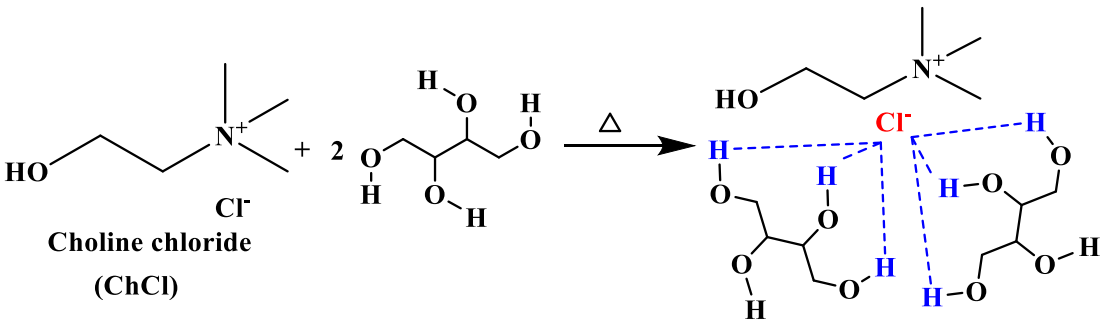


Fig. 5. Synthesis of novel deep eutectic solvent of choline chloride/erythritol (Erythrine).

of hydrogen bonding (using of three of its four hydroxyl groups), these are all-embracing or synergetic hydrogen bonds with robust interaction helps in breaking the ionic lattice of ChCl, and enhancing the disorder in the system, reducing the melting point of the mixture to 50°C compared to pure ChCl (of an elevated melting point of ~302°C),[25,26].

In addition to the improvement of the solution of the catalyst by forming a 3D hydrogen-bonded network as shown in the Erythrine product chemical structure Fig. 5.

Although the melting point of this deep eutectic solvent is higher than room temperature, the power of its actions as a catalyst in producing percentage yields above 90% and the high degree of safety of its components (erythritol and choline chloride) make it preferred to use, in addition to the excellent properties that will be discovered in the coming experiments will increase the importance of this DES, (Erythrine) [27,28].

The FT-IR spectroscopy of the novel deep eutectic solvent derived from the choline chloride and erythritol (Erythrine) gave the following as shown in Fig. 6.

The first important peaks were the fingerprint region of the vibration beaks related to (CN and C-O) of the confined area is characterized by

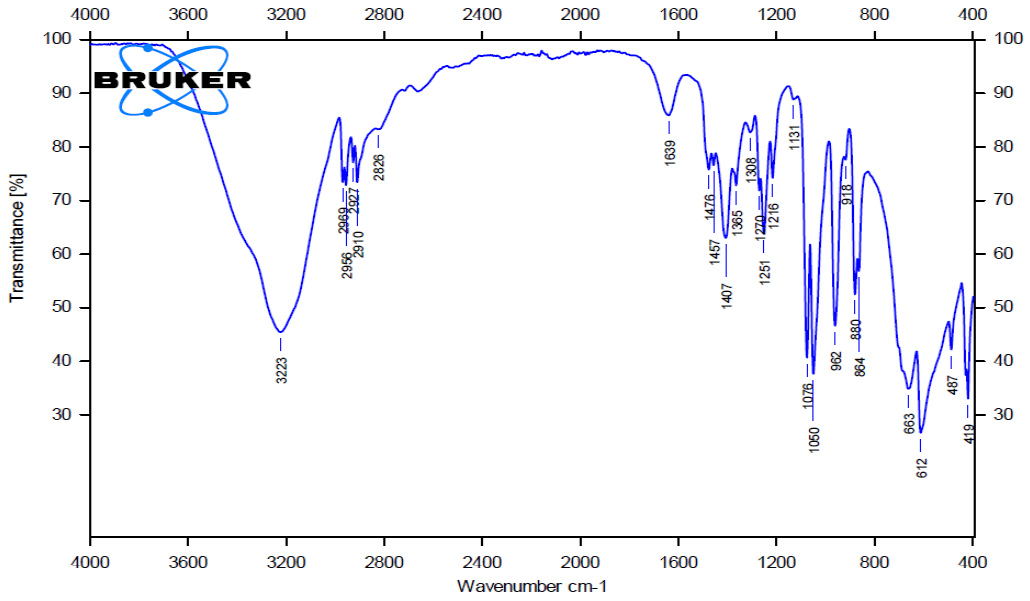
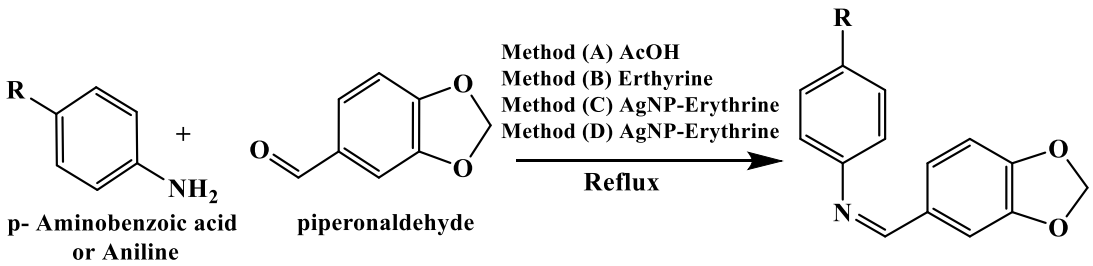


Fig. 6. The FT-IR spectroscopy of the novel deep eutectic solvent (Erythrine).



R = - COOH 4-((benzo[*d*][1,3]dioxol-5-ylmethylene)amino)benzoic acid (I)

R = -H 1-(benzo[*d*][1,3]dioxol-5-yl)-*N*-phenylmethanimine (II)

Fig. 7. Synthesis of Schiff bases.

1457cm^{-1} and 400 cm^{-1}with sharp and precisely peaks that reflect the real structure. Also, the hydroxyl group (OH) at 3223 cm^{-1} appear very wide and strong as a result of the stretching vibration of this group, and the reason for this large width of this beam is the presence of a strong network of condensed hydrogen bonds formed between erythritol and the chloride ion (Cl^-), which is the basis for the formation of deep eutectic solvent (DES).This is in addition to the carbon-hydrogen aliphatic (C-H) bond-binding beaks found in the methyl (CH_3) and methylene (CH_2) groups found in the structure of choline and erythritol. Finally, the appearance of the quaternary ammonium nitrogen 962 cm^{-1} and the sharp peaks of C-O at 1050 cm^{-1} provided conclusive evidence of the occurrence of the physical reaction and the formation of hydrogen bonds characterizing the deep eutectic solvent **(**Erythrine), as s Shown in Fig. 7.

In a synthetic study to find how well this Erythrine speeds up key organic reactions (like synthesis of Schiff bases I and II) and comparing yields percentages to standard conventional catalyst like acetic acid, four experiments were run in this work, the first method (A), a conventional catalyst (acetic acid) was used and comparing it with the using of novel deep eutectic solvent (Erythrine) (Fig. 7).

The catalyzing effects of the few drops of acetic

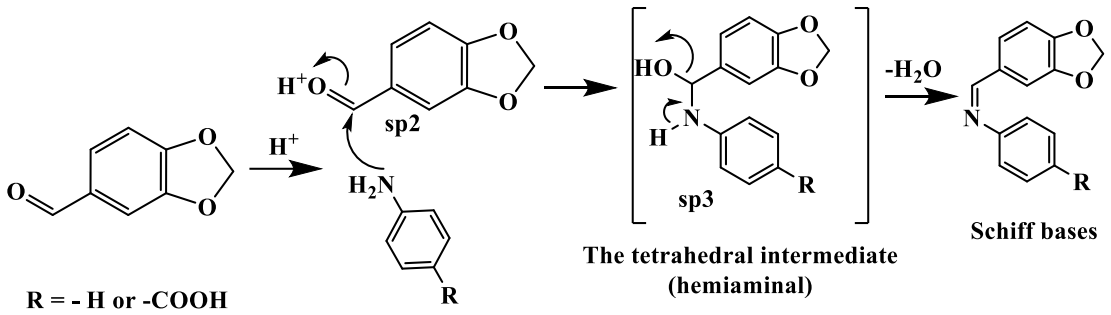


Fig. 8. The acid catalyst tetrahedral mechanism of Schiff bases synthesis.

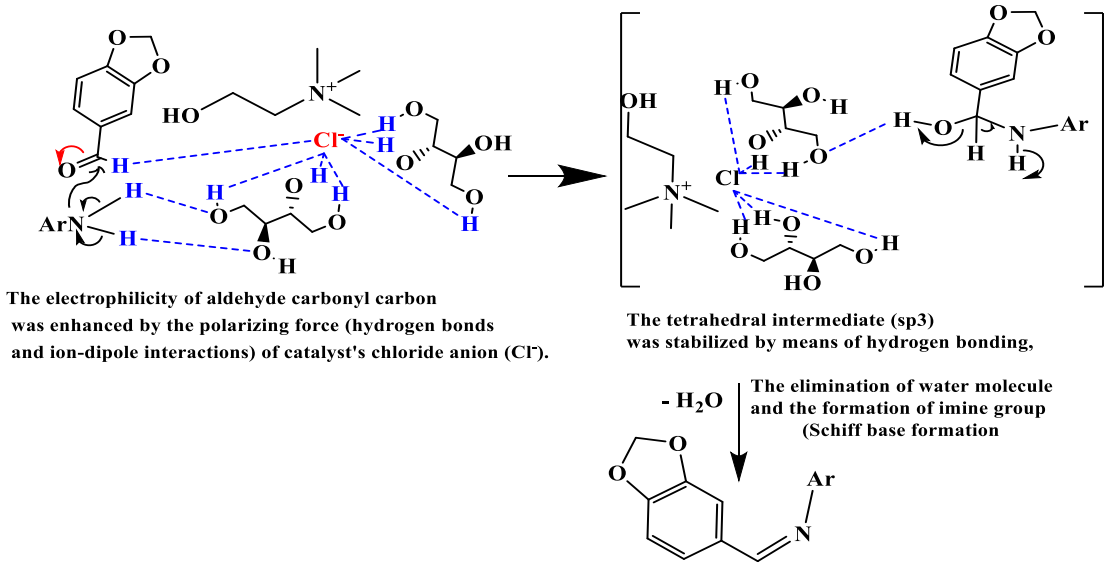


Fig. 9. The Schiff base synthesis by using novel deep eutectic solvent of choline chloride/erythritol (Erythrine).

acid assisted in protonating the carbonyl oxygen of aldehyde making it more reactive before the aniline nitrogen nucleophilic attack. This reaction was proceeded via the tetrahedral mechanism to form hemiaminal intermediate which loses a water molecule to produce the Schiff base as shown in the following mechanism (Fig. 8) [29].

The percentages yields of these two Schiff bases as shown in Table 1 were (80%) and (81%) respectively with long time reaction (5 hours).

In method (B), which is the first time using this novel deep eutectic solvent (Erythrine), 0.1 gram of this novel ionic liquid gave (88%) and (90%) respectively with two hours of the reaction time, while in the methods (C) and (D) the percentage yields were reached to mor than 90% , i.e (91,92) as shown in Table 1.

The mechanism of the actions of this catalyst on the formation of Schiff bases was suggested in Fig. 9. The electrophilicity of aldehyde carbonyl carbon (the first species in this reaction) was enhanced by the polarizing force (ion-dipole interaction) of catalyst's chloride anion (Cl^-). At the same time, the nucleophilicity of amine (the second species) was heightened by the catalyst hydroxyl groups via their alignment with this amine via hydrogen bonding, this facilitated the nucleophilic attacking of the amine to the activated carbonyl carbon, forming a tetrahedral intermediate via the (tetrahedral mechanism), this intermediate was stabilized by means of hydrogen bonding) [23]. Finally, the elimination of water molecules which is the driving force of forming the imine group (Schiff base products) was accelerated by the hygroscopic nature of the catalyst [30].

This novel DES (Erythrine) mixture with the Iron (FeNP) in method (C) increased the yield to (92%). This was because stabilization and anti-agglomeration of nanoparticles, DES acts as a sticky and stabilized medium, preventing the aggregation of nanoparticles (agglomeration), thus keeping the active surface area high, and this directly increases the effectiveness of the catalyst, in addition to the providing a biocompatible and chemically compatible reaction environment, because the DES dissolves many organic and inorganic materials, facilitating the access of reactants (substrates) to the active surface of nanoparticles, and increasing the rate of effective collisions, the important synergistic effect of the DES functional groups, i.e. hydroxyl groups, chloride, or acids can be directly involved in the catalytic reaction or activate bonds in the reactants, in conjunction with the action of nanoparticles. Finally, the effects of improvements recyclability of the DES facilitates the recovery and reuse of the cofactor, maintaining the catalytic power of multiple cycles [31].

The three microscopic images taken by the Electron Scanning Microscope (SEM) of the Forms 3-5 (FeNP) nano iron samples show their morphological structure prior to the mixing process, as it is clearly shown in Fig. 3 of the very

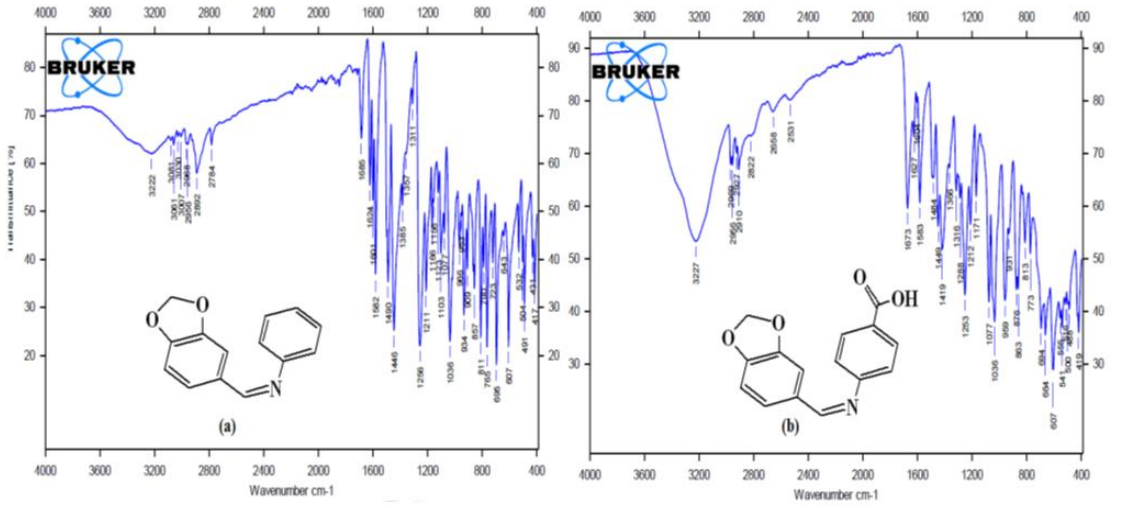


Fig. 10. Spectroscopy a. of the Schiff base (I) and b. of the Schiff base (II).

high magnification 120,000× that the particles have an irregular aspherical shape and their individual diameters are distributed in the pure nanometer range between nanometers and nanometers, while 40.20 and 54.57 , Fig. 3 reveals at a distance of micrometers and Fig. 3. At a micrometer away from a very strong tendency of these particles to clump and clump into large micrometric masses due to the superior surface energy and the magnetic attraction forces exchanged between them. When mixed with 20 Deep Eutectic Solvents, it is scientifically expected that the dense network of solvent's hydrogen bonds and high viscosity will play a pivotal role as a natural dispersant and stabilizer, as the solvent molecules will surround individual nanoparticles to break up the clumps phenomenon in microscopic forms, which prevents their reassembly and slows their deposition rate by gravity, a behavior that ensures a highly stable and homogeneous nanosuspension, especially if the ultrasonic mixing process is incorporated to ensure that the solvent penetrates the iron masses effectively. While after the mixing of these nanoparticles with the novel deep eutectic solvent (Erythrine) with iron (FeNP) nanoparticles as the Fig. 4 there were comprehensive integrated scientific explanation paragraph explaining the changes and the effect of the novel deep eutectic solvent (DES) on the efficiency and strength of the cofactor:

Microscopic images of the Erythrine-modified iron nanocomposite show clear structural changes compared to pure nanoiron before mixing, where the high-magnification a slight growth in individual particle sizes to their measured diameters between nanometers and nanometers was shown in Fig. 4, which was a direct indication of the success of the encapsulation and functional loading process on the surface, while the Fig. 4 were shown the less-transformative magnification dimensions of the cluster pattern, instead of large, random endocrine masses, the particles appear as scattered, more open, and porous clusters. When combined with the novel deep eutectic solvent (DES), this mixing lead to a quantum leap in the strength and efficiency of the catalytic activity, as the polar DES molecules and dense hydrogen bonds act as a diffuse and supporting medium that prevents the particle from agglomerating again, maintaining the active surface area at its highest level, the solvent also provides chemical protection that prevents the oxidation of the active zero valence iron, and the synergistic effect between the functional groups of the solvent and nano-iron facilitates the transfer of electrons and the attraction of the reactants to the active centers of the cofactor, significantly increasing the speed and efficiency of the target chemical reaction

The FT-IR spectroscopy of the Schiff base (I), 1-(benzo[d][1,3] dioxol-5-yl)-N-phenyl methanimine (I) and 4-((Benzo[d][1,3] dioxol-5-ylmethylene) amino) benzoic acid (II), Fig. 10a and b respectively, the comparison between the two spectrums (a) and (b) showed a significant congruence in the overall pattern of absorption of the basic structure of the two Schiff bases, especially in the fingerprint region below(1500cm^{-1}) and the azomethine bonds (C=N) which were clearly visible in both compounds (at 1624 cm^{-1}) in the first compound, and overlapping

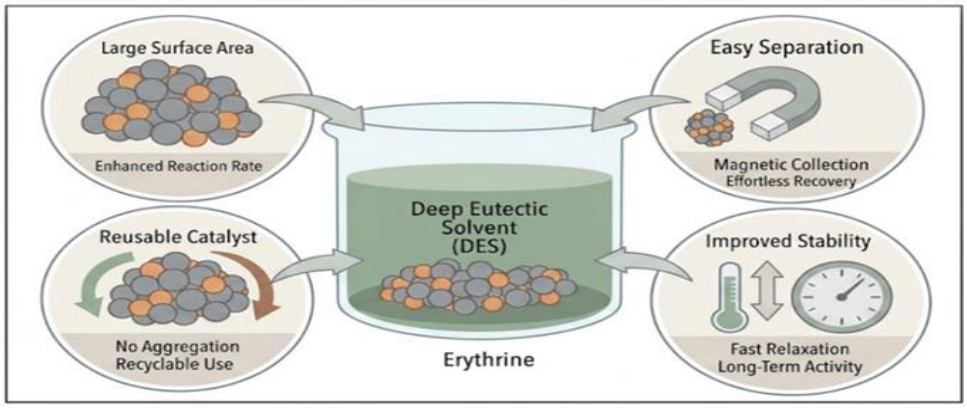


Fig. 11. The synergy of silver nanoparticles with Erythrine.

or appearing very close at (at 1627 cm^{-1}) in the second compound. Also, the benzodioxol group (-O-CH_2-O-) vibration peaks of identical cyclic ethers (CH_2-O-Ar) vibrated in both spectrums showed the same between (1036cm^{-1}) and (1211-1212 cm^{-1}). Finally, the (C=C) aromatic ring vibrations of the bending and stretching of the aromatic rings in the fingerprint region (e.g., sharp beams at the 800-600 cm^{-1}) remain very similar in their distribution and intensity. The only spectral difference is due to the replaced carboxyl group in Schiff base (II), which caused a significant change in the region above (2500 cm^{-1}) due to the broad (O-H) peak and the appearance of the strong carbonyl peak (C=O) at (1673 cm^{-1}), otherwise the spectral footprint of the structure remains constant and exactly identical to the first compound as shown in the Fig. 10 [32-34].

Thermogravimetric Analysis (TGA)

TGA reveals Erythrine's thermal stability range behavior is a balance between component interactions and individual stabilities, with onset decomposition primarily occurring at 318.88°C and the peak decomposition rate of inflection point was 46.10°C (an inflection point is a point on the graph of a function where the concavity changes, i.e. where the function transitions from being concave up to concave down, or vice versa) [1-2]. The completion of decomposition was 366.92°C, thus the DES is thermally stable up to ~320°C, making it suitable for applications below this temperature, this decomposition characteristics with mass loss of 94.63% (Delta m). The TGA kinetics result of activation energy which was equal to 211.84 kJ/mol, while the reaction order (n) ≈ 1.08 (first-order kinetics), this was because the decomposition occurred in a single dominant step with rapid mass loss, consistent with the breakdown of the eutectic structure. The low residue suggests high purity and minimal inorganic contaminants [35]. The TGA study of the decomposition dynamics of the peak decomposition rate was 0.76 %/min at 346.10°C (from DTG inflection point).

Also, the heating rate was 40°C/min (aggressive, yet typical for screening), this concluded that the sharp DTG peak confirms a narrow decomposition window (318–367°C), indicating uniform degradation behavior without intermediate phases. The advantages of this deep eutectic solvent Erythrine **(**choline chloride/erythritol) comparison to typical DES systems indicated that this shows higher and good thermal stability (≤320°C) with a sharp, single-step decomposition profile. Its high purity (94.63% mass loss) and predictable first-order degradation kinetics make it viable for medium-temperature applications. For long-term use, a safety margin below 300°C is recommended, and these results were better than many DESs (e.g., urea-based DES often decompose below 200°C), this stability aligns with erythritol's high melting point (∼120°C) and hydrogen-bonding [23]. Supporting ionic liquids such as the deep eutectic solvents (DESs) on nanoparticles unifies the utility of the high surface area of nanomaterials with the green-chemistry and eco-friendly properties of (DESs). This integration increases the stability of catalyst, selectivity and stability, with increasing the amount and life of the recovery (DESs) when using as reactions catalyst, these benefits improve the nanomaterial properties and improve catalytic performance [23, 35, 36].

This is on the one hand, but in terms of practical benefits**,** advantages and starting with structural stability the DESs avoid nanoparticle "agglomeration" thus extending catalyst lifetime, also, DESs guide reactions toward desired products by modifying the local environment thus enhances the selectivity, [37-38].

The pivotal role of super-magnetism in magnetic nanoparticles, this property gives the particles a large active surface area, which enhances the rate of reaction within deep eutectic solvents (DESs). The ability to efficiently separate particles using an external magnetic field allows for simplified isolation and purification after the reaction, in addition, reusability is guaranteed to the super-paramagnetic nature, as particles do not collect after the magnetic field is removed, allowing multiple cycles of reuse of the adjuvant without loss of efficiency. The dynamics of rapid relaxation prevent the loss of magnetic properties, and maintain catalytic activity for long periods.

Finally, the overall synergy between (DESs) and magnetic nanoparticles combines environmental stability with high catalytic efficiency, resulting in a highly efficient and eco-friendly green system, as shown in in addition to the large surface area, enhanced the reaction rate, easy separation (by magnetic collection), effortless recovery, reusable catalyst, no aggregation, recyclable use, improved stability, fast relaxation and long-term activity of the novel deep eutectic solvent (DES) Erythrine as

shown in the Fig. 11.

CONCLUSION

This paper ensured nanoparticles were evenly dispersed in this novel deep eutectic solvent Erythrine, making them suitable for applications as catalysis and also these promised steps has successfully developed an innovative type of deep eutectic solvent (DES) "Erythrine", as a sustainable and environmentally safe alternative to conventional ionic liquids. This was achieved by replacing urea with a safer hydrogen bond donor, natural sugar alcohol (erythritol), and mixing it with choline chloride salt in a molar ratio (1:2). The results of structural and thermal diagnosis and characterization using FT-IR spectroscopy, thermal and differential gravitational analysis (TGA/DTG) proved the tremendous success of the eutectic effect, which is a very significant reduction in the melting point of the solvent compared to the high melting point of pure choline chloride, as a result of the breakdown of the crystal network of the salt and the formation of a strong and branched hydrogen bond network between the two components, giving Erythrine excellent thermal stability.

In terms of catalytic efficiency assessment, the study revealed that innovative systems were superior and had high chemical safety when applied in the synthesis of biocritical Schiff bases compared to classical methods. The conventional method (A) using acetic acid catalyst resulted in very good yields ranging from 80% to 81% for the two Schiff bases, respectively. In comparison, the use of pure Erythrine catalyst increased the efficiency of the reaction to reach a percentage yield 88% - 90%. The study achieved the catalytic peak when the nanoparticles of iron were combined with the solvent, where the yield ratios jumped to (92%). The research concludes that the fusion of nanoparticles with the innovative eutectic solvents give the system unique synergistic properties that combine the elasticity of the hydrogen bond network with the characteristic surface properties of the nanoparticles, which definitively contributed to the acceleration of reactions, improved solubility, and elevated catalytic activity with excellent production yields, opening new horizons for sustainable organic applications. The distinct morphological modification of iron nanoparticles after being modified with erythrine which is manifested in the increase in the diameters of individual grains to a range of nanometers to nanometers is attributed to the formation of a new enveloping layer and chemical compatibility on the nano surfaces, which corresponds to the success of the functional loading process. This volumetric growth is accompanied by a geometric and structural reorganization of the agglomerations, which have transformed from their previously compact and compact nature into a dispersed structure with a clear porosity that allows for better permeability and diffusion of the reactants. This transformation the structure plays a pivotal role in enhancing the catalytic performance of the system when combined with the new deep eutecticolvent (DES), as the high viscosity and dense network of the solvent's characteristic hydrogen bonds contribute to the stabilization of this dispersion and the prevention of the regrowth of nano-agglomeration. This synergistic behavior ensures that the active sites of the nanoiron remain fully exposed and in direct contact with the reactive medium without vacuum obstructions, as well as the effective protective role provided by the solvent to protect zero valence iron from rapid oxidation, and facilitate the mechanism of the transfer of charges and electrons, which is positively reflected on raising catalytic efficiency and giving the cofactor high operational stability in the long term.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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