

**THE DISSOLUTION OF GOLD COLLOIDS IN  
AQUEOUS THIOSULFATE SOLUTIONS**

**By**

**Xinmin Zhang**

**B. Eng. (Material Sci.) East China University of Technology,  
Shanghai, China**

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*I declare that this thesis is my own account of my research and  
contains as its main content work  
which has not previously been submitted for a degree  
at any tertiary education institute*

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Xinmin Zhang

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## ABSTRACT

The kinetics of the dissolution of gold and silver colloids in ammoniacal thiosulfate solutions has been studied using oxygen, copper(II) or oxygenated copper(II) as oxidants at pH 9 - 11 and temperature 22°C to 48°C. The effects of the concentration of the main reagents such as copper(II), ammonia and thiosulfate as well as various background reagents have been investigated. Gold and silver colloids have characteristic absorption peaks at 530 nm and 620 nm respectively. Thus, the extent of gold or silver dissolution in different lixiviant systems was monitored using an ultraviolet-visible spectrophotometer. A comparison of the behaviour of gold colloids and powders has also been made. The beneficial or detrimental effects of silver colloid, and background reagents such as silver nitrate, and sodium salts of nitrate, carbonate, sulfite, sulfate, trithionate, tetrathionate anions have also been investigated.

Experimental results show that the relative rates and the extent of gold colloid dissolution at 25°C in different lixiviant systems in a given time interval are in the order: oxygen-cyanide > copper(II)-ammonia-thiosulfate  $\approx$  oxygen-copper(II)-ammonia-thiosulfate > oxygen-ammonia-thiosulfate  $\geq$  oxygen-ammonia > copper(II)-ammonia. The analysis of electrode potentials shows that  $\text{Au}(\text{S}_2\text{O}_3)_2^{3-}$  is the predominant gold(I) species in the lixiviant solutions containing oxygen or copper(II) as oxidant and thiosulfate or mixed ammonia-thiosulfate as ligands. During the reaction of copper(II) with thiosulfate in ammoniacal solution without oxygen, the measured potential using a platinum electrode represent the redox couple  $\text{Cu}(\text{NH}_3)_n^{2+}/\text{Cu}(\text{S}_2\text{O}_3)_m^{1-2m}$  ( $n = 4$  or  $3$ ,  $m = 3$  or  $2$ ) depending on the concentrations of thiosulfate and ammonia.

The initial dissolution rates of gold colloid by oxygen in copper-free solutions show a reaction order of 0.28 with respect to the concentration of dissolved oxygen, but independent of the concentration of ammonia and thiosulfate. The reaction activation energy of 25 kJ/mol in the temperature range 25°C to 48°C indicated a diffusion controlled reaction.

The initial dissolution rates of gold colloid by oxidation with copper(II) in oxygen-free solutions show reaction orders of 0.41, 0.49, 0.60, 0.15 and 0.20 with respect to the concentrations of copper(II), thiosulfate, ammonia, chloride and silver respectively. The presence of silver(I) or chloride ions enhances the rate of gold dissolution, indicating their involvement in the surface reaction, possibly by interfering with or preventing a passivating sulfur rich film on gold surface. An activation energy of 40-50 kJ/mol for the dissolution of gold by oxidation with copper(II) in the temperature range 22°C to 48°C suggests a mixed chemically/diffusion controlled reaction. The dissolution of gold by oxidation with copper(II) in oxygen-free solutions appears to be a result of the reaction between gold, thiosulfate ions and the mixed complex  $\text{Cu}(\text{NH}_3)_p(\text{S}_2\text{O}_3)^0$ . The half order reactions support electrochemical mechanisms in some cases.

The initial dissolution rates of gold colloid, massive gold and gold-silver alloys by oxygenated copper(II) solutions also suggest a reaction that is first order with respect to copper(II) concentration. High oxygen concentration in solutions has a negative effect on the initial rate of gold dissolution and overall percentage of gold dissolution, indicating that oxygen affects the copper(II), copper(I) or sulfur species which in turn affects the gold dissolution. The surface reaction produces

$\text{Au}(\text{NH}_3)(\text{S}_2\text{O}_3)^-$  and  $\text{Cu}(\text{NH}_3)_p^+$ . The mixed complexes  $\text{Au}(\text{NH}_3)(\text{S}_2\text{O}_3)^-$  and  $\text{Cu}(\text{NH}_3)_p^+$  re-equilibrate to the more stable complexes  $\text{Au}(\text{S}_2\text{O}_3)_2^{3-}$  and  $\text{Cu}(\text{S}_2\text{O}_3)_3^{5-}$  in solution.

The dissolution of gold powder by oxidation with copper(II) in oxygen-free solutions shows the same trends as that of gold colloid. The presence of silver(I) or chloride ions enhances the initial rate and percentage dissolution of gold colloid and powder. The dissolution kinetics of gold powder and colloid follow a shrinking sphere kinetic model in solutions of relatively low concentrations of thiosulfate and ammonia, with apparent rate constants being inversely proportional to particle radius.

The best system for dissolving gold based on the results of this work is the copper(II)-ammonia-thiosulfate solution in the absence of oxygen or in the presence of oxygen. In the absence of oxygen, copper(II) 1.5-4.5 mM, thiosulfate 20-50 mM, ammonia 120-300 mM and pH 9.3-10 are the best conditions. The presences of carbonate and sulfite have a significant negative effect on the dissolution of gold. The presence of sodium trithionate shows a beneficial effect in the first two hours, while sodium tetrathionate or lead nitrate have a small negative effect and sodium nitrate showed no effect on the dissolution of gold. Silver nitrate and sodium chloride also show beneficial effects. In the presence of oxygen, copper(II) 2.0-3.0 mM, thiosulfate 50 mM, ammonia 240 mM and pH 9.3-9.5 are the best conditions.

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## TABLE OF CONTENTS

<b>Abstract</b>	<b>iii</b>
<b>Acknowledgements</b>	<b>vi</b>
<b>Publications</b>	<b>vii</b>
<b>Table of Contents</b>	<b>viii</b>
<b>List of Figures</b>	<b>xiv</b>
<b>List of Tables</b>	<b>xxvii</b>
<b>Chapter 1 Introduction</b>	<b>1</b>
1.1 Gold cyanidation	1
1.2 Non cyanide lixivants	2
1.2.1 Potential lixivants	2
1.2.2 Stability constants of gold(I)/(III) complex species	3
1.3 Thiosulfate as an alternative lixiviant	7
1.3.1 Advantages	8
1.3.2 Disadvantages	8
1.3.3 Complexity of gold leaching process with thiosulfate	9
1.4 The use of gold colloids for kinetic studies	11
1.5 Objectives	14
<b>Chapter 2 Literature review</b>	<b>15</b>
2.1 History and recent progress on gold extraction using thiosulfate	15
2.2 Gold-copper-sulfur species and electrode potentials	17
2.2.1 Gold-copper species	17
2.2.2 Sulfur species	23
2.3 Production, oxidation and stabilization of thiosulfate	25
2.3.1 Production of thiosulfate	25
2.3.2 Oxidation of thiosulfate by oxygen	27



2.3.3	Oxidation of thiosulfate by copper(II)	28
2.3.4	Oxidation of thiosulfate by oxygenated copper(II)	30
2.3.5	Stabilization of thiosulfate	33
2.4	Leaching of gold	36
2.4.1	Oxide ore	39
2.4.2	Sulfide ore	40
2.4.3	Carbonaceous ore	41
2.4.4	Refractory ore	44
2.4.5	Pure gold	45
2.5	Factors affecting the leaching of gold	48
2.5.1	Effect of thiosulfate concentration	48
2.5.2	Effect of copper concentration	53
2.5.3	Effect of ammonia concentration	55
2.5.4	Effect of pH	59
2.5.5	Effect of temperature	60
2.5.6	Effect of sulfur containing anions	65
2.5.7	Effect of residence time	68
2.5.8	Effect of foreign ions	70
2.5.9	Effect of oxidants	71
2.6	The mechanism of gold dissolution in a $\text{Cu(II)-NH}_3\text{-S}_2\text{O}_3^{2-}$ system	73
2.7	Aims of present study	77
<b>Chapter 3 Experimental</b>		<b>79</b>
3.1	Materials and reagents	79
3.2	Preparation of gold colloid	80
3.2.1	Unstabilized gold colloid	80
3.2.2	Stabilized gold colloid	81
3.2.3	Silver colloid	82
3.3	Determination of particle size	83
3.4	Reaction vessels and experimental measurements	85
3.4.1	Reaction vessels	85
3.4.2	Temperature and pH	88
3.4.3	Oxygen concentration	88

3.4.4 Redox potentials	88
3.4.5 Nitrogen flow rate	89
3.4.6 Thiosulfate concentration	90
3.4.7 Sampling	91
3.5 UV-visible spectrum of gold, silver and copper	91
3.5.1 Measurement of absorbance	91
3.5.2 Comparison of UV-visible spectrum of Au(c), Au(I), Cu(II) and Cu(I)	92
3.5.3 Comparison between unstabilized and stabilized gold colloid	94
3.5.4 Beer-Lambert Law	97
3.5.5 Correction for background absorbance due to copper(II)	99
3.6 Experimental procedure for rate studies	103
3.6.1 Gold colloid-oxygen-cyanide system	103
3.6.2 Gold colloid-oxygen-thiosulfate-ammonia system	103
3.6.3 Gold colloid-copper(II)-thiosulfate-ammonia system	104
3.6.4 Gold colloid-oxygen-copper(II)-thiosulfate-ammonia system	105
3.6.5 Gold powder-copper(II)-thiosulfate-ammonia system	105
3.7 Summary of experimental conditions	106
<b>Chapter 4 Results</b>	<b>110</b>
4.1 Introduction	110
4.2 Dissolution of gold colloids in cyanide solutions	110
4.3 Dissolution of gold in thiosulfate solutions	112
4.3.1 Effect of oxygen concentration	112
4.3.2 Effect of thiosulfate concentration	114
4.3.3 Effect of ammonia concentration	115
4.3.4 Effect of chloride concentration	116
4.3.5 Effect of temperature	117
4.4 Dissolution of gold by copper(II)-ammonia-thiosulfate under nitrogen	118
4.4.1 Gold and platinum electrode potentials	118
4.4.2 Effect of concentration of gold colloid	119
4.4.3 Effect of particle size of gold colloid	119
4.4.4 Effect of ionic strength	120

4.4.5	Effect of copper(II) concentration	121
4.4.6	Effect of thiosulfate concentration	123
4.4.7	Effect of ammonia concentration	124
4.4.8	Effect of pH	126
4.4.9	Effect of concentrations of sodium nitrate and silver nitrate	127
4.4.10	Effect of silver colloid	128
4.4.11	Effect of lead nitrate	130
4.4.12	Effect of sodium chloride	131
4.4.13	Effect of carbonate, trithionate, tetrathionate, and sulfite	132
4.4.14	Effect of temperature	134
4.5	Dissolution of gold in thiosulfate solutions by oxygenated copper(II)	135
4.5.1	Effect of oxygen concentration	135
4.5.2	Effect of copper(II) concentration	138
4.5.3	Effect of thiosulfate concentration	139
4.6	Dissolution of gold powder in thiosulfate solutions under nitrogen	141
4.6.1	Effect of copper(II) concentration	141
4.6.2	Effect of silver and chloride ions	143
4.6.3	Effect of particle size of gold powder	144
<b>Chapter 5 Electrode potentials and gold-copper speciation</b>		<b>145</b>
5.1	Introduction	145
5.2	Relationship between electrode potentials, rates and speciation	147
5.3	Platinum electrode potentials during reaction	152
5.4	Gold electrode potentials	157
5.4.1	Effect of oxygen and copper(II)	157
5.4.2	Effect of ligand concentrations	159
	(a) Ammoniacal-thiosulfate solution	159
	(b) Thiosulfate-free ammonia solution	160
5.4.3	Effect of background salts	163
5.5	Summary and conclusions	165

<b>Chapter 6 Initial rates of gold dissolution</b>	<b>167</b>
6.1 Introduction	167
6.2 Gold dissolution in different lixiviant systems	169
6.3 Oxidation of thiosulfate by oxygen or copper(II)	172
6.4 Determination of initial rate	175
6.4.1 Thiosulfate media	175
6.4.2 Cyanide media	181
6.5 Initial rates of gold, silver and gold-silver alloys in thiosulfate solutions	183
6.6 Comparison with literature data	187
6.7 Effect of temperature on initial rate	190
6.8 Dissolution of gold in the absence of copper(II)	192
6.8.1 Effect of oxygen concentration	192
6.8.2 Effect of ligand concentration	193
6.8.3 Effect of chloride concentration	197
6.9 Dissolution of gold in anaerobic copper(II) solutions	199
6.9.1 Effect of reagent concentration and pH	199
6.9.2 Effects of background salts	202
6.10 Dissolution of gold by oxygenated copper(II) solutions	206
6.10.1 The role of oxygen	206
6.10.2 Effect of initial copper(II) concentration	208
6.10.3 Effect of thiosulfate concentration	209
6.10.4 Effect of oxygen concentration	210
6.11 Effect of concentration ratio of $[\text{NH}_3] / [\text{Na}_2\text{S}_2\text{O}_3]$	214
6.11.1 Non-oxygenated systems	214
6.11.2 Oxygenated systems	219
6.12 Summary and conclusions	221
<b>Chapter 7 Kinetic models for gold dissolution</b>	<b>223</b>
7.1 Introduction	223
7.2 Reaction orders based on initial rates	224
7.3 Rate controlling step for the dissolution of gold by copper(II) in oxygenated solution	231

7.4 Electrochemical model for the dissolution of gold by copper(II) in anaerobic solutions	236
7.5 Shrinking particle kinetic models	241
7.6 Shrinking sphere model in copper(II)-ammonia-thiosulfate media	244
7.6.1 Conditions and limitations	244
7.6.2 Effect of particle size and copper(II) concentration	246
7.6.3 Effect of oxygenation	249
7.6.4 Effect of tetrathionate and trithionate	251
7.6.5 Effect of lead(II), silver(I), chloride and ammonia	255
7.7 Summary and conclusions	264
<b>Chapter 8 Summary and recommendations</b>	<b>266</b>
8.1 Summary	266
8.2 Recommendations for further work	270
<b>Reference</b>	<b>271</b>
<b>Appendix</b>	<b>287</b>
Appendix A1 Comparison between measured and calculated absorbance for the mixture of gold colloid and copper(II)-ammonia solutions	287
Appendix A2 Initial dissolution rates of gold colloid ( $R_i$ ) in difference solutions	288
Appendix A3 Apparent rate constants for shrinking core or shrinking sphere model in difference solutions	292
Appendix A4 Graphs of Results	295
Appendix A5 Reaction order and electrochemistry theory	324
Appendix A6 Optimum reagent concentrations and conditions to leach gold from this work	326
Appendix A7 Front page of the three publications with abstract	327

## List of Figures

Fig. 1.1	A diagrammatic representation of the composition of gold colloid	12
Fig. 2.1	Potential-pH diagrams at low reagent concentrations for copper-gold-ammonia-thiosulfate systems	21
Fig. 2.2	Effect of ammonia concentration on gold(I) species distribution	22
Fig. 2.3	Effect of pH on copper(I)/(II) species distribution	22
Fig. 2.4	Potential-pH diagram for the metastable S-H <sub>2</sub> O system at 25°C	24
Fig. 3.1	Transmission electron micrographs of gold colloid particles	83-84
Fig. 3.2	Experimental set up for gold colloid oxidation	86
Fig. 3.3	Experimental set up for gold powder oxidation	87
Fig. 3.4	Effect of nitrogen flow rate on de-oxygenation of reaction solution	90
Fig. 3.5	Comparison between UV-visible spectra of stabilized gold colloid and Na <sub>3</sub> Au(S <sub>2</sub> O <sub>3</sub> ) <sub>2</sub>	93
Fig. 3.6	Comparison between UV-visible spectra of copper(II) tetraammine and copper(I)-thiosulfate complex	93
Fig. 3.7	UV-visible spectrum for the Cu(II)-NH <sub>3</sub> -S <sub>2</sub> O <sub>3</sub> <sup>2-</sup> system	94
Fig. 3.8	Effect of particle size on UV-visible spectrum of unstabilized gold colloids	95
Fig. 3.9	Effect of particle size on UV-visible spectrum of stabilized gold colloids	95
Fig. 3.10	Effect of storage time on UV-visible spectrum of stabilized gold colloids	96
Fig. 3.11	Effect of storage time on UV-visible spectrum of unstabilized gold colloids	96
Fig. 3.12	Effect of ionic strength and air on UV-visible absorption of stabilized gold colloids	97
Fig. 3.13	Verification of the Beer-Lambert Law for gold colloids	98
Fig. 3.14	Verification of the Beer-Lambert Law for copper(II)	98
Fig. 3.15	UV-visible spectra for gold colloid, silver colloid, Cu(NH <sub>3</sub> ) <sub>4</sub> <sup>2+</sup> and a mixture of colloids and Cu(NH <sub>3</sub> ) <sub>4</sub> <sup>2+</sup>	100

Fig. 3.16	Variation of UV-visible absorption with time in a typical reaction solution	100
Fig. 3.17	Effect of copper(II) concentration on initial absorbance of gold colloids	102
Fig. 4.1	Effect of cyanide on gold dissolution at variable ionic strengths	111
Fig. 4.2	Effect of cyanide on gold dissolution at ionic strength 0.5	112
Fig. 4.3	Effect of oxygen on gold dissolution in non-ammoniacal thiosulfate solutions	113
Fig. 4.4	Effect of oxygen on gold potential in non-ammoniacal thiosulfate solutions	113
Fig. 4.5	Effect of oxygen on gold dissolution in ammoniacal thiosulfate solutions	114
Fig. 4.6	Effect of oxygen on gold potential in ammoniacal thiosulfate solutions	114
Fig. 4.7	Effect of main reagents on gold dissolution in copper(II)-ammonia-thiosulfate solutions	118
Fig. 4.8	Effect of main reagents on gold and platinum electrode potentials in copper(II)-ammonia-thiosulfate solutions	118
Fig. 4.9	The dissolution of gold and silver colloid in copper(II)-ammonia-thiosulfate solutions	129
Fig. 4.10	Effect of silver nitrate on gold dissolution in copper(II)-ammonia-thiosulfate solutions	129
Fig. 4.11	Effect of oxygen on gold dissolution in copper(II)-ammonia-thiosulfate solution	135
Fig. 4.12	Effect of oxygen on electrode potential in copper(II)-ammonia-thiosulfate solution	135
Fig. 4.13	Effect of oxygen on residual copper(II) concentration in copper(II)-ammonia-thiosulfate solutions	137
Fig. 4.14	Effect of oxygen on platinum electrode potential in copper(II)-ammonia-thiosulfate solutions	137
Fig. 4.15	Comparison of gold and platinum electrode potentials in copper(II)-ammonia-thiosulfate solutions	138

Fig. 4.16	Comparison of gold and platinum electrode potentials in oxygenated copper(II)-ammonia-thiosulfate solutions	138
Fig. 4.17	Effect of copper(II) on the dissolution of gold powder in copper(II)-ammonia-thiosulfate solution	142
Fig. 4.18	Effect of copper(II) on gold electrode potential in copper(II)-ammonia-thiosulfate solution	142
Fig. 4.19	Change in residual copper(II) concentration during gold powder dissolution	142
Fig. 5.1	Schematic representation of current-potential curves for gold oxidation and copper(II) reduction	147
Fig. 5.2	Measured and calculated potentials of platinum electrode in copper(II)-ammonia-thiosulfate solution under nitrogen atmosphere	153
Fig. 5.3	Measured potentials of platinum electrode in copper(II)-ammonia-thiosulfate solutions under nitrogen or oxygen atmosphere	153
Fig. 5.4	Measured and calculated potentials of platinum in copper(II)-ammonia-thiosulfate solution under nitrogen atmosphere	154
Fig. 5.5	Measured and calculated potentials of gold electrode in oxygenated ammonia-thiosulfate solutions	158
Fig. 5.6	Measured and calculated potentials of gold electrode in copper(II)-ammonia-thiosulfate solutions under nitrogen	158
Fig. 5.7	Comparison between measured and calculated potentials of gold electrode during gold dissolution	159
Fig. 5.8	Measured and calculated potentials of gold electrode in ammonia solutions without thiosulfate	162
Fig. 5.9	Measured and calculated potentials of gold electrode in copper(II)-ammonia-thiosulfate-silver(I) or chloride solution	164
Fig. 5.10	Measured and calculated potentials of gold electrode in copper(II)-ammonia-thiosulfate-carbonate solution	164
Fig. 5.11	Measured and calculated potentials of gold electrode in copper(II)-ammonia-thiosulfate-sulfite or trithionate solutions	165
Fig. 6.1	Dissolution of gold colloid in different solution systems at 25°C	170



Fig. 6.2	Effect of sparging nitrogen, air or oxygen on gold dissolution in copper(II)-ammonia-thiosulfate solutions	171
Fig. 6.3	Effect of ammonia, thiosulfate, and dissolved oxygen on gold dissolution in non-copper(II) solutions	171
Fig. 6.4	Effect of oxygen and/or copper(II) on gold dissolution in ammonia-thiosulfate solutions	172
Fig. 6.5	Comparison of gold dissolution in cyanide and thiosulfate solutions	174
Fig. 6.6a	A plot of $\ln(A_0/A_t)$ versus time in gold-copper(II)- ammonia-thiosulfate solution	177
Fig. 6.6b	Initial gold dissolution in copper(II)- ammonia-thiosulfate solution	177
Fig. 6.7	Logarithmic plot of initial rates ( $R_i$ ) as a function of particle size of colloids in copper(II)-ammonia-thiosulfate solutions	178
Fig. 6.8	Effect of cyanide concentration on rates of cyanidation of colloidal gold and massive impurity-free gold at 25°C	181
Fig. 6.9	Effect of copper(II) concentration or rotation speed on the rate of dissolution of silver and gold-silver (5%) alloy	186
Fig. 6.10	Effect of temperature on initial rate and final gold dissolution	191
Fig. 6.11	Arrhenius plots based on initial rates	191
Fig. 6.12	Arrhenius plots based on initial rates at a residual copper(II) concentration of 1.75 mM	192
Fig. 6.13	Effect of ammonia and thiosulfate on initial rates of gold dissolution by oxygen	195
Fig. 6.14	Comparison between initial rates and final gold dissolution	195
Fig. 6.15	Effect of chloride on initial rates and final gold dissolution by oxygen	197
Fig. 6.16	Comparison between initial rates and final gold dissolution by oxygen	198
Fig. 6.17	Effect of copper(II) on initial rate of gold dissolution	199
Fig. 6.18	Effect of ammonia, thiosulfate and pH on initial rates of gold dissolution in copper(II) solutions	201
Fig. 6.19	Effect of pH on initial rates of gold dissolution in copper-ammonia-thiosulfate solutions	201
Fig. 6.20	Effect of sulfate on initial rate of gold dissolution and residual copper(II) in ammonia-thiosulfate solutions	203

Fig. 6.21	Effect of chloride on initial rate of gold dissolution and residual copper(II) in ammonia solutions	203
Fig. 6.22	Effect of sulfite or carbonate on residual copper(II) in ammonia-thiosulfate solution	204
Fig. 6.23	Effect of sulfite or carbonate on potential of platinum electrode	204
Fig. 6.24	Effect of sulfite or carbonate on gold dissolution in copper(II)-ammonia-thiosulfate solutions	204
Fig. 6.25	Effect of nitrate, chloride or silver(I) ions on initial rate of gold dissolution	205
Fig. 6.26	Comparison between initial rates and final gold dissolution in the presence of chloride, nitrate or silver(I)	205
Fig. 6.27	Effect of initial copper(II) on initial rate of gold dissolution and residual copper(II) concentration	208
Fig. 6.28	Effect of initial copper(II) on final gold dissolution	208
Fig. 6.29	Effect of thiosulfate on initial rate of gold dissolution and residual copper(II) concentration	209
Fig. 6.30	Effect of thiosulfate on final gold dissolution	209
Fig. 6.31	Effect of oxygen and copper(II) on initial rate of gold dissolution	211
Fig. 6.32	Effect of oxygen and copper(II) on final gold dissolution	211
Fig. 6.33	Effect of oxygen on the rate of gold oxidation by copper(II)	211
Fig. 6.34	Reaction model for the oxidation of thiosulfate and gold by copper(II)	216
Fig. 6.35	Effect of ammonia/thiosulfate concentration ratio on initial rate of copper(II) reduction	217
Fig. 6.36	Effect of ammonia/thiosulfate concentration ratio on initial rate of gold oxidation	217
Fig. 6.37	Effect of decreasing thiosulfate concentration on initial rate of gold oxidation	218
Fig. 6.38	Effect of increasing ammonia concentration or pH on initial rate of gold oxidation	218
Fig. 6.39	Effect of increasing thiosulfate concentration on initial rate of gold oxidation	220
Fig. 6.40	Effect of increasing ammonia concentration or pH on initial rate of gold oxidation	220

Fig. 7.1	Logarithmic plot of initial rate of gold oxidation as a function of oxygen or ammonia concentration in non-copper solutions	227
Fig. 7.2	Logarithmic plot of initial rate of gold oxidation by copper(II) as a function of reagent concentration	227
Fig. 7.3	Logarithmic plot of initial rate of gold oxidation as a function of chloride, nitrate, or silver(I) concentration in anaerobic solutions	228
Fig. 7.4	Logarithmic plot of initial rate of gold oxidation as a function of copper(II) concentration in anaerobic solutions	229
Fig. 7.5	Logarithmic plot of initial rate of gold oxidation as a function of copper(II) and thiosulfate concentration in oxygenated solutions	230
Fig. 7.6	Reaction equilibria involved in the oxidation of thiosulfate by copper(II)	234
Fig. 7.7	Initial rate of gold oxidation as a function of $\{[\text{Cu(II)}] \cdot [\text{X}]\}^{0.5}$ at variable copper(II) concentrations in ammonia-thiosulfate solutions under nitrogen.	240
Fig. 7.8	Initial rate of gold dissolution as a function of $\{[\text{Cu(II)}] \cdot [\text{S}_2\text{O}_3^{2-}]\}^{0.5}$ at variable thiosulfate concentrations in copper(II)-ammonia solutions under nitrogen	240
Fig. 7.9	Initial rate of gold oxidation as a function of $\{[\text{Cu(II)}] \cdot [\text{NH}_3]\}^{0.5}$ at variable ammonia concentrations in copper(II)-ammonia solution under nitrogen	241
Fig. 7.10	Validity of shrinking sphere model for gold colloids in copper(II)-ammonia-thiosulfate solutions under nitrogen	245
Fig. 7.11	Validity of shrinking sphere model for gold powder in copper(II)-ammonia-thiosulfate solutions under nitrogen	245
Fig. 7.12	Validity of shrinking sphere/core model for gold colloid in copper-ammonia (low)-thiosulfate solutions under nitrogen	245
Fig. 7.13	Validity of shrinking sphere/core model for gold colloid in copper(II)-ammonia (high)-thiosulfate solutions under nitrogen	245
Fig. 7.14	Shrinking sphere model for the dissolution of stabilized gold colloids of different particle size in copper(II)-ammonia-thiosulfate solutions under nitrogen	246

Fig. 7.15	Shrinking sphere model for the dissolution of unstabilized gold colloids in copper(II)-ammonia-thiosulfate solutions under nitrogen	246
Fig. 7.16	Logarithmic plot of $k_{ss}$ as a function of $(bC/rp)$ for colloidal gold of different particle sizes	247
Fig. 7.17	Shrinking sphere model for the dissolution of gold powder of different particle sizes in copper(II)-ammonia-thiosulfate solutions under nitrogen	248
Fig. 7.18	Shrinking sphere model for the dissolution of gold powder at different copper(II) concentrations in ammonia-thiosulfate solutions under nitrogen	248
Fig. 7.19	Shrinking sphere model for the dissolution of gold colloid at different thiosulfate concentrations in oxygenated copper(II)-ammonia solutions	250
Fig. 7.20	Effect of tetrathionate or trithionate on residual copper(II) during reaction with thiosulfate.	251
Fig. 7.21	Effect of tetrathionate or trithionate on residual thiosulfate during reaction of copper(II) with thiosulfate.	252
Fig. 7.22	Effect of tetrathionate or trithionate on the dissolution of gold foil in copper(II)-ammonia-thiosulfate solutions	252
Fig. 7.23	Shrinking sphere model for the dissolution of gold colloids by copper(II)-ammonia-thiosulfate solutions in the presence of trithionate or tetrathionate under nitrogen.	253
Fig. 7.24	Formation of tetrathionate and trithionate during the dissolution of gold foil by copper(II)-ammonia-thiosulfate solutions under nitrogen	254
Fig. 7.25	Effect of oxygenation on dissolution of gold foil	255
Fig. 7.26	Shrinking sphere model for the dissolution of gold colloids by copper(II)-ammonia-thiosulfate solutions in the presence of silver(I) under nitrogen	256
Fig. 7.27	Shrinking sphere model for the dissolution of gold colloids by copper(II)-ammonia-thiosulfate solutions in the presence of sodium chloride under nitrogen	256

Fig. 7.28	Shrinking sphere model for the dissolution of gold colloids by copper(II)- thiosulfate solutions at different ammonia concentrations under nitrogen	256
Fig. 7.29	Logarithmic plot of apparent rate constant as a function of silver(I), chloride, or ammonia concentration in copper(II)-ammonia-thiosulfate solutions under nitrogen	257
Fig. 7.30	Effect of silver(I) concentration on silver(I) speciation at 25°C	260
Fig. 7.31	Comparison between calculated silver(I)/silver(0) and gold(I)/gold(0) potentials at different silver(I) and gold(I) concentrations	261
Fig. 7.32	Logarithmic plot of apparent rate constants as a function of reagent concentration	263
Fig. A4.1a	Effect of thiosulfate on gold dissolution in the absence of ammonia	295
Fig. A4.1b	Effect of thiosulfate on gold electrode potential in the absence of ammonia	295
Fig. A4.1c	Effect of thiosulfate on gold dissolution in the presence of ammonia	295
Fig. A4.1d	Effect of thiosulfate on gold electrode potential in the presence of ammonia	295
Fig. A4.2a	Effect of ammonia on gold dissolution in the absence of thiosulfate	296
Fig. A4.2b	Effect of ammonia on gold electrode potential in the absence of thiosulfate	296
Fig. A4.2c	Effect of ammonia on gold dissolution in the presence of thiosulfate	296
Fig. A4.2d	Effect of ammonia on gold electrode potential in the presence of thiosulfate	296
Fig. A4.3a	Effect of chloride on gold dissolution in the absence of ammonia	297
Fig. A4.3b	Effect of chloride on gold electrode potential in the absence of ammonia	297
Fig. A4.4a	Effect of chloride on gold dissolution in the absence of thiosulfate	298
Fig. A4.4b	Effect of chloride on gold electrode potential in the absence of thiosulfate	298

Fig. A4.5a	Effect of chloride on gold dissolution in the presence of both ammonia and thiosulfate	299
Fig. A4.5b	Effect of chloride on gold electrode potential in the presence of both ammonia and thiosulfate	299
Fig. A4.6a	Effect of temperature on gold dissolution in oxygenated-ammonia-thiosulfate solution	300
Fig. A4.6b	Effect of temperature on gold electrode potential in copper(II)-ammonia-thiosulfate solution	300
Fig. A4.7a	Effect of concentration of gold colloid on gold dissolution in copper(II)-ammonia-thiosulfate solution	301
Fig. A4.7b	Effect of concentration of gold colloid on gold electrode potential in copper(II)-ammonia-thiosulfate solution	301
Fig. A4.7c	Change in residual concentration of copper(II) in copper(II)-ammonia-thiosulfate solution	301
Fig. A4.8a	Effect of particle size of stabilized gold colloid on gold dissolution in copper(II)-ammonia-thiosulfate solution	302
Fig. A4.8b	Effect of particle size of stabilized gold colloid on gold electrode potential in copper(II)-ammonia-thiosulfate solution	302
Fig. A4.9a	Effect of particle size of unstabilized gold colloid on gold dissolution in copper(II)-ammonia-thiosulfate solution	303
Fig. A4.9b	Effect of particle size of unstabilized gold colloid on gold electrode potential in copper(II)-ammonia-thiosulfate solution	303
Fig. A4.10a	Copper(II) concentration during reaction of copper(II) and thiosulfate in copper(II)-ammonia-thiosulfate solution in the absence of gold colloids	304
Fig. A4.10b	Platinum electrode potential during reaction of copper(II) and thiosulfate in copper(II)-ammonia-thiosulfate solution in the absence of gold colloids	304
Fig. A4.11a	Effect of ionic strength on gold dissolution in copper(II)-ammonia-thiosulfate solution	305
Fig. A4.11b	Effect of ionic strength on gold electrode potential in copper(II)-ammonia-thiosulfate solution	305

Fig. A4.11c	Effect of ionic strength on copper(II) concentration in copper(II)-ammonia-thiosulfate solution	305
Fig. A4.11d	Effect of ionic strength on platinum electrode potential in copper(II)-ammonia-thiosulfate solution	305
Fig. A4.12a	Effect of copper(II) on gold dissolution in the absence of thiosulfate	306
Fig. A4.12b	Effect of copper(II) on gold electrode potential in the absence of thiosulfate	306
Fig. A4.13a	Effect of copper(II) on gold dissolution in copper(II)-ammonia-thiosulfate solution	307
Fig. A4.13b	Effect of copper(II) on gold electrode potential in copper(II)-ammonia-thiosulfate solution	307
Fig. A4.13c	Effect of initial copper(II) concentration on residual copper(II) in copper(II)-ammonia-thiosulfate solution	307
Fig. A4.13d	Effect of copper(II) on platinum electrode potential in copper(II)-ammonia-thiosulfate solution	307
Fig. A4.14a	Effect of thiosulfate on gold dissolution in copper(II)-ammonia-thiosulfate solution	308
Fig. A4.14b	Effect of thiosulfate on gold electrode potential in copper(II)-ammonia-thiosulfate solution	308
Fig. A4.14c	Effect of thiosulfate on copper(II) concentration in copper(II)-ammonia-thiosulfate solution	308
Fig. A4.14d	Effect of thiosulfate on platinum electrode potential in copper(II)-ammonia-thiosulfate solution	308
Fig. A4.15a	Effect of ammonia on gold dissolution in the absence of thiosulfate	309
Fig. A4.15b	Effect of ammonia on gold electrode potential in the absence of thiosulfate	309
Fig. A4.16a	Effect of ammonia on gold dissolution in copper(II)-ammonia-thiosulfate solution	310
Fig. A4.16b	Effect of ammonia on gold electrode potential in copper(II)-ammonia-thiosulfate solution	310
Fig. A4.16c	Effect of ammonia on copper(II) concentration in copper(II)-ammonia-thiosulfate solution	310

Fig. A4.16d Effect of ammonia on platinum electrode potential in copper(II)-ammonia-thiosulfate solution	310
Fig. A4.17a Effect of pH on gold dissolution in copper(II)-ammonia-thiosulfate solution	311
Fig. A4.17b Effect of pH on gold electrode potential in copper(II)-ammonia-thiosulfate solution	311
Fig. A4.17c Effect of pH on copper(II) concentration in copper(II)-ammonia-thiosulfate solution	311
Fig. A4.17d Effect of pH on platinum electrode potential in copper(II)-ammonia-thiosulfate solution	311
Fig. A4.18a Effect of nitrate on gold dissolution in copper(II)-ammonia-thiosulfate solution	312
Fig. A4.18b Effect of nitrate on gold electrode potential in copper(II)-ammonia-thiosulfate solution	312
Fig. A4.18c Effect of nitrate on copper(II) concentration in copper(II)-ammonia-thiosulfate solution	310
Fig. A4.18d Effect of nitrate on platinum electrode potential in copper(II)-ammonia-thiosulfate solution	312
Fig. A4.19a Effect of silver(I) on gold dissolution in copper(II)-ammonia-thiosulfate solution	313
Fig. A4.19b Effect of silver(I) on gold electrode potential in copper(II)-ammonia-thiosulfate solution	313
Fig. A4.19c Effect of silver(I) on copper(II) concentration in copper(II)-ammonia-thiosulfate solution	313
Fig. A4.19d Effect of silver(I) on platinum electrode potential in copper(II)-ammonia-thiosulfate solution	313
Fig. A4.20a Comparison of gold electrode potentials in the present of silver colloid or silver nitrate	312
Fig. A4.20b Copper(II) concentrations in the present of silver colloid or silver nitrate	314
Fig. A4.20c Platinum electrode potential in the present of silver colloid or silver nitrate	314



Fig. A4.21a Comparison of gold dissolution in the present of lead(II) or silver nitrate	315
Fig. A4.21b Gold electrode potential in the present of lead(II)	315
Fig. A4.21c Copper(II) concentration in the present of lead(II)	315
Fig. A4.21d Platinum electrode potential in the present of lead(II)	315
Fig. A4.22a Effect of chloride on gold dissolution in the absence of thiosulfate	316
Fig. A4.22b Effect of chloride on gold electrode potential in the absence of thiosulfate	316
Fig. A4.22c Effect of chloride on copper(II) concentration in the absence of thiosulfate	316
Fig. A4.23a Effect of chloride on gold dissolution in copper(II)-ammonia-thiosulfate solution	317
Fig. A4.23b Effect of chloride on gold electrode potential in copper(II)-ammonia-thiosulfate solution	317
Fig. A4.23c Effect of chloride on copper(II) concentration in copper(II)-ammonia-thiosulfate solution	317
Fig. A4.24a Effect of $S_3O_6^{2-}$ , $S_4O_6^{2-}$ , $CO_3^{2-}$ and $SO_3^{2-}$ on gold dissolution in copper(II)-ammonia-thiosulfate solution	318
Fig. A4.24b Effect of $S_3O_6^{2-}$ , $S_4O_6^{2-}$ , $CO_3^{2-}$ and $SO_3^{2-}$ on gold electrode potential in copper(II)-ammonia-thiosulfate solution	318
Fig. A4.24c Effect of $S_3O_6^{2-}$ , $S_4O_6^{2-}$ , $CO_3^{2-}$ and $SO_3^{2-}$ on copper(II) concentration in copper(II)-ammonia-thiosulfate solution	318
Fig. A4.24d Effect of $S_3O_6^{2-}$ , $S_4O_6^{2-}$ , $CO_3^{2-}$ and $SO_3^{2-}$ on platinum electrode potential in copper(II)-ammonia-thiosulfate solution	318
Fig. A4.25a Effect of temperature on gold dissolution in copper(II)-ammonia-thiosulfate solution	319
Fig. A4.25b Effect of temperature on gold electrode potential in copper(II)-ammonia-thiosulfate solution	319
Fig. A4.25c Effect of temperature on copper(II) concentration in copper(II)-ammonia-thiosulfate solution	319
Fig. A4.25d Effect of temperature on platinum electrode potential in copper(II)-ammonia-thiosulfate solution	319

Fig. A4.26a Effect of copper(II) on gold dissolution in oxygenated copper(II)-ammonia-thiosulfate solution	320
Fig. A4.26b Effect of copper(II) on gold electrode potential in oxygenated copper(II)-ammonia-thiosulfate solution	320
Fig. A4.26c Variation of copper(II) concentration in oxygenated copper(II)-ammonia-thiosulfate solution	320
Fig. A4.26d Effect of copper(II) on platinum electrode potential in oxygenated copper(II)-ammonia-thiosulfate solution	320
Fig. A4.27a Effect of thiosulfate on gold dissolution in oxygenated copper(II)-ammonia-thiosulfate solution	321
Fig. A4.27b Effect of thiosulfate on gold electrode potential in oxygenated copper(II)-ammonia-thiosulfate solution	321
Fig. A4.27c Effect of thiosulfate on copper(II) concentration in oxygenated copper(II)-ammonia-thiosulfate solution	321
Fig. A4.27d Effect of thiosulfate on platinum electrode potential in oxygenated copper(II)-ammonia-thiosulfate solution	321
Fig. A4.28a Effect of silver(I) and chloride ions on the dissolution of gold powder in copper(II)-ammonia-thiosulfate solution	322
Fig. A4.28b Effect of silver(I) and chloride ions on gold electrode potential in copper(II)-ammonia-thiosulfate solution	322
Fig. A4.28c Effect of silver(I) and chloride ions on copper(II) concentration in copper(II)-ammonia-thiosulfate solution	322
Fig. A4.29a Dissolution of gold powder with difference particle size in copper(II)-ammonia-thiosulfate solution	323
Fig. A4.29b Effect of particle size of gold powder on gold electrode potential in copper(II)-ammonia-thiosulfate solution	323
Fig. A4.29c Effect of particle size of gold powder on copper(II) concentration in copper(II)-ammonia-thiosulfate solution	323

## List of Tables

Table 1.1	Gold lixiviants, conditions and references	5
Table 1.2	Stability constants of selected Au(I) and Au(III) complexes and corresponding standard reduction potentials at 25°C	6
Table 1.3	Relevant properties of lixiviants	7
Table 1.4	Factors investigated in this study	14
Table 2.1	Sulfur-oxygen species and oxidation states	24
Table 2.2	Summary of reactions and conditions for thiosulfate regeneration	25
Table 2.3	Stoichiometry of oxidation and reductive decomposition of thiosulfate	32
Table 2.4	Stoichiometry of the stabilization reaction of thiosulfate	35
Table 2.5	Different types of ores/concentrates used in thiosulfate leaching	36
Table 2.6	Chemical composition of different types of starting material used in thiosulfate leaching	37
Table 2.7	Maximum gold leaching under optimum conditions	38
Table 2.8	Dissolution rates of gold under optimum conditions	46
Table 2.9a	Effect of thiosulfate concentration on gold dissolution rates	49
Table 2.9b,c	Effect of thiosulfate concentration on gold leaching	51-52
Table 2.10	Effect of copper concentration on gold leaching	54
Table 2.11	Effect of ammonia concentration on gold leaching	58
Table 2.12	Effect of pH on gold leaching	60
Table 2.13	Effect of temperature on gold leaching	64
Table 2.14	Effect of sulfur-oxy species on gold dissolution rates	65
Table 2.15	Effect of retention time on gold leaching	69
Table 2.16	Effect of oxidants on gold leaching	71
Table 3.1	List of materials and reagents	79
Table 3.2	Effect of amount of sodium citrate on the particle size and colour of gold colloids	81
Table 3.3	Stability of gold colloid upon storage	96
Table 3.4	Experimental conditions used in Au(c)-oxygen-cyanide systems	106

Table 3.5	Experimental conditions used in Au(c)-oxygen-thiosulfate-ammonia systems	107
Table 3.6	Experimental conditions used in Au(c)-copper(II)-thiosulfate-ammonia-nitrogen systems	108
Table 3.7	Experimental conditions used in Au(c)-copper(II)-oxygen-thiosulfate-ammonia systems	109
Table 3.8	Experimental conditions used in Gold powder-copper(II)-thiosulfate-ammonia systems	109
Table 4.1	Effect of thiosulfate on the dissolution and potentials of gold	115
Table 4.2	Effect of ammonia on the dissolution and potentials of gold	115
Table 4.3	Effect of chloride on the dissolution and potentials of gold	116
Table 4.4	Effect of temperature on the dissolution and potentials of gold	117
Table 4.5	Effect of colloid strength on the dissolution and potentials of gold	119
Table 4.6	Effect of colloid particle size on the dissolution and potentials of gold	120
Table 4.7	Effect of ionic strength on the dissolution and potentials of gold	121
Table 4.8	Effect of copper(II) on the dissolution and potentials of gold	123
Table 4.9	Effect of thiosulfate on the dissolution and potentials of gold	124
Table 4.10	Effect of ammonia on the dissolution and potentials of gold	125
Table 4.11	Effect of ammonia on the residual copper(II) and potentials of platinum electrode	125
Table 4.12	Effect of pH on the dissolution and potentials of gold	126
Table 4.13	Effect of pH on residual copper(II) concentration and platinum electrode potentials	127
Table 4.14	Effect of nitrate on the dissolution and potentials of gold	127
Table 4.15	Effect of silver(I) on the dissolution and potentials of gold	128
Table 4.16	The dissolution of gold and silver colloids in copper(II)-ammonia-thiosulfate solutions	130
Table 4.17	Effects of lead(II) and silver(I) on the dissolution and potentials of gold	130
Table 4.18	Effect of chloride on the dissolution and potentials of gold in the absence of thiosulfate	131

Table 4.19	Effect of chloride on the dissolution and potentials of gold in the presence of thiosulfate	132
Table 4.20	Effect of different anions on the dissolution and potentials of gold and residual copper(II)	133
Table 4.21	Effect of temperature on the dissolution and potentials of gold and residual copper(II)	134
Table 4.22	Effect of oxygen on gold dissolution in oxygenated copper(II)-ammonia-thiosulfate solution	136
Table 4.23	Effect of copper(II) concentration on gold dissolution in oxygenated copper(II)-ammonia-thiosulfate solution	139
Table 4.24	Effect of thiosulfate on gold dissolution in oxygenated copper(II)-ammonia-thiosulfate solution	140
Table 4.25	Effect of initial copper(II) on the dissolution of gold colloid, gold powder and gold electrode potentials	141
Table 4.26	Effect of silver(I) and chloride ions on the dissolution of gold powder and gold electrode potentials	143
Table 4.27	A comparison of gold dissolution with different particle size	144
Table 5.1	Possible chemical reactions involved in gold oxidation	146
Table 5.2	Effect of complex-formation on standard reduction potentials at 25°C	150
Table 5.3	Comparison between measured and predicted $E^{\circ}\{\text{Cu}(\text{NH}_3)_n^{2+}/\text{Cu}(\text{S}_2\text{O}_3)_m^{1-2m}\}$ for different redox couples	155
Table 5.4	Effect of initial reagent concentration on measured equilibrium potential (E, SHE) and $E^{\circ}$ for gold(I)/(0) redox couple	161
Table 5.5	Effect of initial reagent concentrations on measured equilibrium potential (E, SHE) and $E^{\circ}$ for gold(I)/(0) redox couple	163
Table 6.1	Decrease in thiosulfate concentration with time	173
Table 6.2	Effect of gold concentration on initial rate ( $R_i$ )	180
Table 6.3	Effect of particle size of gold on initial rate ( $R_i$ )	180
Table 6.4	Initial rates for cyanidation	182

Table 6.5	Effect of the type of gold and additives on the initial rate of dissolution	188
Table 6.6	Comparison of rate data for massive gold and colloidal gold at 25 °C	189
Table 6.7	Effect of oxygen on the initial rate of gold dissolution	193
Table 6.8	Effect of ammonia and thiosulfate on dissolution rate of massive gold	196
Table 6.9	Comparison between oxidation rate of gold and thiosulfate	212
Table 6.10	Comparison of residual copper(II) concentration after aeration/oxygenation of ammoniacal-thiosulfate solutions	213
Table 6.11	Experimental conditions representing copper(II) reduction and massive gold oxidation	218
Table 6.12	Experimental conditions representing gold colloid oxidation	219
Table 7.1	Reaction orders based on initial rate with respect to each reagent at 25°C	225
Table 7.2	Effect of copper(II) on initial rate of gold colloid and powder oxidation	229
Table 7.3	Calculation of mass transfer coefficient of copper(II) to gold surface	232
Table 7.4	Comparison of initial rates of gold dissolution at 25 °C	236
Table 7.5	Effect of $\text{Cu}(\text{NH}_3)_4^{2+}$ concentrations on the dissolution rate of gold	237
Table 7.6	Rate constant for the dissolution of gold powder based on a shrinking sphere model	249
Table 7.7	Reaction orders for the dissolution of gold colloid by copper(II) based on apparent rate constants	257
Table 7.8	Stability constants of gold and silver complexes at 25°C	259
Table 7.9	Standard Gibbs free energy values ( $\Delta G_{f, 298 \text{ K}}^{\circ}$ ) and equilibrium constants for silver and gold dissolution	259
Table 7.10	Effect of sodium and silver nitrates on apparent rate constants and rates	262

Table A1.1 Comparison between measured and calculated absorbance for the mixture of gold colloid in copper(II)-ammonia solution	287
Table A2.1 Initial dissolution rates of gold colloid ( $R_i$ ) and reaction orders in oxygenated ammonia-thiosulfate solution: effect of reagent concentration	288
Table A2.2 Initial dissolution rates of gold colloid ( $R_i$ ) and reaction orders in copper(II)-ammonia-thiosulfate solution: effect of reagent concentration	289
Table A2.3 Initial dissolution rates of gold colloid ( $R_i$ ) and reaction orders in copper(II)-ammonia-thiosulfate solution: effect of reagent concentration	290
Table A2.4 Initial dissolution rates of gold colloid ( $R_i$ ) and reaction orders in oxygenated copper(II)-ammonia-thiosulfate solution: effect of reagent concentration	291
Table A2.5 Effect of temperature on initial dissolution rates of gold colloid ( $R_i$ )	291
Table A3.1 Apparent rate constants for shrinking core and shrinking sphere models in copper(II)-ammonia-thiosulfate solutions	292
Table A3.2 Apparent rate constants for shrinking sphere model in copper(II)-ammonia-thiosulfate solutions	293
Table A3.3 Apparent rate constants for shrinking sphere model in oxygenated copper(II)-ammonia-thiosulfate solutions	294
Table A3.4 Apparent rate constants for shrinking sphere model in copper(II)-ammonia-thiosulfate solutions: effect of particle size of gold colloid and gold powder	294
Table A6.1 Copper(II)-ammonia-thiosulfate system without oxygen	326
Table A6.2 Oxygenated copper(II)-ammonia-thiosulfate system	326