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Gold (Au) Selective Adsorption Using Polyeugenol Based Ionic Imprinted Polymer with Ethylene Glycol Dimethacrylate Crosslink

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Abstract. The Ionic imprinted polymer (IIP) Au (III) adsorbent has been synthesized using polyeugenol as a base polymer with ethylene glycol dimethacrylate (EGDMA) as a crosslinking agent to adsorb Au (III) metal ions from water media. The synthesis procedure includes polymerization, binding of mold ions, crosslinking and mold ions release. As a comparison, the non-imprinted polymer (NIP) material was also synthesized. The polymer yielded was characterized using FTIR, SEM-EDX, and TGA-DTA and then continued with adsorption. Selective adsorption was tested for a mixture of Au (III) binary metal ions with Cd (II), Pb (II) and Fe (III). The adsorption mechanism of Au (III) in all adsorbents is dominant because of the coordination bond between the -OH group and the Au (III) hydrate. Adsorption capacity is expressed as a percentage of IIP-Au (III) of 99.88% over the capacity of other adsorbents; IIP-Au (III) is more selective than NIP with successive selectivity coefficients of Fe (III) 263.03, Cd (II) 53.18, and Pb (II) 6.06.

INTRODUCTION

Gold's unique characteristics have made it applicable generally in various fields including jewelry, electronics, as a catalyst and environment treatment. The development of gold technology is used in the electronics field as a catalyst in various chemical reactions and corrosion-resistant materials. The high demand for gold made gold consumption increase and resulted in gold reserves decline, gold reserves are only about 89 million kg. In this case, it requires the best efforts to recycle gold from electronic waste, to recover gold and reduces environmental pollution [1].

Electronic scrap generally contains Au metal among other metals such as Ag, Pd, Pt, Cu, Fe, Ni, Sn, Zn, and Pb. In recent years, adsorption has been carried out for metal recovery and has been proven to be an effective method as well as to increase adsorbent selectivity for target metals. One technique in making selective adsorbents for target metal ions is the technique of making metal ion molds [2].

Previous research using polyeugenolic based membranes have been done to selectively transport Fe (III) metal ion using polyethylene glycol diglycydil ether as a crosslinker [3]. Research on the effect of template types on polyeugenol to the adsorption selectivity of IIP Fe (III) metal ion has also been done [4].

In this research, ionic imprinted polymer (IIP Au (III)) will be synthesized using polyeugenol as a functional polymer and ethylene glycol dimethacrylate (EGDMA) as a crosslinking agent. The novelty of this research is to apply polyeugenolic based membrane to selectively adsorb and discover a new potential candidate for gold metals recovery.

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EXPERIMENTAL

Materials and Instrumental

The materials used in the research based on pro-analysis from Merck and Sigma Aldrich include, eugenol p.a., anhydrous Na_2SO_4, thiourea, EGDMA (ethylene glycol dimethacrylate), AIBN (2,2', azo-bis-(2-methylpropionitrile), HCl, Methanol, NaOH, chloroform, $BF_3O(C_2H_5)_2$, $AuCl_4$ std 1000 mg/L (Merck), $Pb(NO_3)_2$ std 1000 mg/L (Merck), $Cd(NO_3)_2$ std 1000 mg/L (Merck), Fe $(NO_3)_2$ std 1000 mg/ L (Merck), and aquabidest.

The tools used are laboratory glass equipment, analytical balance (pioneer), hotplate Stirrer (LabTech Co. Ltd.), pH meter (Trans Instrument), reflux equipment, Atomic Absorption Spectroscopy (AAS) (Perkin Elmer Analyst 400), FTIR (Shimadzu Prestige 21), SEM-EDX (Phenom Pro X Desktop with EDX), and TGA (DTA/TG Exstar SII 7300).

Synthesis of Polyeugenol

Eugenol (5.8 grams/0.035 mol) poured into a three-neck flask and 0.25 mL BF_3-diethyl ether was added once every hour while stirred at room temperature. After the reaction lasted for 12-16 hours the polymerization is stopped by adding 1 mL of methanol. The formed gel is then dissolved with chloroform and washed with distilled water to a neutral pH. The solution was dried by adding anhydrous Na_2SO_4. Afterward, it was evaporated at room temperature. The precipitate formed was weighed and analyzed by FTIR. Molecular weight measurement of synthesized polyeugenol was done by Ubbehlohde viscometer to detect whether polymer formed or not.

Synthesis of IIP-Au (III)

Polyeugenol (0.5 grams) were added into a beaker and stirred with a standard Au (III) solution of 500 mg/L for 24 hours. The resulted polyeugenol-Au (III) was taken 0.22 grams to be crosslinked with EGDMA crosslinking agent as much as 0.4 mL, mixed with 1.67 mL chloroform and was added 0.48 mL AIBN initiator. The reaction was carried out within the temperature range of 80-90 °C. The precipitate is then dried and sieved with 100 mesh sieve. Au (III) ions from 1 gram of resin produced were then released using 0.8 M thiourea in 0.1 M HCl for 168 hours and by changing solvent for every 24 hours. It was then neutralized with aquabidest to produce IIP-Au (III). The release of Au (III) ions was analyzed using atomic absorption spectroscopy (AAS).

Synthesis of NIP

The NIP synthesis was carried out in the same manner as IIP-Au (III) without contacted to Au (III) metal ions.

Au (III) Metal Ion Adsorption Test

IIP-Au (III) (50 mg) and NIP adsorbents were contacted with a mixture of 10 ml of the 25 mg/L Au (III) for 24 hours at constant stirring speed. The mixture was filtered with fine filter paper and an AAS analysis was performed on the filtrate to determine the levels of Au (III) adsorbed.

Selectivity Test

Adsorbent (50 mg) is contacted with a mixture of 10 mL binary metal ions with a concentration of 25 mg/L consisting of a mixture of binary metal ions Au (III)/Cd (II), Au (III)/Pb (II), and Au (III)/Fe (III) for 24 hours of contact time at constant stirring speed. The mixture was filtered with fine filter paper and an AAS analysis was carried out on the filtrate to determine the levels of Au (III) and other metals that were adsorbed.

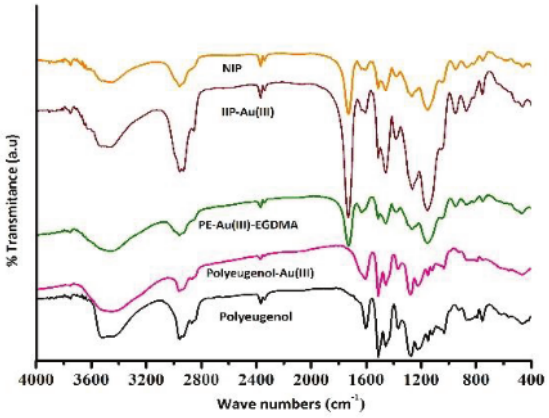
Characterization

Synthesized polymers were characterized by FTIR (Shimadzu Prestige 21), TGA (DTA/TG Exstar SII 7300), and SEM-EDX (Phenom Pro X Desktop with EDX).

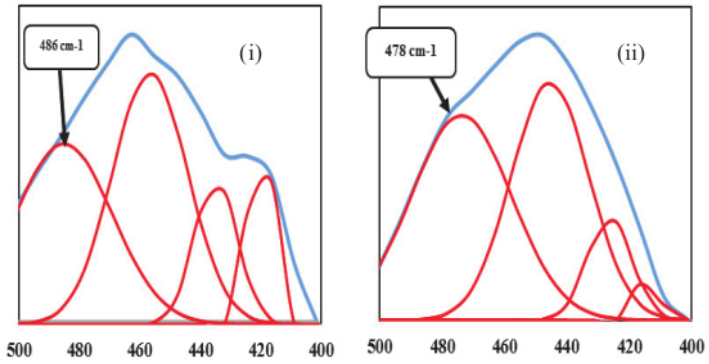
RESULTS AND DISCUSSION

Synthesis of IIP-Au (III)

The formation of polyeugenol was proven by the loss of the allyl group in the absorption of 1637 cm^{-1} and the presence of the peak of the carbonyl group (C=O) in the absorption band 1700 cm^{-1}, which were absent initially. This indicates that the carbonyl group in the crosslinking agent EGDMA was successfully linked to the polymer. The result from molecular weight measurements using Ubbehlohde viscometer was 6127.162 Dalton with polymerization degree (n) of 37. The formation of the polymer could also be proven by the transformation of the appearance from which is initially liquid into a more solid form.



(a)



(b)

FIGURE 1. (a) FTIR IIP-Au (III) and NIP, (b) 500-400 cm^{-1} absorption spectra on label (i) and Polyeugenol-Au (III) on label (ii)

The formation of polyeugenol was proven by the loss of the allyl group in the absorption of 1637 cm^{-1} and the presence of the peak of the carbonyl group (C=O) in the absorption band 1700 cm^{-1}, which were absent initially. This indicates that the carbonyl group in the crosslinking agent EGDMA was successfully linked to the polymer. The result from molecular weight measurements using Ubbehlohde viscometer was 6127.162 Dalton with polymerization degree (n) of 37. The formation of the polymer could also be proven by the transformation of the appearance from which is initially liquid into a more solid form.

There was a shift in the absorption peak from 486 cm^{-1} to 478 cm^{-1}. The emergence of the absorption peak of 478 cm^{-1} characterizes the presence of Au-O. The Au-O absorption peak occurs at 470 cm^{-1}.

The polyeugenol produced was contacted with 500 mg/L Au(III) metal ions and then analyzed using AAS and the adsorbed concentration was 27% compared from the initial (147 mg/L). The washing of metal ion templates was done using thiourea solution acidified in HCl, the process was repeated until there are no Au(III) metal ions left in it [5].

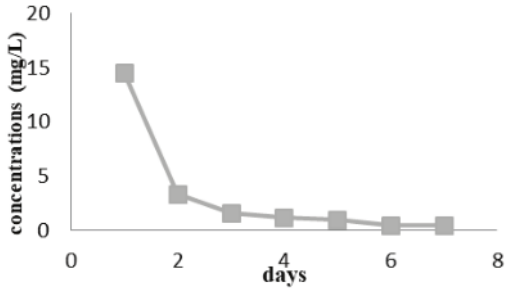
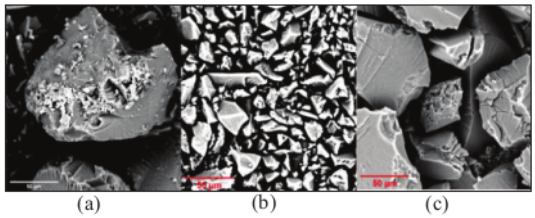


FIGURE 2. Washing of Au(III) ion template for a week

A decrease in Au (III) concentration from the first day to the seventh day showed that the metal ion Au (III) has been bonded with a washing solution so that the metal ion Au (III) has been released from the polymer. The bond between polyeugenol and Au (III) metal ions is not known with certainty, but it can be predicted that it is the coordination interaction through valence bond theory that can form a quadrilateral planar complex as a result of dsp^2 hybridization.

SEM-EDX characterization

The characterization of polyeugenol-Au-EGDMA, IIP-Au(III), and NIP using SEM is shown in Fig. 3. The surface appearance of PE-Au(III)-EGDMA (a) appears to have a less uniform and irregular surface. IIP-Au(III) (b) has a smaller, uniform and regular particle size, compared to NIP (c). The change in morphological form between PE-Au(III)-EGDMA with IIP-Au(III) is likely to occur due to the release of Au(III) metal ions [6]. The purity of the polymer is not tested yet in this research and would be a good point to be analyzed in future research.



(a) (b) (c)

FIGURE 3. Analysis of SEM PE-Au-EGDMA 1500x (a), IIP 1500x (b), NIP 1500x (c) with a scale of 50 µm

TABLE 1. Percentage of Elemental Mass in Polymers

Element	PE-Au-EGDMA	IIP	NIP
	Mass (%)	Mass (%)	Mass (%)
C	38,7	49,77	40
O	5,4	7,34	26
N	21,9	42,89	34
Au	34,1	-	-

TGA-DTA analysis

Thermogravimetric analysis (TGA) in PE-Au(III)-EGDMA, IIP-Au(III) and NIP can be seen in Fig. 4. Thermal testing takes place at the temperature of 50-500 ºC, nitrogen atmosphere flow rate of 20 mL/min, heating rate 10

ºC/minute. PE-Au(III)-EGDMA material experienced decreased of mass during a temperature range of 238-361 ºC, IIP-Au(III) during 272-348 ºC while in NIP is during 233-368 ºC. At a temperature of 100 ºC, it loses weight less than 1% due to the release of water. The subsequent weight loss of 10-70% at a temperature of 300-400 ºC indicates the breakdown of the depolymerization/breakdown of benzene rings that are abundant in the polymer chain [7].

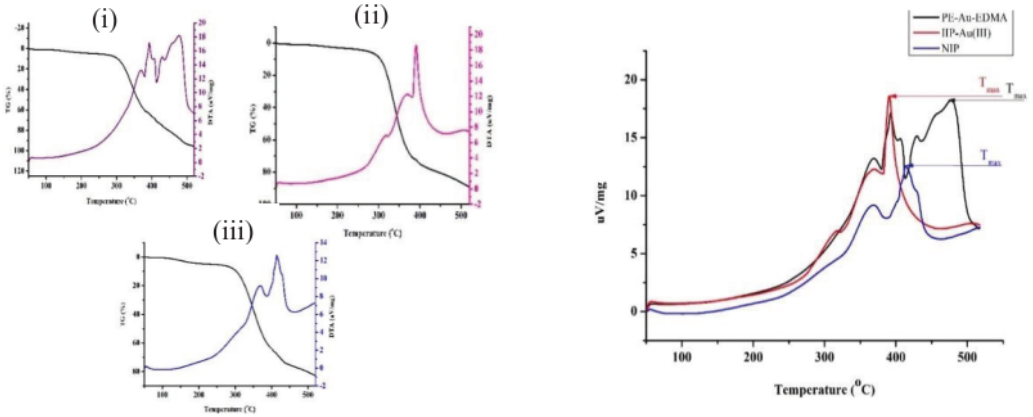


FIGURE 4. TGA-DTA curves of (i) polyeugenol-Au-EGDMA, (ii) IIP-Au(III) and (iii)-NIP, polymer DTA curves at a heating rate of 10ºC / min.

Determination of inflection temperature (T_{max}) using the DTA curve, the change in inflection temperature in the polymer has been presented in Table 2. The inflection temperature of the polymer occurs at the exothermic peak all three DTA curves (thermogram) can be clarified in Fig. 4. The IIP-Au (III) DTA curve shows a sharp weight loss at 390 ºC indicating an empty cavity in the polymer and free from the template. This did not happen with NIP [8]. According to the EDX

TABLE 2. TGA Polymer Profile

Polymer	$T_{5\%}$ (ºC)	$T_{10\%}$(ºC)	$T_{50\%}$(ºC)	T_{max}(ºC)
PE-Au-EGDMA	238	303	361	477
IIP-Au (III)	272	304	348	390
NIP	233	303	368	410

Au (III) Metal Ion Adsorption Capability

IIP-Au (III) and NIP adsorbents were tested with 25 mg/L Au(III) at pH 3 adsorbate to determine the adsorption ability of Au(III) metal ions. Through AAS analysis, it was known that the ability of IIP-Au (III) is higher than NIP, which is clarified in Fig 5. The adsorption efficiency of the adsorbent depends on its surface functional group, which is effective in forming complexes with metal ions. Various functional groups including carboxylates, hydroxyl, sulfates, phosphates, amides, and amino groups have been used for metal adsorption [9].

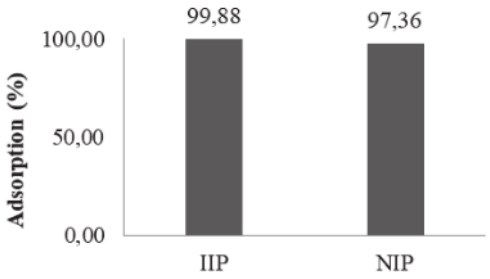


FIGURE 5. IIP-Au(III) and NIP Adsorption Ability

Capability of Au (III) Metal Ion Adsorption in binary Au (III)/ Cd (II)

The adsorption ability test was carried out in a mixture of binary metal Au (III) with Cd (II), Cd (II) was chosen because it has soft acid properties and has a radius of hydrated ions smaller than Au (III).

Figure 6 shows that the ability of Au (III) adsorption is more in IIP-Au (III) compared to NIP and the Cd (II) adsorption ability is more in NIP than in IIP-Au (III). It can be seen that the value of Au (III) adsorption percentage is greater than Cd (II). This can be interpreted that the role of the Au (III) template in IIP works by recognizing its template which is Au (III) so that the IIP-Au (III) were able to be more selective towards Au (III) compared to its competitor metal, Cd(II). The IIP get its recognizing ability through the contacting process with Au (III).

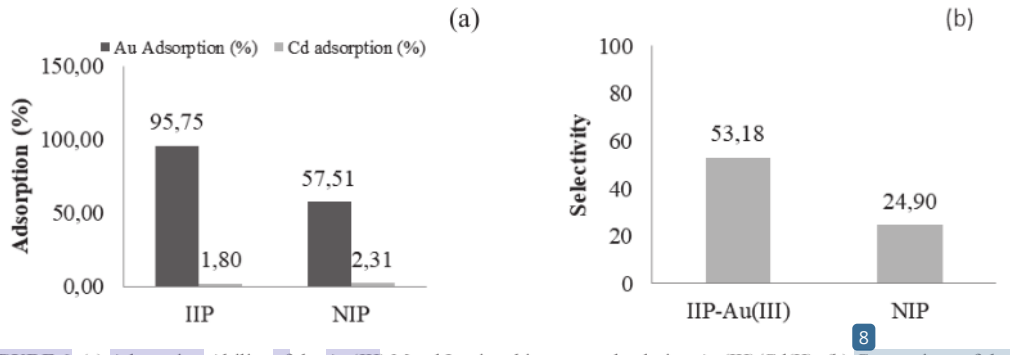


FIGURE 6. (a) Adsorption Ability of the Au(III) Metal Ion in a binary metal solution Au(III)/Cd(II), (b) Comparison of the selectivity of the Au(III) metal ion in the Au(III)/Cd(II) binary metal solution.

Capability of Au (III) Metal Ion Adsorption in binary Au (III)/ Pb (II)

Pb (II) metal was chosen as a competitor because Pb (II) metal has a hydrated ion radius which is smaller than (III), besides Pb (II) metal is included in the borderline acid group in HSAB theory [10]. The ability of adsorption of Au (III) metal ions in a mixture of binary metals with Pb (II) is shown in Fig.7.

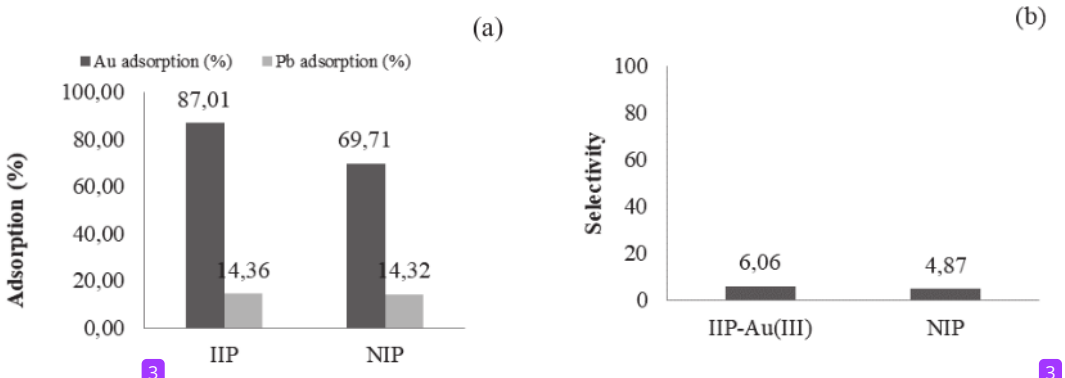


FIGURE 7. (a) The Adsorption Ability of the Au(III) Metal Ion in an Au(III)/Pb(II) binary metal solution, (b) Comparison of the Au(III) metal ion selectivity in the Au(III)/Pb(II) binary metal solution.

The results of the adsorption ability of Au (III) metal ions in a mixture of Au (III)/Pb (II) metal ions can be seen that the percentage value of Au (III) adsorption on IIP-Au (III) is greater than NIP. This is due to the role of template metal ions in IIP-Au (III) whose active groups already have a memory to recognize the target metal ions, in contrast to the NIP which does not have the memory effect resulting in less selectivity [11]. The selectivity difference between IIP Au (III) and Pb (II) was not too visible due to the Pb (II) smaller ion radius compared to Au (III) which is about 0,118 nm [12]. It is also caused by Au (III) small hydrated ion radius which makes Au (III) is more acid than Pb (II), so there is a possibility that Pb (II) will hinder Au (III) metal ions to enter the template.

Capability of Au (III) Metal Ion Adsorption in binary Au (III)/Fe (III)

The choice of Fe (III) metal as a competitor is because it has a hydrated ion radius greater than Au (III) metal and is classified as a strong acid-base in the HSAB classification [10].

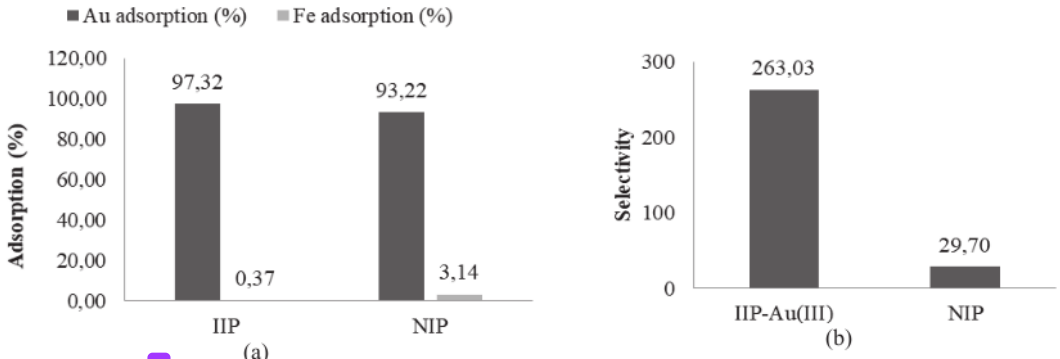


FIGURE 8. (a) The Adsorption Ability of the Au(III) Metal Ion in an Au(III)/Fe(III) binary metal solution, (b) Comparison of the Au(III) metal ion selectivity in the Au(III)/Fe(III) binary metal solution.

The ability of Au (III) metal adsorption on IIP-Au (III) is greater than that of NIP. And the adsorption of Fe (III) is greater in NIP than IIP-Au (III). This happens because of the role of metal templates in IIP-Au (III) whose active group has a memory effect recognizing target metal ions, whereas in NIP it has no memory effect or recognizing ability on the active group so that it absorbs fewer Au (III) metal ions [11].

CONCLUSION

Polyeugenol was successfully synthesized with a yield of 95.37% and a molecular weight of 127.162 Dalton. The ability of IIP-Au (III) adsorption selectivity for Au (III) metal ions is greater than NIP, in the order of Au (III)/Fe (III)> Au (III)/Cd (II)> Au (III)/Pb (II).

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