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Study of the "Oxidation-Complexation" Coordination Composite Ionic Liquid System for Dissolving Precious Metals

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Featured Application: This work can be applied to the recovery of precious metals in WEEE and waste catalyst.

Abstract: The extraction of precious metals is of significant importance for recycling valuable precious metal resources, however, most of the current methods for extraction are inefficient, energy-intensive, or environmentally unfriendly. Herein, we report a new chemical dissolution system for precious metals employing the trichloroisocyanuric acid (TCCA) as an oxidant and tributyl monomethyl ammonium chloride ($N_{4441}Cl$) as a complexant. The leaching yield was achieving 100 wt% for the metals of Au, Pd, Cu, and Ag at 25 °C, and the average dissolution rate of gold was reaching 375 mg/h, 107.1 times higher than that of traditional cyanide method. Based on the analysis of UV-Vis and XPS results, a possible reaction mechanism was proposed: the precious metals were probably oxidized by TCCA, then the formed precious metal ions coordinate with $N_{4441}Cl$ strongly to promote the dissolution process. Applied to gold dissolution from ores, waste electronic and electrical equipment (WEEE) and waste catalysts, the leaching yield of the TCCA-$N_{4441}Cl$ coordinative composite reached 97, 100, and 100 wt%, respectively, demonstrating that this method is not only efficient and environmentally friendly, but also with great adaptability and high potential for real applications.

Keywords: precious metals recovery; ionic liquid; electronic waste; waste catalyst; oxidation-complexation

1. Introduction

Precious metals are widely used in various industrial fields, including modern electronics, medicine and chemical catalysis [1–7]. The high value and depletion of precious metal reserves make the recovery of noble metals from the secondary resources highly desirable [8]. Waste electronic and electrical equipment (WEEE) and the spent catalysts are among the most important raw materials for precious metal recycling due to massive annual production and high element abundance. According to the statistics, the annual production of waste catalysts is about 500,000–700,000 tons [9,10]. The rapid urbanization and change in our lifestyle have resulted in a large and still increasing number of WEEE. By 2021, about 52.2 million tons of WEEE will be generated [11,12]. The recyclable noble metals in these waste materials are much more abundant than the natural ores, for example, the estimated concentration of gold in the WEEE is about 0.15 wt%.

Currently, cyanide leaching is the major method to recycle noble metals from solid wastes, which possesses both simplicity and economic benefits. However, cyanide is extremely toxic to the human body and environment [5,13]. Therefore, new metal leaching systems based on environmental benign reagents, such as thiourea, thiosulphate, and KI/I_2 have been extensively studied [14–18]. However, problems still exist, such as inefficiency, high cost and the requirements of strict operation conditions have limited application of these systems in real circumstances.

Ionic liquids (ILs) have attracted growing interest due to their unique physicochemical properties [19]. ILs are "green" solvents for extraction of precious metal cations and the common extraction mechanisms can be electrostatic interaction, cation exchange, and anion exchange [20–22]. In addition, ILs can form extended hydrogen bonding and have unique interactions both electronically and sterically with several precious metal species, which can stabilize the formation of several precious metal nanoparticles [23,24]. The aforementioned interactions made ionic liquids have the potential to become an ideal metal leaching agent.

Moreover, trichloroisocyanuric acid (TCCA) is a readily available, stable, non-volatile, and inexpensive oxidant [25], which is a content in the majority of disinfectants for swimming pools, medical, and catering fields [26,27].

Therefore, this study reports a novel, efficient, mild, low-toxic precious metals extraction system using a composite ionic liquid combining TCCA and tributyl monomethyl ammonium chloride ($N_{4441}Cl$) together, for the leaching of precious metals from ore, spent catalyst, and WEEE. During the leaching process, TCCA acted as a precious metal oxidant, the oxidized metal ions were then captured by $N_{4441}Cl$ which coordinates well with the metal ions.

2. Materials and Methods

2.1. Materials

$N_{4441}Cl$ at a purity of 99% was purchased from Lanzhou Institute of Physical Chemistry, Chinese Academy of Sciences. TCCA at the purity of 99.5%, gold, palladium, silver, and copper powder, 36%–38% hydrochloric acid (HCl), 68% nitric acid (HNO_3), acetone (CH_3COCH_3), silica (SiO_2), chloroauric acid ($HAuCl_4$), 80% hydrazine hydrate ($N_2H_4{\cdot}H_2O$) were provided by Aladdin. Gold ores were obtained from Suichang Gold Mine located in Suichang, Lishui, Zhejiang, China and they were ground to less than 0.00374 mm (400 mesh) powder before use. CPU chips were acquired from waste CPU Intel XEON^{TM}, and the metal pins were cut off for dissolution.

2.2. Experimental Procedures

2.2.1. Preparation of Au@SiO_2 Catalyst

The impregnation method was used for the preparation of Au@SiO_2 [28]. A total of 10 g silica support and 100 mL deionized water were loaded into a 250 mL round-bottom flask and stirred for 10 min with a speed of 200 r/min at 80 °C. Then, 10 mL of 0.0193 g/mL $HAuCl_4$ solution was then added dropwise into the resulting solution and stirred for 4 h. After that, NaOH solution was used to adjust the pH of the solution to 7 before 10 mL of 80% $N_2H_4{\cdot}H_2O$ was added dropwise into the flask and stirred for another 30 min at 40 °C. After being filtered and washed with deionized water until the filtrate is neutral, the synthesized catalyst was dried under vacuum at 110 °C for 12 h.

2.2.2. Analysis of Metal Content in Gold Ore Powder, Waste CPU Pins, and Au@SiO_2 Catalyst

In order to accurately measure the leaching yield, the analysis of original metal content in the different samples was necessary. For gold ore powder and Au@SiO_2 catalyst, the metal content can be simply detected by X-ray fluorescence (XRF) [29,30]. However, for CPU pins XRF was not applicable since they cannot be processed into powders. CPU pins are composed of Cu cores and Au coating, so by measuring the mass of selectively dissolved Cu in 0.01 g CPU pins, its gold content can be estimated [31]. Experimental details for selective dissolution of gold are as follows: in a 10 mL glass bottle with a lid, 0.01 g of CPU pins were added to 5 mL of 36%–38% HCl and stirred for 1 h with 600 rpm at room temperature. When the reaction was completed, the solution was filtered and analyzed by AAS (Atomic Absorption Spectroscopy).

2.2.3. General Metal Leaching Procedure by TCCA/$N_{4441}Cl$

Typically, in a 10 mL glass bottle with a lid, metal sources (except for Au@SiO_2 catalyst) were added to TCCA/$N_{4441}Cl$ solution with acetone as solvent (TCCA = 3 g (0.16 mol/L), $N_{4441}Cl$ = 0.3 g (1.59 mol/L), acetone = 5 mL), then the reaction was conducted for a certain period of time with constant stirring at 600 rpm. After the reaction, the product was filtered by a 0.22 um filter membrane to obtain the solution after dissolving metal. Finally, AAS was utilized to measure the concentration of gold or other metals in the solution. The leaching yield for each metal was then calculated based on Equation (1).

$$\text{Leaching yield} = \frac{\text{Metal mass in solution measured by AAS}}{\text{Initial metal mass}} \times 100\% \tag{1}$$

2.2.4. Metal Leaching from Catalyst by TCCA/$N_{4441}Cl$

Due to the adsorption of the catalyst carrier, the leaching methods adopted for Au catalysts were slightly different from the general procedure. After being mixed and stirred with TCCA/$N_{4441}Cl$ in acetone (TCCA = 3 g (0.16 mol/L), $N_{4441}Cl$ = 0.3 g (1.59 mol/L), acetone = 5 mL), the mixture was then separated by Soxhlet extraction (acetone as solvent), so that the ionic liquid adsorbed in pores of the catalyst and the metal coordinated to the ionic liquid can be washed off from the catalyst powder. Finally, AAS was utilized to measure the concentration of metal in the mixed solution composed of the leaching solution and Soxhlet extraction solution. The leaching yield was then calculated based on Equation (1).

2.3. *Characterization and Method*

AAS (Atomic Absorption Spectroscopy) was obtained by air acetylene method using TAS-990 atomic absorption spectrometer (PUXI, China). A standard curve was determined by a series of corresponding metal standard solutions. The samples were diluted with deionized water and digested with 68% nitric acid before being examined by AAS. The concentration and metal mass were determined using the standard curve.

X-ray fluorescence (XRF) test was carried out on ARLADVANT'S X-4200 X-ray fluorescence spectrometer (ThermoFisher, American). Before the test, the samples were dried at 105 °C for more than 2 h and pressed into round-shaped discs by a tablet press machine. Finally, the software UNIQUANT was used for data analysis.

X-ray diffraction (XRD) patterns were measured by an X'Pert PRO X-ray diffractometer (PANalytical, Netherlands) using a Cu Kα radiation with an X-ray source (λ = 0.154056 nm) at 40 kV.

X-ray photoelectron spectra (XPS) were acquired with a Kratos AXIS Ultra DLD spectrometer (KRATOS, Japan). XPS analysis was performed with the monochromatized aluminum X-ray source, and the pass energy of the electron analyzer was 15 eV. The pressure in the sample analysis chamber was lower than 6×10^{-9} Torr during data acquisition. Binding energies were referred to the C1s line at 284.8 eV.

Ultraviolet-visible spectrophotometry (UV-vis) was performed at room temperature on Cary-100 Ultraviolet-visible spectrophotometer (VARIAN, American). With acetone as reference, the absorption spectrum of the sample in the range of 200–800 nm was determined after the background of the solvent was removed.

3. Results and Discussion

3.1. Metal Dissolving Effect of TCCA/$N_{4441}Cl$

The TCCA-ILs composite solution showed a good solubility for Au^0, Pd^0, Cu^0, Ag^0 at room temperature (25 °C, $N_{4441}Cl$ = 3 g, TCCA = 0.3 g, acetone = 5 mL, metal = 0.01 g). The solutions after dissolution appeared to be clear and transparent (Figure 1). The AAS results showed 100 wt% leaching yield for all the metals examined (Table 1). In addition, the UV-Vis spectrum of the dissolved gold samples (Figure 2) showed that there are no absorption bands above 500 nm where the absorption peaks of Au^0 are supposed to be located [32], which indicated the absence of Au^0 (dispersed Au particles) in the samples. There is only one 328 nm absorption peak in the dissolved gold sample. In order to determine the existence form of the gold species in the absorption peak, a comparative experiment was carried out. It was found that the UV-Vis spectrum obtained by directly mixing $HAuCl_4$ with $N_{4441}Cl$ only has 328 nm absorption peak as well. Combined with the literature of Usher et al. [33], it was speculated that the dissolved gold exists in the form of Au(III). Solvent-free leaching was also tested. Although the leaching yield was also high, the viscosity of $N_{4441}Cl$ had a negative influence on the mass transfer, which reduced the dissolving rate. Heating is necessary to obtain similar performance. While, the protonic solvent such as the water or alcohol will induce the formation of cyanuric acid (the product of TCCA reaction), causing the separation problem of subsequent deposited precious metal powder.

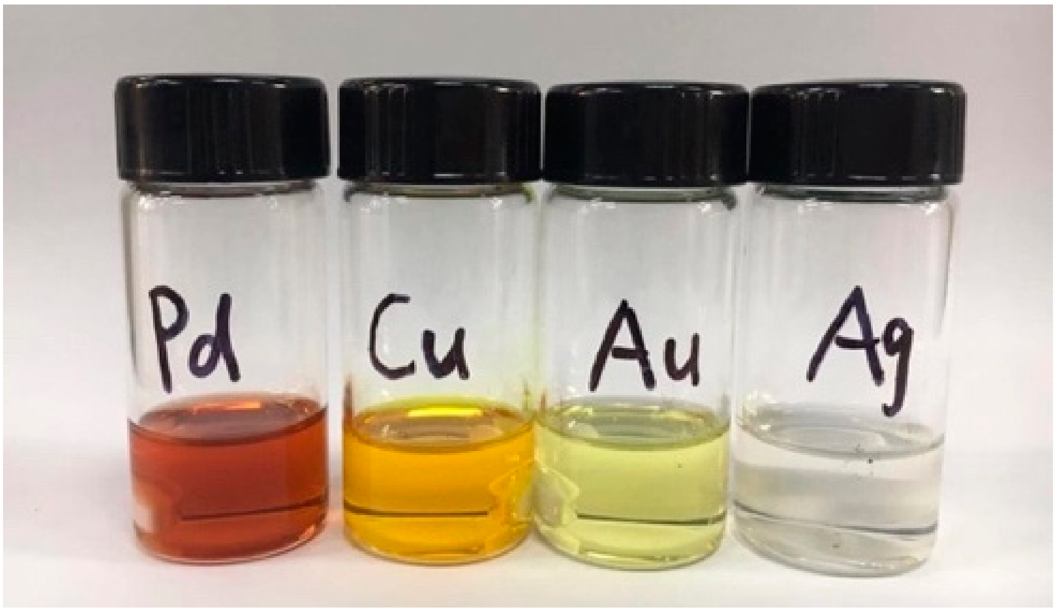


Figure 1. Metals dissolved by trichloroisocyanuric acid (TCCA)/$N_{4441}Cl$ (the dissolved metals are Pd, Cu, Au, Ag from left to right, respectively).

Table 1. AAS (Atomic Absorption Spectroscopy) results of TCCA/$N_{4441}Cl$ to dissolve Au^0, Pd^0, Cu^0, Ag^0.

Metal	Metal Mass/g	AAS Result/g	Leaching Yield/wt%
Au	0.01	0.01	100
Pd	0.01	0.01	100
Cu	0.01	0.01	100
Ag	0.01	0.01	100

Reaction conditions: 25 °C, 10 min, $N_{4441}Cl$ = 3 g, TCCA = 0.3 g, acetone = 5 mL.

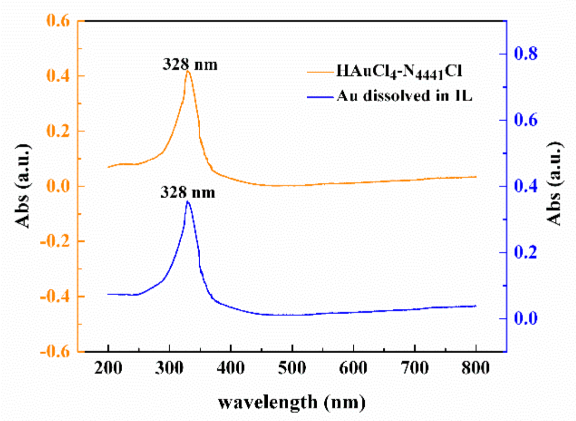
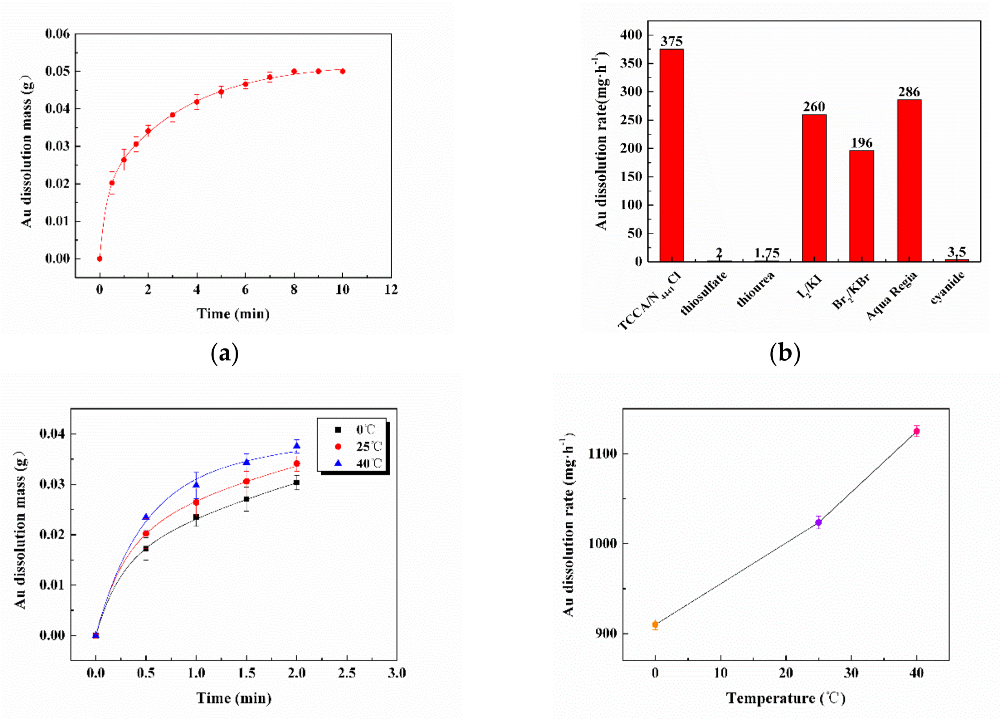


Figure 2. UV-Vis result of the ionic liquid system after dissolving gold and the mixture of $N_{4441}Cl$ and $HAuCl_4$.

3.2. The Rate of Dissolving Gold

As shown in Figure 3a, the dissolving rate of gold in TCCA/$N_{4441}Cl$ was rapid. An average 375 mg/h dissolving rate was obtained in the first 8 min. The kinetic model based on this data is shown in Figure S1. Additionally, the deceleration of the reaction rate can be explained with the decrease of exposed gold surface (resulting in the decrease of the amount of gold that can participate in the reaction) and the consumption of TCCA in the reaction. In addition, the equilibrium solubility of the ionic liquid system was as shown in the supporting information.



(**a**) (**b**)

(**c**) (**d**)

Figure 3. (**a**) The dissolution rate curve of Au by TCCA/$N_{4441}Cl$ at 25 °C; (**b**) comparison of average dissolution rates at 25 °C obtained with the literature [34] (the reagents of the methods mentioned are TCCA 0.16 M, $N_{4441}Cl$ 1.59 M; $(NH_4)_2S_2O_3$ 0.3 M, NH_3 0.3 M, $CuSO_4$ 0.01 M; CH_4N_2S 0.263 M, $Fe_2(SO_4)_3$ 0.02 M, H_2SO_4 0.15 M; I_2 0.08 M, KI 0.80 M; Br_2 0.20 M, KBr 0.67 M; Aqua Regia (HCl:HNO_3 = 3:1); KCN 0.14 M, respectively); (**c**) the dissolution rate curve of Au by TCCA/$N_{4441}Cl$ at different temperatures; (**d**) correlation diagram between the rate of Au dissolution and temperature.

Even though the dissolving rate dropped as the reaction proceeded, the average rate was still higher than those of the other systems reported previously. According to the literature [34], the average dissolution rate in 8 h of thiosulfate, thiourea, I_2/KI, Br_2/KBr, Aqua Regia and cyanide are 2, 1.75, 260, 196, 286, 3.5 $mg \cdot h^{-1}$, respectively, at 25 °C. Obviously, TCCA/N_{4441}Cl has the fastest gold dissolution rate, which is 107.1 times and 1.3 times higher than the aqua regia and cyanide (Figure 3b).

In addition, the dissolution rates of gold at different temperatures were measured. At 0, 25, and 40 °C, the dissolution rates of gold in the first 2 min were 910.2, 1023.6, and 1126.8 mg/h, respectively, thus the reaction temperature was positively correlated to the dissolution rate of gold (Figure 3c,d).

3.3. Reaction Mechanism

In order to further explore the structure of dissolved Au in the ionic liquid, XPS was used to characterize the samples. The binding energy of Au $4f_{7/2}$ in the resultant sample was 84.3 eV, while that of the mixture of N_{4441}Cl and $HAuCl_4$ was 84.7 eV (Figure 4), which was close to the sample. Therefore, the dissolved gold in the sample was in the form of $[(C_4H_9)_3CH_3N][AuCl_4]$ ion pair.

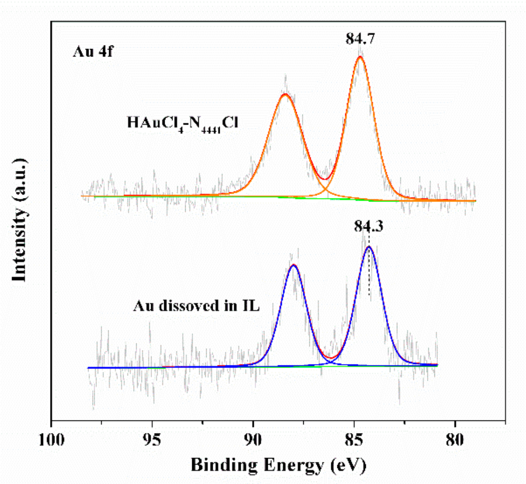
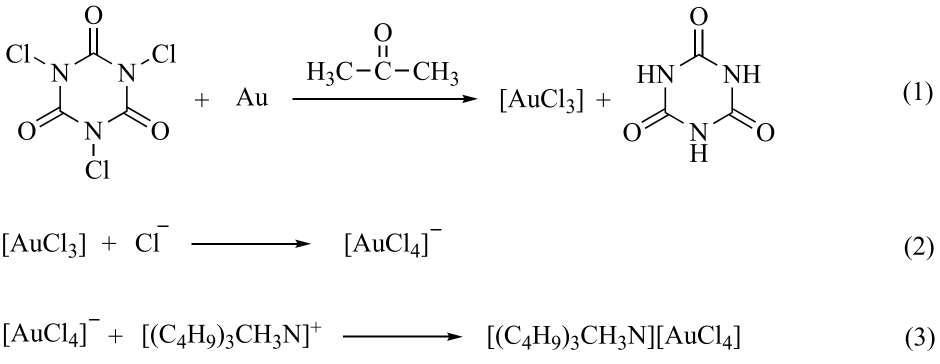


Figure 4. XPS result of the ionic liquid system after dissolving gold and the mixture of N_{4441}Cl and $HAuCl_4$.

According to the above characterization results, the possible reaction mechanism can be inferred as Scheme 1 and Figure 5. Metallic gold (Au^0) on the surface of gold sources is oxidized by TCCA at Step (1) and the resultant $[AuCl_3]$ then coordinates with Cl^- in IL to form $AuCl_4^-$ at Step (2), after that, a more stable ion pair is formed with N_{4441}^+ through electrostatic attraction, hydrogen bond and other forces at Step (3), and finally diffuses into the bulk phase of ionic liquids.



$$[AuCl_3] + Cl^- \longrightarrow [AuCl_4]^- \qquad (2)$$

$$[AuCl_4]^- + [(C_4H_9)_3CH_3N]^+ \longrightarrow [(C_4H_9)_3CH_3N][AuCl_4] \qquad (3)$$

Scheme 1. Mechanism of dissolution of gold by TCCA/N_{4441}Cl.

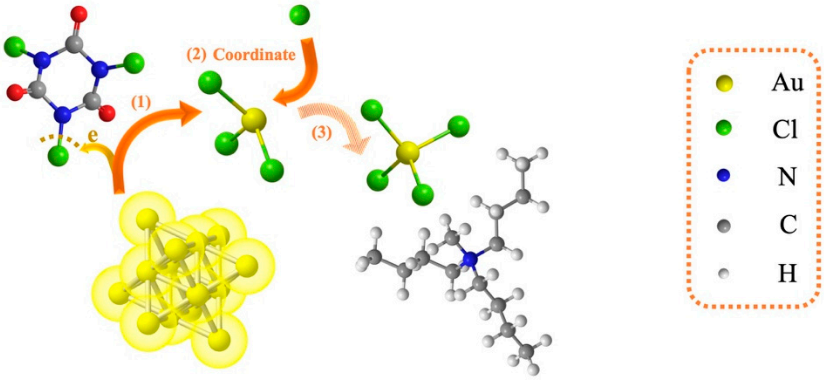


Figure 5. Reaction mechanism diagram.

Based on the proposed mechanism, both TCCA and $N_{4441}Cl$ were indispensable for the high leaching yield. Control experiments using pure TCCA and pure $N_{4441}Cl$ both showed negligible leaching yield (1.3 and 0 wt%), while for the TCCA/$N_{4441}Cl$ mixture, the leaching yield could reach 100 wt% (Figure 6). This comparative study further confirms that the synergistic effect of TCCA and $N_{4441}Cl$ in the composite system is necessary for effective metal dissolving.

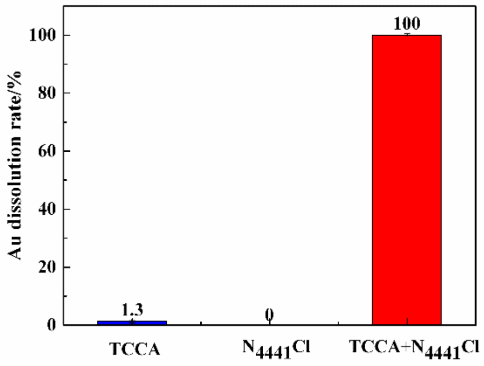


Figure 6. Difference of gold dissolving rate of single reagent and combined TCCA and $N_{4441}Cl$.

The optimal ratio of $N_{4441}Cl$:TCCA:Au for the most effective gold leaching was investigated as well. The effect of the amount of ionic liquid was not investigated since it was already added in large excess, thus the concentration of ligands was much higher than that of Au^{3+} during the coordination process. Therefore, the rate of the coordination reaction was hence believed to depend solely on the concentration of Au^{3+} generated from oxidation of metallic Au. As shown in Figure 7, when the mass ratio of $N_{4441}Cl$:TCCA:Au was set at 300:10:1, 300:15:1, 300:30:1, 300:60:1, the average dissolution rates were 81.6, 276.6, 375, 414 mg/h, respectively. The dissolution rate of Au accelerated rapidly with the increasing TCCA amount when the TCCA concentration was low, but gradually flattened out with the further increasing TCCA:Au ratio. This observation was rationalized to the saturation of adsorbed TCCA on the surface of Au particles. When the concentration of oxidant is low, the rate of reaction would respond sensitively to the change in the concentration of oxidant since there are still plenty of empty sites for adsorption. While at higher oxidant concentration, the number of Au atoms exposed becomes limited. After all the Au sites obtained saturated adsorption, even if the concentration of TCCA in the liquid phase increased, the dissolution rate of Au would depend only on the number of surface Au atoms. Therefore, $N_{4441}Cl$:TCCA:Au = 300:30:1 is the appropriate mass ratio.

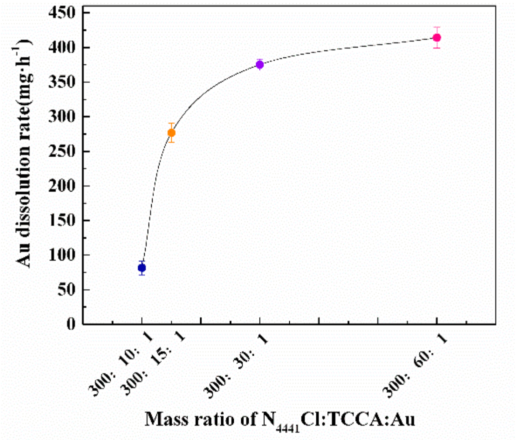


Figure 7. Effect of $N_{4441}Cl$ and TCCA ratio on dissolution rate.

4. Application of Precious Metal Extraction

In order to further evaluate the potential of the reported extraction system in the practical application circumstance, its extraction efficiency was further assessed for ores, WEEE, and spent catalysts.

4.1. Extraction from Ores

The XRF spectroscopy (Table S1) indicated that a variety of metal elements were present in the gold ores, including gold (0.0044 wt%), iron (2.07 wt%), and copper (0.413 wt%). The maximum Au leaching yield obtained using the TCCA/$N_{4441}Cl$ leaching system was approximately 97 wt%. Based on the experimentally obtained yield, theoretically, 1 ton of normal-grade gold ore would afford about 40 g of gold. Though the concentrations of Fe in the gold ores were 470 times higher than that of Au, only 27.3 wt% of iron was dissolved by TCCA/$N_{4441}Cl$ (Figure 8), indicating that the TCCA/$N_{4441}Cl$ leaching system exhibits a satisfying leaching selectivity for Au which would be helpful to simplify the subsequent purification procedure during the metal refinery procedure.

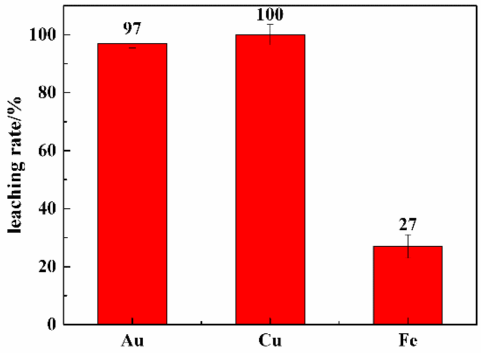
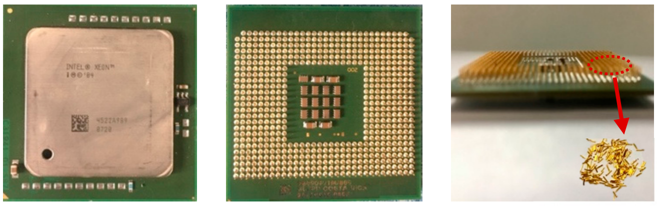


Figure 8. AAS results of TCCA/$N_{4441}Cl$ to dissolve gold ore.

4.2. Extraction from WEEE

As illustrated in Figure 9, the metal pins on the CPU chip were cut off before leaching. AAS results indicated that the dissolved Cu accounts for 93.6 wt% of the metal pins and no Au was detected in the solution, hence the undissolved part, which is 6.4 wt%, can be considered as the quantity of gold. At 25 °C, 0.01 g CPU could be completely dissolved by 3 g $N_{4441}Cl$ and 0.3 g TCCA in 5 mL acetone within 10 min, the leaching yield of Au and Cu was 100 wt% as revealed by AAS results.



(**a**) (**b**) (**c**)

Figure 9. The waste CPU utilized for the gold leaching: (**a**) backside; (**b**) front side; (**c**) CPU chips. Reaction condition: 0.01 g CPU pins dissolved in 3 g $N_{4441}Cl$ and 0.3 g TCCA in 5 mL acetone within 10 min.

4.3. Extraction from Spent Catalyst

As shown in Figure 10, the leaching efficiency of TCCA/$N_{4441}Cl$ for the waste catalysts was also high. The gold content of the prepared catalyst was 0.983 wt% calculated from XRF analysis (Table S2). After stirring 1 g Au@SiO_2 in 0.3 g TCCA, 3 g $N_{4441}Cl$ and 5 mL acetone for 10 min at 25 °C, Soxhlet extraction was carried out for 24 h, and finally the leached catalyst was obtained. XRF analysis result showed that the gold content remained in the catalyst powder was 0 wt%, which indicated that the extraction yield of Au was theoretically 100 wt%. Furthermore, the catalyst support after leaching and extraction can be used to prepare new catalysts.

(**a**) (**b**) (**c**)

Figure 10. (**a**) Au@SiO_2 leaching process; (**b**) Au@SiO_2 before leaching; (**c**) Au@SiO_2 after leaching.

5. Conclusions

In this study, by using the TCCA and $N_{4441}Cl$ composite ionic liquid, a 100 wt% leaching yield of precious metals and the average gold dissolution rate of 375 mg/h was obtained at room temperature, which is 107.1 times higher than that of the traditional cyanide method. Moreover, the method was able to be applied in the recovery of precious metals from ores, WEEE, and spent catalysts. Compared with the traditional recovery method, this method is more efficient, simple, and environmentally friendly and showed significant adaptability for the practical applications.

6. Patents

According to the research results of this paper, two Chinese invention patents (publication numbers: CN201910796215.1 and CN201910797057.1) have been applied for.

Supplementary Materials: The following are available online at http://www.mdpi.com/2076-3417/10/10/3625/s1, Table S1: The XRF of gold ores, Table S2: The XRF of Au@SiO_2: (**a**) before leaching; (**b**) after leaching, Figure S1: (**a**) The dissolution rate curve of Au by TCCA/$N_{4441}Cl$ at 25 °C; (**b**) fitting diagram of kinetic model (x = leaching yield of gold).

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Conflicts of Interest: The authors declare no conflict of interest.

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