Fe (III) adsorption using Fe (III) ionic imprinted polymer from polyeugenoxy acetate crosslinked with ethylene glycol dimethacrylate (EGDMA)

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Abstract. Ionic Imprinted Polymer (IIP) with polyeugenoxy acetate (PA) as the base polymer, EGDMA as the crosslinking agent, and AIBN as the initiator have been synthesized to adsorb Fe(III) metal ions. NIP (non-imprinted polymer) was used as a comparison for this study. The produced polymer was characterized using FTIR spectroscopy and SEM-EDX. Adsorption rate and capacity were examined through adsorption isotherm and kinetics studies for all adsorbents. The adsorption selectivity test was done with mixed-metal solutions of Zn(II), Cu(II), Cd(II), and Ag(I). The results of SEM analysis on IIP-PA show a more uniform and regular size than NIP-PA. Adsorption of Fe(III) in IIP-PA and NIP-PA follows pseudo-second-order kinetics, and the adsorption isotherm both follow the Langmuir theory. The adsorption capacity of Fe(III) metal ions in IIP-PA is 10.88 mg/g, and the adsorption capacity of NIP-PA is 7.94 mg/g. The mechanism of adsorption of Fe(III) metal ions in IIP-PA and NIP-PA was dominated by hydrogen bonds between the -COOH groups and Fe(III). In binary solutions, IIP-PA adsorption selectivity coefficient is higher than NIP-PA with the order of Fe(III)/Zn(II)>Fe(III)/Cu(II)>Fe(III)/Cd(II)>Fe(III)/Ag(I).

1. Introduction
Water pollution is caused by domestic waste containing toxic heavy metal ions [1]. Iron metal is a heavy metal pollutant that is often found in water. The total concentration of iron dissolved in the waters ranges from 0.05 to 6.4 mg L⁻¹ (0.89-114.3 M) [2]. One simple method to reduce the level of ferrous metal in water with a relatively high yield of effectiveness is adsorption [3], though the selectivity of adsorbents on targeted metal ions needs to be further developed. One material development that has high adsorption selectivity for the presence of iron in water is Ionic Imprinted Polymer (IIP) [4-6].

The use of eugenol as a biomaterial for heavy metals extraction has been widely developed, such as the synthesis of eugenol into polyeugenol oxide acetate using solvent extraction methods [7], Ionic Imprinted Polymer (IIP) from polyeugenol has been used as an adsorbent with PEGDE crosslinking agent (polyethylene glycol diglycidil ether) to adsorb Fe metal ions [8], ionic imprinted polymer (IIP) from eugenol derivative compounds in the form of polyeugenol (PE), polyeugenol acetate (PA), and poly thiophene-2-methyl-eugenox-acetate (PFMEA) have been used as adsorbents with EGDMA crosslinking agent for the adsorption of Fe metal ions [9].

In this research, IIP was synthesized using polyeugenoxy acetate (PA) with EGDMA (ethylene glycol dimethacrylate) to find out adsorption rate and capacity through adsorption kinetics and
isotherm studies, Fe(III) metal ion adsorption mechanism, and adsorption selectivity of Fe(III) metal ions with other metal competitors.

2. Research methods

2.1. Equipment
Laboratory glass equipment, reflux equipment, filter paper, analytic balance (Mettler-200), magnetic stirrer, pH meter (HACH E C20), UV-Vis spectrophotometer (Shimadzu), AAS (Perkin Elmer), FTIR (Nicolete Avatar 360), SEM (JSM-6510).

2.2. Materials
Eugenol p.a, BF₃-diethyl ether, EGDMA (Ethylene glycol dimethacrylate) and AIBN (2,2’,Azobis(2-methylpropionitrile) were purchased from SIGMA Aldrich. Na₂SO₄ technical, HCl, NaHCO₃ technical, NaOH p.a, CICH₂COOH p.a, chloroform p.a, diethyl ether p.a, KNO₃, KSCN, aquabidest, Fe(NO₃)₃, Cd(NO₃)₂, Zn(NO₃)₂, Cu(NO₃)₂, AgNO₃ and Na₂EDTA were purchased from Merck.

2.3. Research Procedure

2.3.1. Poly(eugenol) (PE) synthesis. A total of 5.8 g of eugenol (0.035 mol) was added with 1 mL BF₃-diethyl ether and stirred for 4 hours with the addition of 0.25 mL (0.002 mol) of BF₃-diethyl ether every 1 hour. The polymerization occurred for 12-16 hours, and then 1 mL of methanol was added to stop the polymerization process. The gel formed was dissolved in chloroform and washed with aquabidest until it reached a neutral pH. The water content in the solution was then removed by adding anhydrous Na₂SO₄. Afterward, the solution was kept at room temperature until the precipitate dried. The precipitate was then ground to form poly(eugenol) and weighed. It was then analyzed by FTIR.

2.3.2. Poly(eugenox acetate) (PA) synthesis. Poly(eugenol) as much as 5 g (0.03 mol) was added with 17.5 mL of 33% (0.825 mol) NaOH solution and then stirred for about 30 minutes. It was added with 12.5 mL of 50% chloroacetic acid (0.529 mol) gradually whilst continued being stirred. It was then heated in a water bath with a temperature of 80-90°C for 2 hours. After the mixture cooled down, it was added with 6 M HCl until the mixture reached pH 1. Extraction was done using diethyl ether for 3 times, the result then extracted with sodium bicarbonate 5% b/v (0.059 mol) for 3 times, and then acidified to pH 1 using 6 M HCl. Subsequently, it was filtered, dried at room temperature, and weighed. The results were analyzed using FTIR.

2.3.3. Ionic imprinted polymer (IIP-PA) synthesis. 0.5 g of poly(eugenox acetate) (0.003 mol) was mixed with Fe(III) ions of a certain concentration under pH of 3 for 24 hours. The product was filtered and then air dried. The resulting polymer deposit was freed from Fe(III) ion which was bound to acid for 24 hours.

2.3.4. Non-imprinted polymer (NIP-PA) synthesis. NIP-PA synthesis was done by the same method as IIP-PA, but without the binding of Fe(III) metal ions.

2.3.5. Material characterization. Characterizations of IIP-PA and NIP-PA adsorbents were carried out using FTIR and SEM-EDX.

2.3.6. Adsorption test. An adsorption test was done by the batch method in the condition set to pH 3, with 50 mg of IIP-PA and NIP-PA adsorbent, 10 mg/L Fe(III) solution, and variation of contact time (0-1440 minutes) to determine adsorption kinetics. To determine the adsorption isotherm, adsorption
test was conducted with Fe(III) metal ion concentrations ranging from 0-150 mg/L. Afterward, the mixture was filtered, and the concentration of Fe(III) in the filtrate was analyzed by AAS.

2.3.7. Adsorption selectivity test. The adsorption selectivity test was carried out by adding 50 mg of adsorbent to 10 mL of each mixed-metal solutions consisting of a mixture of Fe(III)/Cd(II), Fe(III)/Cu(II), Fe(III)/Zn(II), and Fe(III)/Ag(I) with ion concentration of 10 mg/L. The adsorption test was carried out in a batch system using a magnetic stirrer at pH 3. This test was carried out for IIP-PA and NIP-PA to provide comparative results.

3. Results and discussion

3.1. Polyeugenol synthesis

The polymerization of eugenol into polyeugenol is a cationic addition polymerization process. Eugenol undergoes an addition reaction through its allyl group. The mechanism of the polymerization reaction is presented in Figure 1.

![Figure 1. Eugenol formation mechanism](image)

In the initiation stage, BF₃-diethyleter acts as Lewis acid catalyst that causes an addition reaction causing the allyl group in eugenol to undergo termination of the double bond. Termination of the double bond occurs because the empty orbitals in BF₃ are transferred to the eugenol monomers thus, carbonium ions are formed [10]. The addition reaction at the initiation stage follows Markovnikov's law which states that the carbonium ion stability determines the reactivity and its ability to merge with the next monomer [11]. The carbonium ion produced will then cause an additional reaction to occur in another monomer at the propagation stage [12].

In the propagation phase, there is a formation of chains from eugenol monomers. The process is continued until a long chain that consists of monomers is obtained. The termination phase is the stopping stage of the polymerization reaction process. It is done by adding methanol. At this stage, carbonium ions will bind to their partner anions, namely the CH₃O⁻ group causing polymerization to stop and establish the formation of a methoxy group at the end of the polymer chain [13-14]. The experiment produced polyeugenol (PE) in the form of orange powder with a yield of 96.4%, where it was later analyzed using FTIR. The results of polyeugenol FTIR spectra analysis can be seen in Figure...
2. The formation of polyeugenol can be implied from the reduced spectra of vinyl eugenol groups at wavenumbers 995.27 cm\(^{-1}\) and 910.40 cm\(^{-1}\). This indicates polymerization.

![Figure 2. FTIR graph comparison of eugenol dan polyeugenol](image)

3.2. Polyeugenoxy Acetate (PA) synthesis

Polyeugenoxy acetate (PA) has an active acid group (COOH) which is supposed to have an affinity towards Fe(III) metal ions. According to the hard and soft acids and bases (HSAB) theory, an acid group is a hard base group that will form a strong complex with a hard acid, such as Fe(III) metal ion.

In the synthesis process, polyeugenol would react with NaOH through the hydroxyl group forming Na-polyeugenolate salt. The protons in OH of polyeugenol would be easily released because the resonance of the benzene ring stabilized the anions. Na-polyeugenolate was then reacted with chloroacetic acid, resulting in polyeugenoxy acetate compound [13,15]. The PA formation reaction is shown in Figure 3.

![Figure 3. PA formation mechanism](image)

Polyeugenoxy acetate compound which had been successfully synthesized in the form of dark brown precipitate with a yield of 91.09% was then analyzed using FTIR. The results of FTIR analysis of polyeugenoxy acetate is shown in Figure 4.

![Figure 4. Graph of PE and PA FTIR result comparison](image)
In the FTIR spectra above, there are absorption bands in wavenumber of 3448.72 cm\(^{-1}\) indicating hydroxyl groups (-OH), absorption bands in the range of 2900-2800 cm\(^{-1}\) indicating saturated carbon groups (Csp\(^3\)-H), the absorption band of 1735.93 cm\(^{-1}\) shows the presence of acid carbonyl group (C=O), absorption bands of 1597.06 cm\(^{-1}\) and 1512.19 cm\(^{-1}\) indicate an aromatic group (C=C), the absorption band of 1458.18 cm\(^{-1}\) indicating methylene groups (-CH\(_2\)-), the absorption band of 1149.57 cm\(^{-1}\) indicating acid CO groups, and the absorption band of 810.10 cm\(^{-1}\) band indicating that the aromatic is substituted. It could be concluded that the synthesis of polyeugenoxy acetate compounds was successful.

3.3. IIP-PA and NIP-PA synthesis
The synthesized PA compound was contacted with a template of Fe(III) metal ion 50 ppm under pH 3 condition for 24 hours. The resulting PA-Fe was then cross-linked with EGDMA crosslinker in chloroform solvent, added with AIBN as the initiators, and then refluxed. The results were dried and analyzed with FTIR. The release of Fe(III) metal ion template was then conducted through acidification for 24 hours using HCl. UV-Vis spectrophotometry analysis was used to determine that the Fe(III) had been released. Testing of Fe(III) metal ion level release was done by adding KSCN solution as a complexing agent. The reaction is as follows:

\[
\text{Fe}^{3+} + 3\text{SCN}^- \rightleftharpoons \text{Fe(SCN)}_3 \quad \text{(red)}
\]

The test to determine the amount of Fe(III) metal ions released was done until filtrate cleared with the addition of KSCN indicating that there were no longer Fe(III) metal ion templates released.

![Figure 5. Absorbance results of released Fe(III) templates](image)

Based on Figure 5, it can be implied that the PA compound is able to bind with Fe(III) metal ions. These results are following the HSAB theory that the acetate group (-COOH) in the PA compound, which belongs to the hard base group, can form complexes with hard acids in this case, is Fe(III) metal ion. PA compound which has been freed from the metal template, is neutralized and dried to obtain IIP-PA polymer that can be used to adsorb Fe(III) metal ions. In the synthesis of NIP-PA, the procedure carried out was the same as the synthesis of IIP-PA, but without the Fe(III) metal ion template bonding.

3.4. FTIR analysis of PA derivative compounds
Figure 6 shows that the carbonyl acid spectrum of 1735.93 cm\(^{-1}\) in polyeugenoxy acetate (PA) becomes increasingly sharp after Fe(III) metal ion binding and EGDMA crosslinking. This is possible because EGDMA also has C=O acid.
3.5. SEM-EDX Analysis

Results of analysis using SEM can be seen in Figure 7. If the surface appearance of the materials is compared, IIP pores seem to be more uniform and regular. This can be caused by Fe(III) which was bound and released from the adsorbent.

Figure 7. SEM micrograph of (a) PA-Fe-EGDMA, (b) IIP-PA, (c) NIP-PA

EDX analysis is used to analyze the composition in the polymeric matrix PA-Fe-EGDMA and IIP-PA. The EDX analysis results are shown in Table 1. The initial content of Fe metal in the aggregate of PA-Fe-EGDMA was 5%. However, after template release using HCl solution, only 0.2% of the content of Fe(III) metal is left. This shows that the acid solution used has released as much as 96% of the content of Fe(III) metal ions in the matrix.

Table 1. Mass percentage of elements in the adsorbents

| Elements | PA-Fe-EGDMA | IIP-PA |
|----------|-------------|--------|
|          | Mass (%)    | Atom   | Mass (%) | Atom   |
| C        | 64.1        | 0.97   | 75.2      | 0.99   |
| O        | 30.8        | 0.93   | 24.6      | 0.98   |
| Fe       | 5.0         | 0.79   | 0.2       | 0.62   |

3.6. Metal ion adsorption kinetics

The adsorption (efficiency) of Fe(III) increased quickly because the adsorbent’s active sites were mostly still available. The adsorption balance of IIP-PA and NIP-PA was reached in 120 minutes, and as the duration increased, the adsorption tended to reach a constant point. At this point, IIP-PA and NIP-PA were saturated. Under this condition, the adsorbents were unable to adsorb more Fe(III). A slight
decrease in efficiency is possibly due to some Fe(III) metal ions that had been adsorbed were released back into the solution due to stirring [3] (Figure 8).

![Figure 8. IIP-PA and NIP-PA Adsorptions of Fe(III)](image)

Adsorption kinetics were identified by modeling the data into the first-order and second-order kinetic equations. The first pseudo level kinetic equation is [16]:

\[
\log (q_e - q_t) = \log (q_e) - \frac{k}{2.303} t
\]

With \( q_e \) and \( q_t \) as adsorption capacities (mg g\(^{-1}\)) at equilibrium and t (min) time, and k is the rate constant for pseudo-first-order adsorption (L min\(^{-1}\)).

The pseudo-second-order kinetic equation is [17]:

\[
\frac{t}{q_t} = \frac{1}{kq_e^2 + \frac{1}{q_e} t}
\]

Table 2 shows the adsorption kinetics parameters results for Fe(III) metal ions in IIP-PA and NIP-PA. Low correlation coefficient for the log \( (q_e - q_t) \) vs t plot in the pseudo-first-order equation were given by all adsorbents whereas the \( t/q_t \) vs t plot gives a high correlation (> 0.99). It could be concluded that besides the trait of adsorbent, it is also influenced by the adsorbates (metal ions) natures [3]. The value of the rate constant \( (k_2) \) in IIP-PA is higher than the \( k_2 \) value in NIP-PA. The higher amount of pores in IIP-PA allows Fe(III) metal ions to reach the active site faster, then bind to the active site [18].

**Table 2. Fe(III) adsorption kinetics parameters**

| Parameter                     | Adsorbent |       |       |
|-------------------------------|-----------|-------|-------|
|                               |           | IIP-PA | NIP-PA |
| **Pseudo-first-order**        |           |       |       |
| \( q_e \) (mg/g)              |           | 0.1280 | 0.1580 |
| \( k_1 \) (L/min)             |           | 0.0138 | 0.00069|
| \( R^2 \)                     |           | 0.2623 | 0.11300|
| **Pseudo-second-order**       |           |       |       |
| \( k_2 \) (g/mg min) x \( 10^{-2} \) |   | 42.03  | 35.19  |
| \( R^2 \)                     |           | 1      | 0.9999 |
3.7. Adsorption isotherm

Adsorption isotherm portrays concentrations based on the equilibrium distribution of metal ions between solution and solid phase [19]. The common equation used is the Langmuir and Freundlich equations. The Langmuir isotherm occurs in a homogeneous monolayer [20]. The equation is [19]:

\[
\frac{C_e}{q_e} = \frac{1}{Q_m b} + \frac{C_e}{Q_m}
\]

With \( C_e \) as the concentration at equilibrium (mg L\(^{-1}\)), \( q_e \) as the number of adsorbates adsorbed at equilibrium (mg g\(^{-1}\)), \( Q_m \) adsorption capacity (mg g\(^{-1}\)), and \( b \) is the adsorption intensity known as the Langmuir constant (L mg\(^{-1}\)).

The Freundlich equation assumes that adsorption occurs multi-layer on the surface of the adsorbent and that the adsorption increases with increasing concentration [21]. The equation is [22]:

\[
\log q_e = \log K_f + \frac{1}{n} \log C_e
\]

With \( K_f \) as the relative adsorption capacity (Freundlich constant) and \( \frac{1}{n} \) as the adsorption intensity. In Table 3, it can be seen that IIP-PA and NIP-PA have the same adsorption properties where both follow the Langmuir model. The adsorption happens on the monolayer surface, which is overall homogeneous with the adsorption capacities of 10.881 mg/g and 7.936 mg/g for IIP-PA and NIP-PA, respectively. The number of pores corresponding to Fe(III) metal ions causes IIP-PA to have a greater adsorption capacity than NIP-PA. The energy of adsorption of adsorbate onto the adsorbent surface is:

\[
E = -\Delta G^0 = R T \ln K
\]

With \( \Delta G^0 \) is the change in Gibbs free energy (J mol\(^{-1}\)), \( R \) is the gas constant (8.314 J mol\(^{-1}\) K\(^{-1}\)), \( T \) is the temperature (K), and \( K \) is the equilibrium constant of adsorption (L mol\(^{-1}\)).

The physical adsorption released energy of approximately -20 kJ/mol, and the chemical adsorption energy release is about -40 to -800 kJ/mol [7]. The \( \Delta G^0 \) value for IIP-PA is -21.11 kJ/mol, and NIP-PA is -20.84 kJ/mol. This indicates that the adsorptions of Fe(III) in IIP-PA and NIP-PA are through physical and chemical adsorption. Furthermore, the template binding process causes the interaction of Fe(III) and adsorbents to be stronger [14].

| Table 3. Langmuir and freundlich isotherm constants of Fe(III) adsorption |
|-----------------|-----------------|
| Parameter       | Adsorbent \( Q_m \) (mg/g) | Adsorbent \( b \) (L/mg) | Adsorbent \( B \) (L/mol) \( \times 10^2 \) | Adsorbent \( R^2 \) | Adsorbent \( n \) | Adsorbent \( K_f \) | Adsorbent \( R^2 \) |
| Langmuir        | 10.881          | 0.0846          | 47.41            | 0.924           | 1.910            | 1.280           | 0.553           | 0.599           |
| Freundlich      | 7.936           | 0.0758          | 42.47            | 0.886           | 1.720            | 1.340           | 0.599           |

3.8. Fe(III) metal ion adsorption

To find out the mechanism that occurs in the process of adsorption of Fe(III) metal ions onto IIP-PA and NIP-PA, desorption was done sequentially against each adsorbent that had previously been interacted with Fe(III) solution. The use of a solution that will carry out the desorption process sequentially is hoped to dissolve metals adsorbed both physically and chemically [23].
Figure 9. Percentage of desorption of Fe(III) metal ions

IIP-PA and NIP-PA adsorption process are dominated by hydrogen bonds and complex formation, meaning that the Fe(III) adsorption mechanism in both adsorbents occur chemically. The mechanism of Fe(III) adsorption in both adsorbents happens by the mechanism of hydrogen bond and complex formation between the adsorbates and the active site of the adsorbent.

3.9. Adsorption Selectivity of Metal Ions

Adsorption selectivity determination was done by adding the adsorbent into the solutions containing Fe(III)/Cd(II), Fe(III)/Cu(II), Fe(III)/Zn(II), and Fe(III)/Ag(I). Adsorption was done for 24 hours under pH 3 conditions. The distribution ratio (D) and selectivity coefficient (α) are determined as [24]:

\[
D = \frac{q_e}{C_e}
\]

\[
\alpha = \frac{D(\text{Fe(III)})}{D(M_c)}
\]

Where \(M_c\) is the competitor metal ion. The selectivity of adsorbent for a specific kind of metal ion is affected by the existence of competing metal ions [25]. The data of selectivity values for each adsorbent with different metal ions is shown in Figure 10.

Figure 10. Graph of adsorbent adsorption selectivity

From Figure 10, it can be seen that IIP-PA and NIP-PA are selective towards Fe(III) metal ions with the order of effectiveness for both mediums as follows Zn\(^{2+}\) > Cu\(^{2+}\) > Cd\(^{2+}\) > Ag\(^+\). According to the theory of HSAB, hard acids will bind to hard bases, while soft acids will bind to soft bases. Fe(III) metal ion is a hard acid with a hydrated radius of 0.66 Å, metal ions Zn (II) and Cu (II) fall into the borderline (medium) acid group with their respective hydrated radii of 0.74 Å and 0.94 Å. Whereas metal ions Cd(II) and Ag(I) fall into the group of soft acids with hydrated radii of 0.96 Å and 0.98 Å, respectively [26].
The highest selectivity of Fe(III) adsorption is achieved by the mixed-metal solution of Fe(III)/Zn(II). The adsorbent formed has characteristics that are in accordance with the metal ion template [27]. Thus, the adsorbent should only be effective for Fe(III) metal ions separations. Even though Zn(II) has a hydrated radius similar to the radius of the Fe(III), the mold factor of the adsorbent is selective towards Fe(III) ions. Metal ions Cd(II) and Ag(I) are within the soft acid group, but the hydrated radius of Cd(II) is less than the radius of hydrated Ag(I). The selectivity of ion-imprinted adsorbents is strongly influenced by the size of the hydrated radius of the atom [25]. The size of the hydrated radius affects the acidity of the two metals [27] making metal ions Cd(II) have a higher acidity level compared to the metal ion Ag(I).

Therefore, the adsorption selectivity of Fe (III) in the mixed-metal solution of Fe(III)/Cd (II) metal is slightly greater than the selectivity of adsorption in the mixed-metal solution of Fe(III)/Ag(I) metal. The larger the hydrated radius size of the competitor metal, the more Fe(III) metal ions will be blocked from entering the mold obtained from the templates, which will reduce the selectivity of the Fe(III) metal ion to be small. According to the research conducted by Arini on the eugenol derived crosslinking polymer EDMA and Kartikasari on the PEGDE Fe-IIP-PE crosslinker, the selectivity sequence of Fe(III) metal ion acquired based on the order of the hydrated radius of metal ions is as follows Fe(III)/Cr(III) > Fe(III)/Cd(II) > Fe(III)/Pb(II) [9, 28].

The distribution of Fe(III) and Zn(II) ions in IIP-PA and NIP-PA in a mixed-metal solution of Fe(III)/Zn(II) has the highest ratio value. The distribution ratio value (D) depicts the ratio of the total concentration of a metal ion that can be separated through an adsorbent in a mixture of solutions consisting of several types of metal ions [27]. Hence, it could be concluded that the greater the metal ion distribution is, the easier it is to separate a metal over another.

| Metal Ions   | Adsorbent |         |         |
|--------------|-----------|---------|---------|
|              | IIP-PA    | NIP-PA  |         |
|              | D (L/g)   | D (L/g) |         |
| Fe(III)      | Mc        | Fe(III) | Mc      |
| Fe(III)/Zn(II)| 0.692     | 0.018   | 0.531   | 0.022   |
| Fe(III)/Cu(II)| 0.235     | 0.007   | 0.215   | 0.011   |
| Fe(III)/Cd(II)| 0.450     | 0.060   | 0.338   | 0.095   |
| Fe(III)/Ag(I)| 0.567     | 26.786  | 0.457   | 40.433  |

4. Conclusion
Adsorption of Fe(III) metal ions in IIP-PA and NIP-PA follows the pseudo-second-order kinetics and both of the adsorption isotherm follow Langmuir. The adsorption capacity of Fe(III) in IIP-PA is 10.88 mg/g, while the adsorption capacity of NIP-PA is 7.94 mg/g. The mechanism of adsorption of Fe(III) metal ions in IIP-PA and NIP-PA is dominated by hydrogen bonds between the -COOH groups and Fe(III) metal ions. IIP-PA adsorption selectivity coefficient is higher than NIP-PA in a binary solution in the order of Fe(III)/Zn(II) > Fe(III)/Cu(II) > Fe(III)/Cd(II) > Fe(III)/Ag(I).

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