別紙第１号様式　　No.1

博　士　論　文　の　要　旨

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博士論文題名　Development of the Low Cost Adsorbents from Polysaccharides-based Biomass for the Recovery of Gold

金回収のための多糖類系バイオマスからの低コスト吸着剤の開発

要旨（２，０００字程度にまとめること。）

The wide application of gold to make traditional jewellery as well as in different electronic devices such as computer, mobile phone and other high tech electronic appliances increases its demand every year thus the gold ore are continuously mined to fulfill its demand which will invites the trouble of gold ore exhaustion in near future. The customer are expecting more and more advanced or improve technology thus more sophisticated and technologically advanced electronic devices were invented that replaces the old electronic devices thus large amount of electronic waste is generated every year. In addition, the conventional cyanide treatment for the extraction of gold is toxic that produces large volume of toxic cyanide solution which is not environmentally benign. The invention of new recovery method of gold from primary (gold ore) and secondary (wasted electronic devices) sources with convenient, low cost, and environmentally benign technology is required. From the literature it was found that cellulose is most abundant material in the world which is found mostly in plants. In the present work, new recovery method was invented for the recovery of gold from trace concentration by using chemically modified cellulose derived from agricultural plants, and compared with commercially available cellulose together with some other polysaccharides contains similar functionality such as dextran, alginic acid, and pectic acid. It was found that natural and commercial cellulose containing hydroxyl functional group showed very high affinity and selectively adsorbed gold from trace concentration even in the presence of other coexisting base metals and precious metals. The traditional and industrial values of gold, conventional way of recovering gold their disadvantages are systematically described together with the advantages of novel, economic and environmentally benign technology investigated in this research work in **Chapter 1**.

The novel material for Au(III) recovery was developed from commercial cellulose by cross linking with concentrated sulphuric acid. The cellulose is most abundant material in the world. The aim of this research work is effective utilization of cellulose and its derivatives for the recovery of gold from aqueous medium. The research work was

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conducted with aiming the development of new adsorbent which may be successful to adsorbed and subsequently reduced the Au(III) to its elemental form without adding any types of reducing agents. For that, commercially available cellulose powder was first cross-linked with concentrated sulphuric acid in order to convert the new coordinating sites for Au(III) adsorption. During the treatment, crystalline form of commercial cellulose which have very low affinity with Au(III) ion was converted into amorphous form with very high affinity for Au(III) ion. The trivalent gold was selectively adsorbed onto the cross-linked cellulose gel from the mixture of other precious metals and base metal. It was visually observed that yellow shining metallic particle was formed and floating at the surface of the reactor at the short contact where as it was found to be aggregate then forming heavy particle after long time contact which is settle down on the bottom later. It was further confirmed from the observation of crystalline peaks of elemental gold [reduced gold Au(0)] in XRD spectrum Au(III) loaded cross linked cellulose gel . So, based on the results, we further expect to attempt whether such type of quantitative and selectiveness of gold adsorption is common to all kinds of polysaccharide or not, we employed commercially available polysaccharide such as dextran, alginic acid and pectic acid also hydroxyl as a major functional group by cross-linking with concentrated sulfuric acid similar to the case of cellulose gel. In all the cases the is high selectivity for Au(III) ion from the mixed solution thus the polysaccharide based adsorbent investigated in this study can be expected to be promising material to be employed in large scale industrial processing for gold recovery.

Although commercial cellulose after cross linking yield very effective adsorbent for Au(III), we have tried to extent our technology for the other low cost cellulose rich biomass thus adsorbent was prepared from cotton by the similar method of cross-linking as in Chapter 2 and investigate its adsorption behavior in **Chapter 3**. In **Chapter 4** more abundantly distributed waste paper was utilized for the recovery of gold together with the mixture solution of other precious and base metals from acidic chloride media. Both the materials after cross-linking with concentrated sulfuric acid selectively adsorbed gold with extremely high adsorption capacity. Comparing with cotton, waste paper was much cheaper and abundant. So we are intended to elaborate our technology

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to more abundant cellulose rich material like paper because Japan is the third largest country for paper production in the world thus raw paper for adsorbent preparation was easily available in low cost in Japan in comparison to cotton biomass.

Although we have succeeded to recover the trace concentration of Au(III) from hydrochloric acid media by using cross-linked polysaccharide gels, our final target is to apply our technology in actual industrial processing. The sulphite and cyanide salts of Au(I) are most common chemicals used in gold plating industries where large amount of waste solution containing trace amount of Au(I) is produced. Moreover, for the extraction of gold from its ore or some other secondary sources like waste electronic appliances, cyanide leaching is most effective and common. The gold plating waste solution and cyanide leached liquor of gold ore contains gold in the forms of $Au(CN)_2^-$ or $Au(SO_3)_2^-$, thus recovery of gold from such a solution is required. The gold cyanide is very toxic compared to gold sulphite solution. It was found from the literature that $Au(CN)_2^-$ or $Au(SO_3)_2^-$ possesses chemical similarity thus forms similar types of complex in aqueous solution. From the result of **Chapter 2, Chapter 3 and Chapter 4**, it was found that the cross-linked polysaccharide adsorbents were very much effective for Au(III) recovery in hydrochloric acid media but the industrial gold plating waste solution contains anionic complex of Au(I) sulphite or Au(I) cyanide. So that we further tried to recover mono-valent gold using cross linked cellulose gel from Au(I) sulfite solution in sodium hypochlorite media in **Chapter 5.** Hence, the new way of recovering trace concentration of Au(I) from less toxic $Au(SO_3)_2^-$ solution have been investigated and expected also to be successful to recover Au(I) also from cyanide medium too. By using cross linked pure cellulose (CLPC) gel, the Au(I) was successfully recovered in oxidizing environment in sodium hypochlorite medium. It was believed that Au(I) in $Au(SO_3)_2^-$ solution was oxidized into Au(III) with the aid of NaClO which is effectively adsorbed onto the cellulose gel according to the method described in earlier Chapters. For the application of investigated polysaccharide based adsorbent in actual practice, the real leached liquor of gold and silver was prepared from both primary source (Mongolian gold ore soil sample) and secondary source (scraps of plasma TV monitor) and successfully recovered the gold and silver by leaching with acidic-thiourea followed by its adsorptive

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recovery using cross linked cellulose cotton gel is presented in **Chapter 6** and **Chapter 7,** respectively. Finally, the overall concluding remarks and an outlook are suggested in **Chapter 8**. At last part of this thesis, a list of publications related to author's works, list of presentation and contributions to scientific forum are summarized in appendices.