

การเก็บกลับคืนโลหะมีค่าและพลาสติกจากซากเครื่องใช้ไฟฟ้าขนาดเล็กโดยการคัดแยกด้วยจิก



นายธีระยุทธ เฟื่องสะอาด

บทคัดย่อและแฟ้มข้อมูลฉบับเต็มของวิทยานิพนธ์ตั้งแต่ปีการศึกษา 2554 ที่ให้บริการในคลังปัญญาจุฬาฯ (CUIR)  
เป็นแฟ้มข้อมูลของนิสิตเจ้าของวิทยานิพนธ์ ที่ส่งผ่านทางบัณฑิตวิทยาลัย

The abstract and full text of theses from the academic year 2011 in Chulalongkorn University Intellectual Repository (CUIR)  
are the thesis authors' files submitted through the University Graduate School.

วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิศวกรรมศาสตรมหาบัณฑิต

สาขาวิชาวิศวกรรมทรัพยากรธรณี ภาควิชาวิศวกรรมเหมืองแร่และปิโตรเลียม

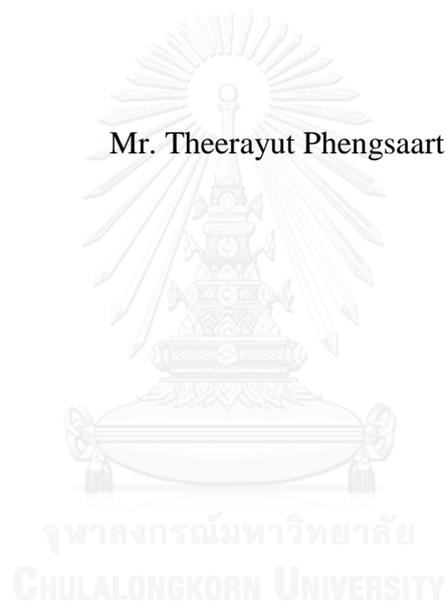
คณะวิศวกรรมศาสตร์ จุฬาลงกรณ์มหาวิทยาลัย

ปีการศึกษา 2558

ลิขสิทธิ์ของจุฬาลงกรณ์มหาวิทยาลัย

RECOVERY OF VALUABLE METALS AND PLASTICS FROM END-OF-LIFE  
SMALL HOME APPLIANCES BY JIG SEPARATION

Mr. Theerayut Phengsaart



A Thesis Submitted in Partial Fulfillment of the Requirements  
for the Degree of Master of Engineering Program in Georesources Engineering  
Department of Mining and Petroleum Engineering  
Faculty of Engineering  
Chulalongkorn University  
Academic Year 2015  
Copyright of Chulalongkorn University



ธีระยุทธ เฟื่องสะอาด : การเก็บกลับคืน โลหะมีค่าและพลาสติกจากซากเครื่องใช้ไฟฟ้าขนาดเล็ก โดยการคัดแยกด้วยจิ๊ก (RECOVERY OF VALUABLE METALS AND PLASTICS FROM END-OF-LIFE SMALL HOME APPLIANCES BY JIG SEPARATION) อ.ที่ปรึกษาวิทยานิพนธ์หลัก: รศ. ดร. ดาวลัย วิวรรณนะเดช, 68 หน้า.

การคัดแยกด้วยจิ๊ก เป็นเทคนิคการคัดแยกวัสดุโดยใช้ความแตกต่างด้านความหนาแน่นของวัสดุ และเป็นหนึ่งในวิธีเก่าแก่ที่ถูกใช้อย่างกว้างขวางในกระบวนการแตงแร่ เนื่องจากกระบวนการไม่ซับซ้อน ประสิทธิภาพสูง และต้นทุนต่ำ ต่อมาได้ถูกนำมาประยุกต์ใช้ได้ผลดีในการเก็บกลับคืนโลหะและพลาสติกจากซากเครื่องใช้ไฟฟ้าและอิเล็กทรอนิกส์ เช่น เครื่องใช้ไฟฟ้าขนาดเล็กในครัวเรือน และคอมพิวเตอร์ เป็นต้น ในการศึกษาครั้งนี้ ได้มีการนำการคัดแยกด้วยจิ๊ก (RETAC® jig) มาประยุกต์ใช้ในการนำกลับคืนโลหะและพลาสติกจากซากเครื่องใช้ไฟฟ้าขนาดเล็กในครัวเรือน หลังผ่านกระบวนการคัดแยกด้วยแม่เหล็กและกระแสไฟฟ้าวน ผลการศึกษาพบว่า ประสิทธิภาพการคัดแยกลดลงตามปริมาณลวดทองแดง ทั้งนี้เนื่องจากลวดทองแดงเกิดการพันกันได้ง่ายในห้องแยกขณะทำการคัดแยกด้วยจิ๊ก และลวดทองแดงที่พันกันนี้ได้กีดขวางการเคลื่อนที่ของพลาสติกและชิ้นส่วนพีซีบีในห้องแยก แต่ยังไม่ทราบแน่ชัดว่าปัจจัยใดบ้างที่ส่งผลกระทบต่อประสิทธิภาพการคัดแยกที่นำเสนอในนี้ จึงได้ทำการตรวจสอบผลกระทบจากปัจจัยต่างๆ อาทิ ความยาวและปริมาณของลวดทองแดง โดยใช้ตัวอย่างจำลองที่มีสัดส่วนพลาสติกและลวดทองแดงแตกต่างกัน ผลการศึกษาพบว่าทั้งสัดส่วนเชิงน้ำหนักและความยาวของลวดทองแดงที่มากขึ้นก่อให้เกิดการพันตัวได้มากขึ้น นอกจากนี้ยังพบว่าการปรับปรุงคุณภาพเบื้องต้นโดยการใช้หม้อบดสามารถเก็บกลับคืนลวดทองแดงที่พันกันในหม้อบด และเพิ่มการเก็บกลับคืนลวดทองแดงในการคัดแยกด้วยจิ๊ก กล่าวคือ ทองแดงบริสุทธิ์ 93% ถูกเก็บกลับคืนในกระบวนการปรับปรุงสภาพเบื้องต้นในหม้อบด และอัตราการเก็บกลับคืนของทองแดงในขั้นที่ 6 ของการคัดแยกด้วยจิ๊กเพิ่มขึ้นจาก 66% เป็น 87%

ภาควิชา วิศวกรรมเหมืองแร่และปิโตรเลียม ลายมือชื่อนิสิต .....

สาขาวิชา วิศวกรรมทรัพยากรธรณี ลายมือชื่อ อ.ที่ปรึกษาหลัก .....

ปีการศึกษา 2558



## ACKNOWLEDGEMENTS

Foremost, I would like to thank my thesis advisor, Assoc. Prof. Dr. Dawan Wiwattanadate for the recommendation of this study. Sincere thanks are expressed to my Japanese advisor, Assoc. Prof. Dr. Mayumi Ito, for kind suggestion on the experimental study during my study in Japan. It is difficult to overstate my appreciation to Assoc. Prof. Dr. Pinyo Meechumna, Assoc. Prof. Dr. Quanchai Leepowpanth, and Assoc. Prof. Somsak Saisinchai for teaching me the knowledge background on Mineral Processing and Resources Recycling. Sincere thanks are also expressed to thesis committee members, Dr. Songwut Arthitthong, for his kind recommendation.

I am deeply grateful to the PARE Program of Hokkaido University for giving me the scholarship for study exchange and funding this research. I am indebted to Re-Tem Corporation for providing the sample.

I also would like to express my appreciation to my Japanese professors at Hokkaido University, Prof. Dr. Naoki Hiroyoshi and Assist. Prof. Carlito Baltazar Tabelin for their valuable suggestions during the series of dialoged on progress report. I gratefully thank Mr. Naoki Hamaya and other members in the Plastics Research Group for assistance on this research experiments.

Many thanks are extended to all members in Department of Mining and Petroleum Engineering, Faculty of Engineering, Chulalongkorn University and Laboratory of Mineral Processing and Resources Engineering, Graduate School of Engineering, Hokkaido University, whose friendship and support are beyond a place of study.

I am forever grateful my parent and my family who have always supported and encouraged me to do my best in all matters of life.

The author alone assumes responsibility for the conclusion of this thesis and any errors it may contain.

## CONTENTS

	Page
THAI ABSTRACT .....	iv
ENGLISH ABSTRACT.....	v
ACKNOWLEDGEMENTS .....	vi
CONTENTS.....	vii
LIST OF TABLES .....	ix
LIST OF FIGURES .....	x
CHAPTER I INTRODUCTION.....	1
1.1 General introduction.....	1
1.2 Objectives of this study .....	3
1.3 Scope and outline of this study.....	3
1.4 Expected benefits.....	4
1.5 Order of presentation .....	4
CHAPTER II BACKGROUNDS AND LITERATURE REVIEWS .....	6
2.1 Description of small home appliances.....	6
2.2 Jig separation technology .....	10
2.3 Recovery and recycling of end-of-life small home appliances .....	20
2.4 Literature reviews .....	24
CHAPTER III CHARACTERIZATION OF SAMPLES.....	26
3.1 Characterization of real sample .....	26
3.1.1 Experimental .....	26
3.1.2 Results and discussion.....	29
3.2 Characterization of model samples .....	30
3.2.1 Experimental .....	30
3.2.2 Results and discussion.....	33
CHAPTER IV EFFECTS OF COPPER WIRES ON JIG SEPARATION .....	36
4.1 Jig separation of real sample .....	36
4.1.1 Experimental .....	36
4.1.2 Results and discussion.....	37

	Page
4.2 Jig separation of model samples.....	39
4.2.1 Experimental .....	39
4.2.2 Results and discussion.....	39
CHAPTER V DEVELOPMENT OF PRE-TREATMENT METHOD.....	48
5.1 Pre-treatment of model samples .....	48
5.1.1 Experimental .....	48
5.1.2 Results and discussion.....	50
5.2 Pre-treatment of real sample.....	52
5.2.1 Experimental .....	52
5.2.2 Results and discussion.....	52
CHAPTER VI CONCLUSION .....	56
6.1 Conclusion.....	56
6.2 Recommendation.....	58
REFERENCES .....	60
APPENDIX.....	62
VITA.....	68

## LIST OF TABLES

Table 2. 1 E-waste categories according to the South Korea government regulation .....	9
Table 2. 2 Recovery and purity of copper in the 6 <sup>th</sup> layer of the mixture products.....	23
Table 4. 1 Recovery and purity of copper in the 6 <sup>th</sup> layer at mass ratios variation; 5 %, 10 %, and 15 % (30 mm long copper wires, separation time 3 minutes) .....	44
Table 4. 2 Recovery and purity of copper in the 6 <sup>th</sup> layer at lengths of copper wires variation; 10 mm, 30 mm, and 50 mm (10 % copper content, separation time 3 minutes).....	46
Table 5. 1 Recovery and purity of copper in pre-treatment product of model sample (+16 mm) at rotating time variation; 5, 10, and 15 minutes (10 % copper content, 50 mm long of copper wires) .....	52

## LIST OF FIGURES

Figure 1. 1 Schematic flowchart of the present study.....	4
Figure 2. 1 E-waste category as Japanese definition .....	10
Figure 2. 2 Idealized jig separation process.....	12
Figure 2. 3 Movement of the piston in a jig.....	12
Figure 2. 4 Speed of flow through bed during jig cycle .....	13
Figure 2. 5 Basic Jig Construction.....	14
Figure 2. 6 Outline of BATAC jig.....	15
Figure 2. 7 Schematic outline of the desktop type batch-wise RETAC jig.....	15
Figure 2. 8 Separation process of RETAC jig .....	16
Figure 2. 9 Schematic outline of the desktop type batch-wise reverse jig.....	16
Figure 2. 10 Schematic outline of the desktop type batch-wise hybrid jig.....	17
Figure 2. 11 Schematic outline of the desktop type batch-wise reverse hybrid jig.....	17
Figure 2. 12 Schematic outline of the continuous RETAC jig .....	18
Figure 2. 13 Laboratory scale RETAC jig; batch (left) and continuous type (right) .....	19
Figure 2. 14 Pilot scale RETAC jig .....	19
Figure 2. 15 Recycling flowchart of small home appliances of Re-Tem Corporation (Pilot scale).....	20
Figure 2. 16 Mixture of plastics and metals products; (a) coarse group, (b) medium group, and (c) fine group.....	21
Figure 2. 17 Particle size distributions of the mixture products .....	21
Figure 2. 18 Chemical compositions of mixture products.....	22
Figure 2. 19 Components of each layer after jig separation; (a) coarse group, (b) medium group, and (c) fine group.....	23

Figure 3. 1 Crushed end-of-life small home appliances sample (the fine fraction) .....	26
Figure 3. 2 Three fractions of real sample after hand sorting; (a) copper wires, (b) others metals, and (c) plastics & PCB .....	27
Figure 3. 3 Analysis flowchart of real sample .....	28
Figure 3. 4 ONE TPH-02 type low temperature grinding equipment.....	28
Figure 3. 5 Microwave digestion equipment (ETHOS PLUS) .....	29
Figure 3. 6 Particle size distribution of the real sample.....	30
Figure 3. 7 Chemical composition of the real sample .....	30
Figure 3. 8 Orient mill .....	31
Figure 3. 9 Mixture of model samples; (a) ABS, (b) PVC, and (c) copper wires .	32
Figure 3. 10 Model sample .....	32
Figure 3. 11 Particle size distributions of the plastics mixture in model samples .	33
Figure 3. 12 Copper wires in the model samples; lengths (a) 10 mm, (b) 30 mm, and (c) 50 mm.....	34
Figure 3. 13 Absolute maximum lengths of entangled copper wires: $L_e$ and real length of straight copper wires: $L_s$ ; (a) 10 mm (b) 30 mm (c) 50 mm.....	34
Figure 3. 14 Relationship between Entangle factor of copper wires [E.F.] and Real length of straight copper wire (mm) .....	35
Figure 4. 1 Photo of recovered product after jig separation .....	37
Figure 4. 2 Pictures of real sample in separation chamber; (a) before and (b) after jig separation .....	38
Figure 4. 3 Components of each layer after jig separation of real sample.....	38
Figure 4. 4 Jig products of real sample; (a) the 5 <sup>th</sup> layer and (b) the 6 <sup>th</sup> layer .....	39
Figure 4. 5 Example pictures of model samples in separation chamber; (a) before jig separation, (b) after jig separation, and (c) upon divided into 6 layers .....	40

Figure 4. 6 Pictures of jig product of model sample (without copper wire) at separation time variation; (a) 1 minute (b) 2 minutes (c) 3 minutes.....	40
Figure 4. 7 Components of each layer after jig separation of model samples (without copper wire) at separation time variation; (a) 1 minute (b) 2 minutes (c) 3 minutes.....	41
Figure 4. 8 Pictures of jig products of model samples with copper wires (10 % copper wires, 30 mm long copper wires) at separation time variation; (a) 1 minute (b) 2 minutes (c) 3 minutes.....	42
Figure 4. 9 Components of each layer after jig separation of model samples with copper wires (10 wt% copper wires, 30 mm long copper wires) at separation time variation; (a) 1 minute (b) 2 minutes (c) 3 minutes.....	42
Figure 4. 10 Pictures of jig products of model samples with copper wires (30 mm long copper wires, separation time 3 minutes) at mass ratio variation; (a) 5 % (b) 10 % (c) 15 % .....	43
Figure 4. 11 Components of each layer after jig separation of model samples with copper wires (30 mm long copper wires, separation time 3 minutes) at mass ratio variation; (a) 5 % (b) 10 % (c) 15 % .....	44
Figure 4. 12 Relationship between copper content in feed and copper recovery in 6 <sup>th</sup> layer .....	44
Figure 4. 13 Pictures of jig products of model samples with copper wires (10% copper wires, separation time 3 minutes) at lengths of copper wires variation; (a) 10 mm (b) 30 mm (c) 50 mm .....	45
Figure 4. 14 Components of each layer after jig separation of model samples with copper wires (10 % copper wires, 3 minutes) ) at lengths of copper wires variation; (a)10 mm (b) 30 mm (c) 50 mm .....	46
Figure 4. 15 Relationship between entanglement factor [E.F.] and copper recovery in the 6 <sup>th</sup> layer .....	46
Figure 4. 16 Entanglement phenomena during jig separation .....	47
Figure 5. 1 Mill pot; inner diameter 190 mm.....	49

Figure 5. 2 Sieve; aperture size 16 mm.....	50
Figure 5. 3 Outline of trommel .....	50
Figure 5. 4 Model sample; (a) before and (b) after pre-treatment by mill pot.....	51
Figure 5. 5 Pre-treatment product of model sample (+16 mm) .....	51
Figure 5. 6 Real sample; (a) before and (b) after pre-treatment by mill pot.....	53
Figure 5. 7 Pre-treatment product of real sample (+16 mm) .....	53
Figure 5. 8 Pictures of real sample (with pre-treatment) in separation chamber; (a) before and (b) after jig separation .....	54
Figure 5. 9 Components of each layer after jig separation of real sample; (a) without and (b) with pre-treatment.....	55
Figure 5. 10 Jig products of real sample (with pre-treatment); (a) 5 <sup>th</sup> layer and (b) 6 <sup>th</sup> layer .....	55
Figure 6. 1 Outline of special trommel .....	57
Figure 6. 2 Recycling flowchart of small home appliances (suggestive process) .	58

# CHAPTER I

## INTRODUCTION

### 1.1 General introduction

Nowadays, depletion of natural resources as well as increasing of resources demand and wastes generation is problem of concerns. Therefore, resources recovery and recycling have become important to cope with these concerns. In Japan, recycling laws for home appliances likes air conditioners, televisions, refrigerators, and washing machines were enacted in 2001 and then extended to automobiles and small home appliances such as video game and camera in 2005 and 2013 (Ito 2008), (Ito 2013).

Demand for electric and electronic equipment (EEE) is increasing around the world. Electronic devices like cell phones, televisions, computers and high-tech gadgets, which fall into this category, have very short lifetimes due to the rapid advances in computer technology; hence, the amount of wastes from electrical and electronic equipment (WEEE) or electronic wastes (e-wastes) generated per year is rapidly and steadily increasing each year. Management of e-wastes has recently become an important issue not only in developed countries, but also in majority of the developing countries including Thailand. In addition, telecom companies sometimes require their customers to change their devices/appliances in order to be compatible with the company's mode of transmission (e.g., analog to digital for TVs) and data transfer protocols (e.g., 2G to 3G for mobile phones). Due to these changes, the volume of e-wastes is expected to rapidly increase approximate 10% per year. Otherwise, the amount of e-wastes and their disposal would become a big problem to the local authorities (Khanchang 2014).

E-wastes have recently become considered as "urban ores" due to their high contents of valuable metals (i.e. platinum, gold, silver, copper, etc.) and plastics so that the e-wastes would be considered as secondary resources, not wastes for disposal anymore. Hence, some countries have enacted recycling laws for e-waste management.

In Europe, for example, the EU Directive on Waste from Electronic and Electronic Equipment (WEEE) outlined the reuse and recycling at least 65% of total weight of one appliance or IT equipment (Khanchang 2014). In Japan, a recycling law for home appliances like air conditioners, televisions, refrigerators, and washing machines was enacted in 2001. According to this recycling law, about 80% of total weight of one appliance is recycled. More recently, a new law was enacted in 2013 to recycle smaller home appliances such as video game consoles, cameras, and mobile devices (Ito 2013).

Prior to the passage of these recycling laws, it is fairly common for e-wastes to end up in landfills or be burnt in incinerators. Such processes could not recover valuable materials and the potential risks of releasing toxic substances such as cadmium (Cd), lead (Pb), and other hazardous materials into the air, soil, or water are relatively high (Khanchang 2014).

Jig separation, which is a well-known method for particle separation based on density differences, has become widely used in mineral processing due to its simplicity of operation, low cost, and high efficiency. It is still widely used in mineral processing, especially for coal cleaning, because of its high separation efficiency and cost effectiveness (Wills 2006). Recently, this physical separation technique has been successfully applied in the recycling of electronic wastes like home appliances and computers.

The RETAC jig (R&E, Co., Ltd.) used in this study is a modified TACUB (Takakuwa air chamber under bed) jig, commercially known as BATAC® jig. It is an excellent example of a jig that is well-suited for the separation of metal-plastic and plastic-plastic mixtures because of the “wave form” could be precisely controlled during operation (Ito 2010), (Hori 2009), (Tsunekawa 2005).

Tsunekawa et al. have applied this jig in pilot scale studies to separate kinds of plastic from plastic mixtures generated from scrapped copy machines. The copy machines were dismantled to recover valuable parts and then the residue was crushed and metals were firstly removed, the residue containing plastics was treated by jig separation to separate the different kinds of plastic. Good results were obtained under suitable conditions (purities: 99.8% PS, 99.3% ABS, and 98.6% PET) as the products

in the upper, middle, and bottom layers of the output of the jig separation, respectively (Tsunekawa 2005).

In this study, the RETAC jig was applied for recovery of valuable materials from end-of-life small home appliances. A variety of small home appliances were crushed and large pieces of metals were removed by magnetic and eddy current separation, then the residue containing plastics and small sized metal components was treated by jig separation. However, the separation efficiency was low due to copper wires containing in the small home appliances. Copper wires entanglement in the separation chamber causes decreasing in the recovery of valuable metals and the efficiency of plastics separation. Therefore, effects of copper wires on the jig separation were investigated by using model samples of plastics and copper wires mixture. The study found that the separation efficiency and recovery can be increased if the copper wires were removed by pre-treatment process prior jig separation.

## **1.2 Objectives of this study**

To study effects of copper wires on recovery of valuable metals and plastics from crushed end-of-life small home appliances by jig separation, and also to evaluate the possibility to increase separation efficiency and recovery of copper wires by pre-treatment prior jig separation.

## **1.3 Scope and outline of this study**

The scope of this study is to investigate the effects of copper wires on the recovery of valuable metals and plastics from crushed end-of-life small home appliances by jig separation and to develop the effective method to increase the separation efficiency and recovery by using real and model samples.

The schematic flowchart of this study is shown in figure 1.1.

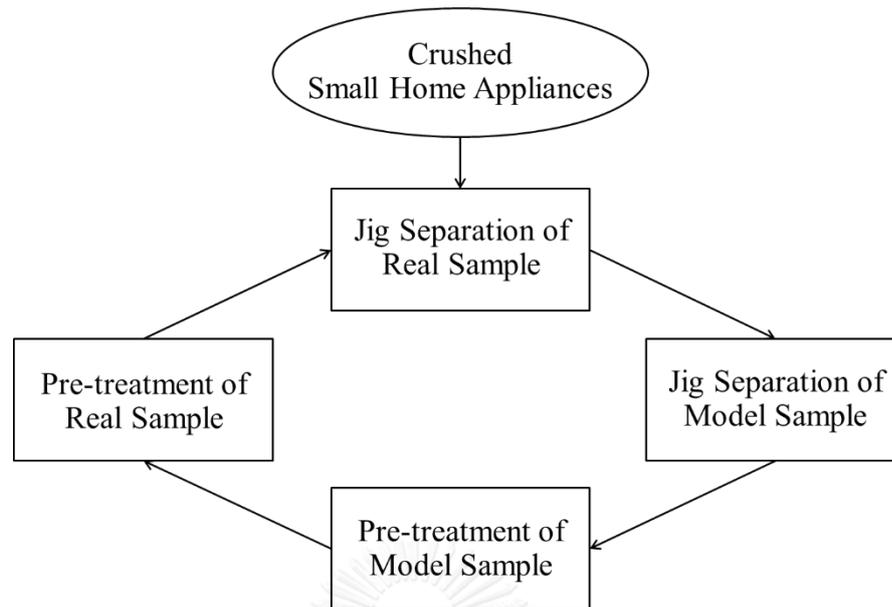


Figure 1. 1 Schematic flowchart of the present study

#### 1.4 Expected benefits

Result of this study is expected to be applied for industrial resources recycling processes of small home appliances wastes and also to increase purity of the separated products (valuable metals and plastics) and can be one solution to reduce the environmental issues.

#### 1.5 Order of presentation

In order to present this research and make it easy to understand for readers, the author has divided this research into the following 6 chapters;

**Chapter 1 Introduction:** General introduction, objectives of this study, scope and outline of this study, and expected benefits.

**Chapter 2 Backgrounds and literature reviews:** Description of small home appliances, jig separation technology, recovery and recycling of end-of-life small home appliance, and literature reviews.

**Chapter 3 Characterization of samples:** Characterization of real sample and characterization of real sample model samples.

**Chapter 4 Effects of copper wires on jig separation:** Jig separation of real sample and jig separation model samples.

**Chapter 5 Development of pre-treatment method:** Pre-treatment of model samples and pre-treatment of real sample.

**Chapter 6 Conclusion:** Conclusion and recommendation.



## CHAPTER II

### BACKGROUNDS AND LITERATURE REVIEWS

#### 2.1 Description of small home appliances

There are so many definitions of e-wastes. Small home appliance, small household appliance, or small appliance is one kind of e-wastes. The small home appliances also have many descriptions depend on the definition of e-wastes. Examples of e-waste definition for are as follow.

##### **WEEE (Waste from Electrical and Electronic Equipment)**

In Europe, the EU Directive on Waste from Electronic and Electronic Equipment (WEEE) outlined the reuse and recycling of at least 65% of total weight of one appliance or IT equipment (Khanchang 2014).

The first WEEE Directive (Directive 2002/96/EC) entered into force in February 2003. In December 2008, the European Commission proposed to revise the Directive in order to tackle the fast increasing waste stream. The new WEEE (Directive 2012/19/EU) entered into force on 13 August 2012 and became effective on 14 February 2014. (Exexutive 2015), (Information 2015), (LTD 2015).

There are ten broad categories of WEEE currently outlined within the regulations, namely:

**Category 1: Large household appliances** e.g. microwaves, electric heating appliances, electric radiators, electric fans, air conditioner appliances, large cooling appliances, refrigerators, freezers, washing machines, clothes dryers, dish washing machines, cookers, electric stoves, electric hot plates, and other large appliances used for heating rooms, beds, seating furniture, fanning, exhaust ventilation and conditioning equipment, refrigeration, conservation, storage of food, cooking, and other processing of food.

**Category 2: Small household appliances** e.g. vacuum cleaners, carpet sweepers and other appliances for cleaning, appliances used for sewing, knitting, weaving and other processing for textiles, irons and other appliances for ironing, mangling and other care of clothing, toasters, fryers, grinders, coffee machines and equipment for opening or sealing containers or packages, electric knives appliances for hair-cutting, hair drying, tooth brushing, shaving, massage and other body care appliances, scales, clocks, watches and equipment for the purpose of measuring, indicating or registering time.

**Category 3: Information technology (IT) and telecommunications equipments** e.g. centralized data processing, mainframes, minicomputers, printer units, personal computers (CPU, mouse, screen and keyboard included), laptop computers (CPU, mouse, screen and keyboard included), notebook computers, printers, copying equipment, electrical and electronic typewriter, pocket and desk calculators and other products and equipment for the collection, storage, processing, presentation or communication of information by electronic means, user terminals and systems, facsimile, telex, telephones (including pay telephones, cordless telephones & mobiles), answering systems.

**Category 4: Consumer equipments** e.g. radios set, televisions set, hi-fi equipment, camcorders, video cameras, video recorders, audio amplifiers, electric toothbrushes, musical instruments, and other products or equipment for the purpose of recording or reproducing sound or images, including signals or other technologies for the distribution of sound and image than by telecommunications.

**Category 5: Lighting equipment** e.g. straight and compact fluorescent tubes and high intensity discharge lamps including pressure sodium lamps and metal halide lamps, low pressure sodium lamps, luminaires for fluorescent lamps with the exception of luminaires in households, and other lighting or equipment for the purpose of spreading or controlling light with the exception of filament bulbs.

**Category 6: Electrical & electronic tools** (except large-scale stationary industrial tools) e.g. drills, saws and sewing machines, electric lawnmowers, equipment for turning, milling, sanding, grinding, sawing, cutting, shearing, drilling, making holes, punching, folding, bending or similar processing of wood, metal and other materials, tools for riveting, nailing or screwing or removing rivets, nails, screws or similar uses, Tools for welding, soldering or similar use, equipment for spraying, spreading, dispersing or other treatment of liquid or gaseous substances by other means, and tools for mowing or other gardening activities.

**Category 7: Toys, leisure and sports equipment** e.g. electric trains or car racing sets, games consoles, sports equipment with electric or electronic components, coin slot machines, and computers for biking, diving, running, rowing, etc.

**Category 8: Medical devices** (with the exception of all implanted and infected products) e .g. dialysis machines, analyzers, medical freezers and cardiology, pulmonary ventilators, nuclear medicine, fertilization tests, radiotherapy equipment, Laboratory equipment for in-vitro diagnosis, and other appliances for detecting, preventing, monitoring, treating, alleviating illness, injury or disability.

**Category 9: Monitoring and control instruments** e .g. smoke detectors, thermostats, heating regulators, Measuring, weighing or adjusting appliances for household or as laboratory equipment, and other monitoring and control instruments used in industrial installations (e.g. in control panels).

**Category 10: Automatic dispensers** e.g. hot drinks, hot or cold bottles or cans, solid products, money automatic dispensers and all appliances which deliver automatically all kind of products.

Scope of the regulations will be extended from January 2019 to cover further categories of electric and electronic equipment (EEE).

### South Korea government regulation

In South Korea, e-wastes are categorized by weight as shown in table 2.1. If less than 5 kg is categorized as small household appliances; if 5 – 10 kg is categorized as medium household appliances, and large household appliances if more than 10 kg (Choi 2015).

Table 2. 1 E-waste categories according to the South Korea government regulation

Category	Criteria	Items
Small household appliances	< 5kg	Electric iron, Blender, Elec. fan, Humidifier, Video player
Medium household appliances	5 ~ 10kg	Elec. cooker, Vacuum cleaner, Air cleaner, Bidet, Elec. Heater
Large household appliances	> 10kg	Dish washer, Microwave oven, Elec. oven, Water purifier, Garbage disposal

### Japanese Definition

In Japan, e-wastes are categorized by size. Home appliances (such as air conditioners, televisions, refrigerators, and washing machines), and small home appliances (such as laptops, video game consoles, cameras and mobile devices) as shown in figure 2.1 (Ito 2013).

For this study, Japanese definition was used.

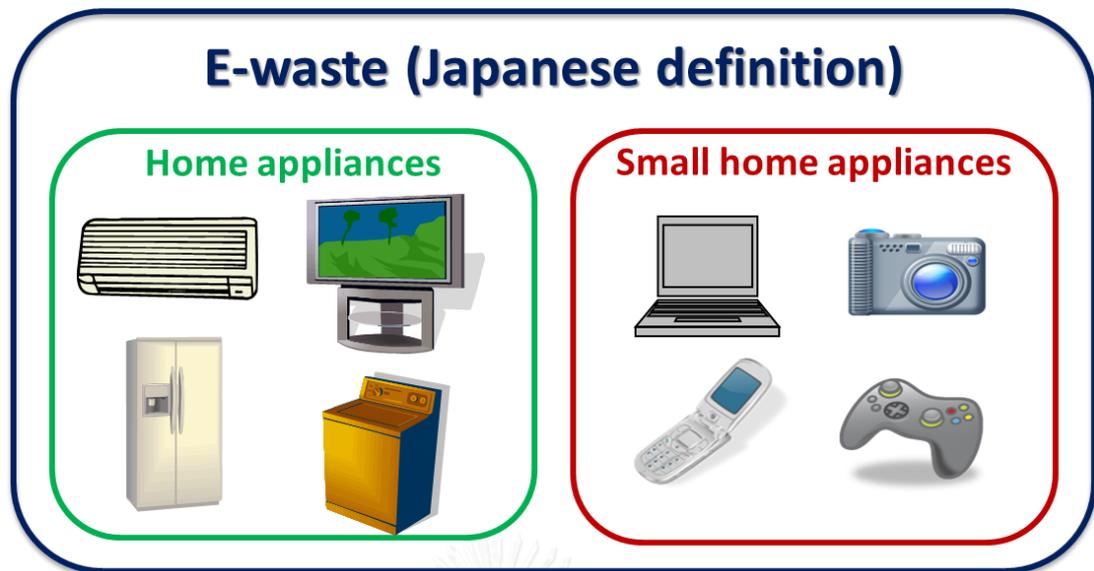


Figure 2. 1 E-waste category as Japanese definition

## 2.2 Jig separation technology

Jig separation, which is a well-known method to separate particles based on density differences, has become widely used in mineral processing due to its simplicity of operation, low cost, and high efficiency. It is still widely used in mineral processing especially for coal cleaning because of its high separation efficiency and cost effectiveness (Will 2006).

Jig is normally used to concentrate relatively coarse material and, if the feed is moderately closed sized (3-10 mm), it is not difficult to achieve good separation of a moderately narrow specific gravity range of materials in the feed. When the specific gravity difference is large, good concentration is possible with a wider size range. Many large jig circuits are still used in the coal, cassiterite, tungsten, gold, and iron-ore industries.

In the jig, gravity separation is performed in a bed by a pulsating current of water to produce stratification. The aim is to enlarge the bed being treated and to control the expansion so that the heavier, smaller particles seep the space of the bed and the larger high specific gravity particles fall under a condition probably like hindered settling.

On the pulsion stroke the bed is lifted, then the velocity decreases it tends to enlarge, the bottom particles falling first until the bed is loosened. On the suction stroke, it then closes slowly again and this is repeated at every stroke, the frequency usually varying between 55 and 330 cycles/min.

The initial acceleration of the particles grains is thus independent on size but depending on densities of solid and fluid only. In theory, if the falling time is short and the repeating frequent enough, the total distance travelled by the particles will be more affected by the differential initial acceleration, and by density, than by their terminal velocities and by size. In other words, to separate small heavy mineral particles from large light particles a short cycle is needed. Although relatively short fast strokes are used to separate fine minerals, more control and better stratification can be achieved by using longer, slower strokes, especially with the coarser particle sizes. Therefore, screening before jig separation and treat the different size separately is better.

If the particles are examined after a longer time, they would reach their terminal velocities and will be moving at a rate depending on their size and specific gravity. Due to the bed is loosely packed mass with interstitial water providing a very thick suspension of high density, hindered-settling conditions control, and the settling ratio of heavy to light material is higher than free settling. The upward flow can be adjusted overcomes the downward velocity of the fine light particles and take them away. It can be increased large heavy particles settle only, but it is clear that it will not be possible to separate the small heavy and large light particles of similar terminal velocity. Hindered settling has an effect on the coarse particles separation, for which longer, slower strokes should be used, although in practice, with coarser feeds, it is impossible that the larger particles have time to reach their terminal velocities.

At the end of a pulsion stroke, as the bed begins to compact, the larger particles interlock, allowing the smaller grains to move downwards through space. The fine grains may not settle as quickly during this consolidation trickling phase as during the initial acceleration, but if consolidation trickling can be made to last long enough, the effect, especially in the fine heavy materials recovery, can be significant. Idealized jig separation process by the described phenomena is shown in figure 2.2.

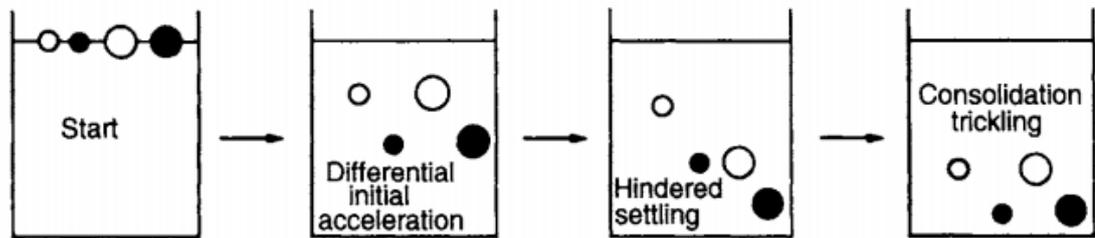


Figure 2. 2 Idealized jig separation process

In the jig, the pulsating water currents are caused by a piston having a movement which is a harmonic waveform. Figure 2.3 shows the movement of the piston in a jig.

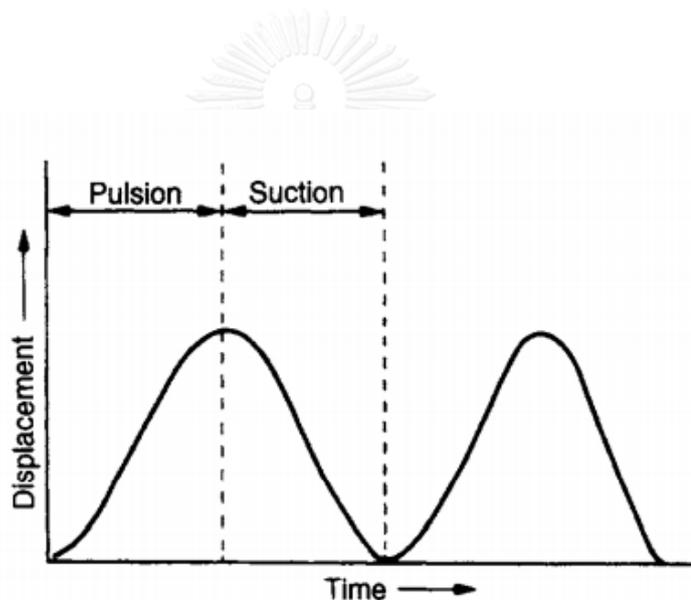


Figure 2. 3 Movement of the piston in a jig

The vertical speed of flow through the bed is equivalent to the speed of the piston. The speed of flow through bed during jig cycle is shown in figure 2.4. The upward speed of flow increases after point A, the beginning of the cycle. As the speed increases, the grains will be loosened and the bed will be forced open, or enlarge. At point B, the grains are in the phase of hindered settling in an upward flow, and due to the speed of flow from B to C still increases, the fine grains are pushed upwards by the flow. At point D, first the coarser grains and later on the remaining fine grains will fall back. Due to the combination of initial acceleration and hindered settling, it is mainly

the coarser grains that will lie at the bottom of the bed. At the point of transition between the pulsion and the suction stroke, at point E, the bed will be compacted. Consolidation trickling can now happen. In closely sized particles, the heavy grains can seep difficultly through the bed and might be lost to the tailings.

The jig bed consists of a layer of coarse, heavy particles, or ragging, placed on the jig screen. The feed flows across the ragging and the separation takes place in the jig bed so that grains with a high specific gravity seep through the ragging and screen to be the concentrate while the light grains are take away by water flow to be the tailings. The basic jig construction is shown figure 2.5.

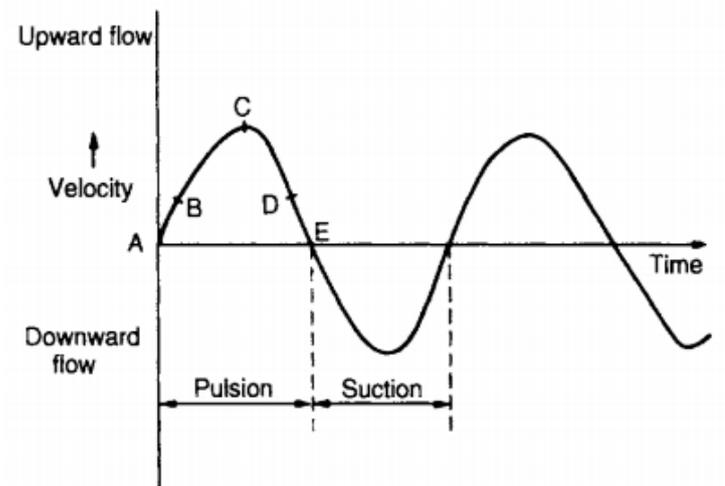


Figure 2. 4 Speed of flow through bed during jig cycle

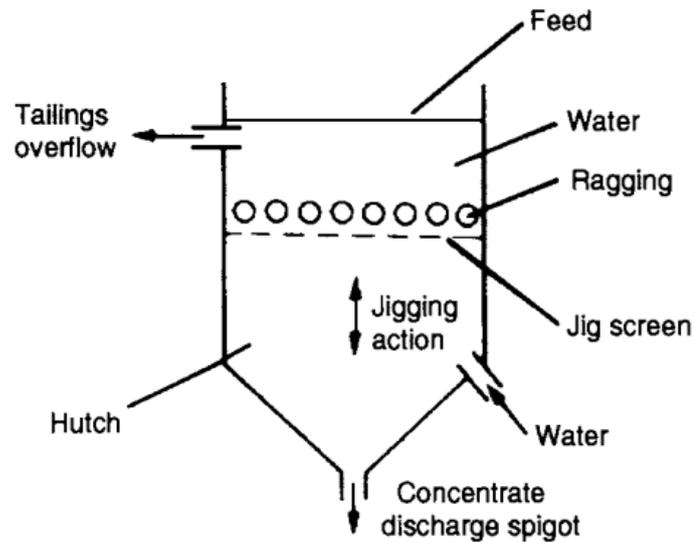


Figure 2. 5 Basic Jig Construction

### Advance jig separation technology

In this study, advance jig separation technology was used. Jig are ideally suited and widely used in coal cleansing because coal is lighter than its accompanying gangue minerals, commonly quartz and alumina-silicates. A number of distinct jig separators have been developed.

Among these, the TACUB (Takakuwa air chamber under bed) Jig, commercially marketed as the BATAAC® jig, was developed by Professor emeritus K. Takakuwa in Laboratory of Mineral Processing and Resources Recycling, Division of Sustainable Resources Engineering, Graduate School of Engineering, Hokkaido University, Japan, renowned in the mineral processing and coal cleansing industries for its high efficiency and reliability. The outline of BATAAC jig is shown in figure 2.6. More recently, Professor emeritus M. Tsunekawa in the same laboratory modified the TACUB jig to RETAC jig for recycling processes. The outline of a desktop type batch-wise RETAC jig is shown in figure 2.7.

RETAC jig is an excellent example of a jig that is well-suited for the separation of metal-plastic and plastic-plastic mixtures because of the “wave form” could be precisely controlled during operation (Ito 2010), (Hori 2009), (Tsunekawa 2005). The separation process of RETAC jig is shown in figure 2.8.

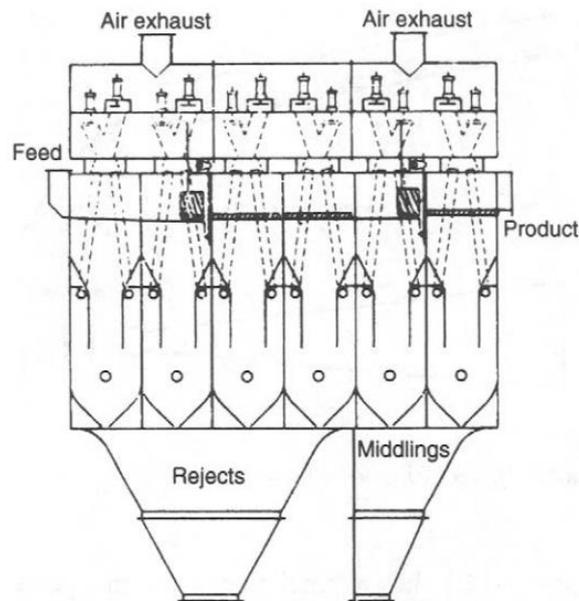


Figure 2. 6 Outline of BATAAC jig

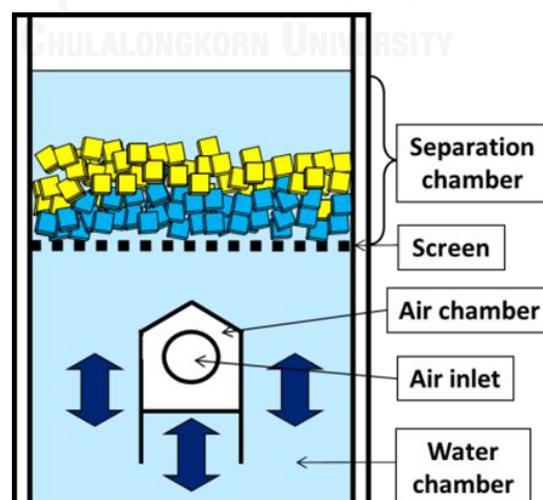


Figure 2. 7 Schematic outline of the desktop type batch-wise RETAC jig

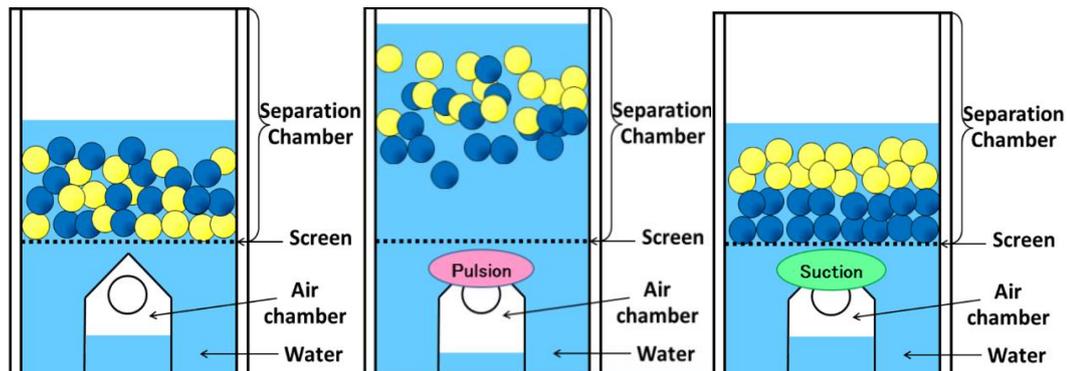


Figure 2. 8 Separation process of RETAC jig

The reverse jig (R&E, Co., Ltd.) is a modified RETAC jig (a top screen has been added to the chamber) designed to separate particles having lighter densities than water. Figure 2.9 shows an outline of the reverse jig where particles move up and down under the top screen and the particles are separated based on differences in levitation velocity.

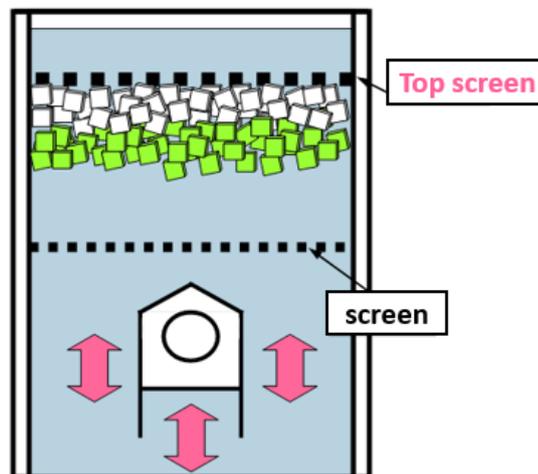


Figure 2. 9 Schematic outline of the desktop type batch-wise reverse jig

The hybrid jig (R&E, Co., Ltd.) is a modified RETAC jig designed to separate particles having similar densities but different wettability. Figure 2.10 shows an outline of the hybrid jig. An aeration tube was installed under the screen (particle bed) and an air pump connected to the tube to bubble air into the water chamber. When

bubble attach on particles the apparent density becomes lower and the hybrid jig can separate particles using the apparent density differences.

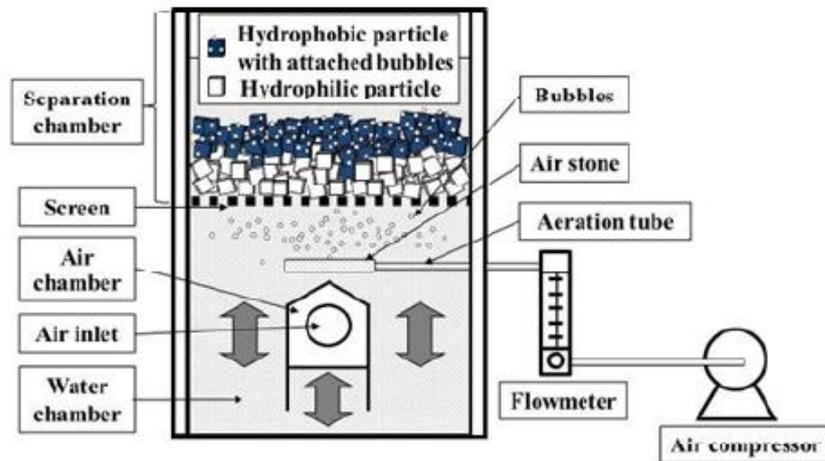


Figure 2. 10 Schematic outline of the desktop type batch-wise hybrid jig

The reverse hybrid jig (R&E, Co., Ltd.) is a modified hybrid jig (a top screen has been added to the chamber) designed to separate particles having lighter densities than water, which having similar densities but different wettability. Figure 2.11 shows an outline of the reverse hybrid jig.

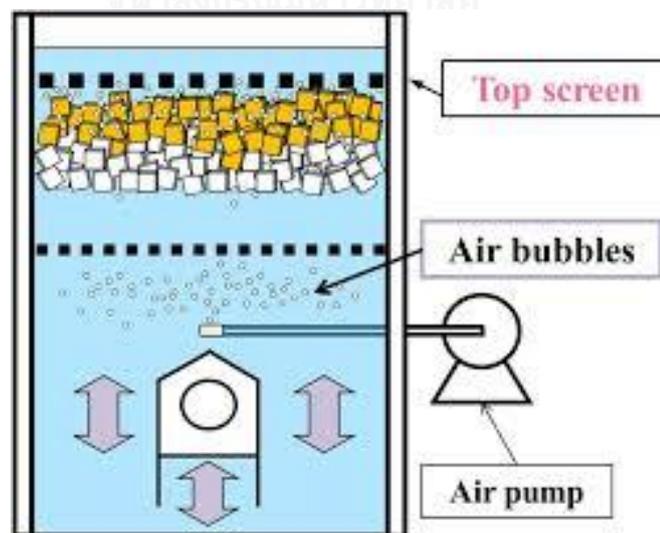


Figure 2. 11 Schematic outline of the desktop type batch-wise reverse hybrid jig

The continuous type RETAC jig (R&E, Co., Ltd.) is a modified RETAC jig designed to separate particles continuously. Figure 2.12 shows an outline of the continuous type RETAC jig.

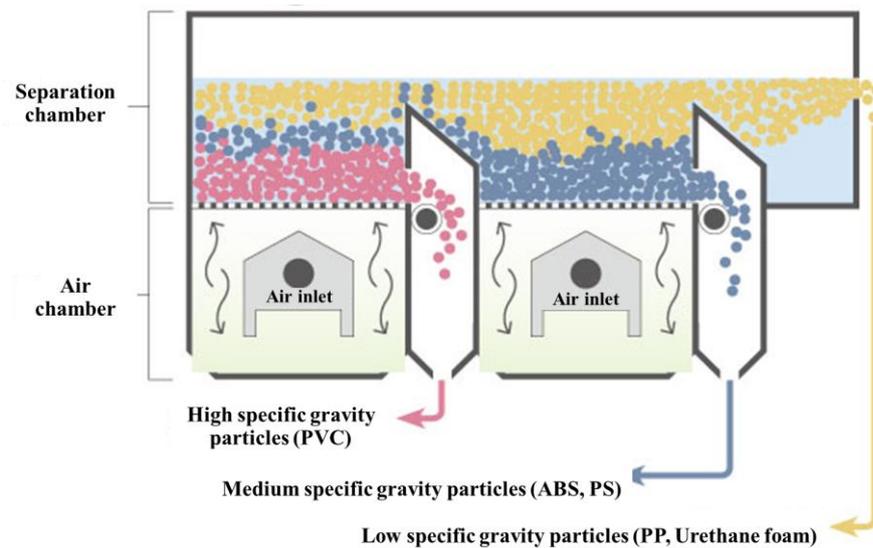


Figure 2. 12 Schematic outline of the continuous RETAC jig

The laboratory scale (batch and continuous type) and pilot scale RETAC jig are shown in figure 2.13 and 2.14



Figure 2. 13 Laboratory scale RETAC jig; batch (left) and continuous type (right)



Figure 2. 14 Pilot scale RETAC jig

In this study, a desktop type batch-wise RETAC jig, which a 145 mm long, 155 mm wide, and 320 high separations chamber was used for the experiment.

### 2.3 Recovery and recycling of end-of-life small home appliances

According to small home appliances recycling law in Japan, Re-Tem Corporation, copy machine and ATM recycling company tried to recycle the end-of-life small home appliances by using the copy machine and ATM recycling process. The recycling flowchart of small home appliances of Re-Tem Corporation (Pilot scale) is shown in figure 2.15. After dust removal of crushed end-of-life small home appliances, coarse magnetic matters were removed by magnetic separation and coarse metals were removed by eddy current separation. There are three groups of the mixture of plastics and metals products; coarse, medium, and fine groups. The mixture products of small home appliances recycling process are shown in figure 2.16.

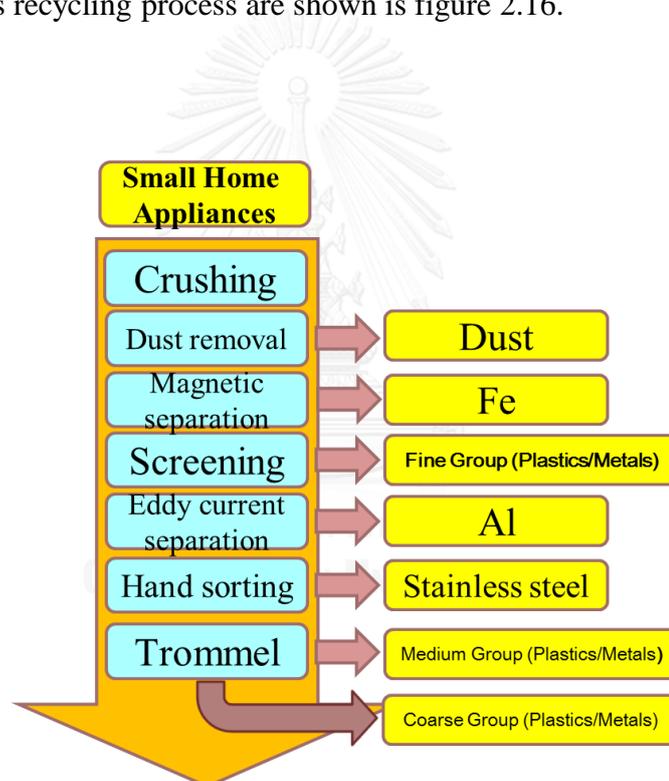


Figure 2. 15 Recycling flowchart of small home appliances of Re-Tem Corporation (Pilot scale)



Figure 2. 16 Mixture of plastics and metals products;  
 (a) coarse group, (b) medium group, and (c) fine group

Particle size distribution of the mixture products are shown in figure 2.17 and the chemical composition of mixture products are shown in figure 2.18.

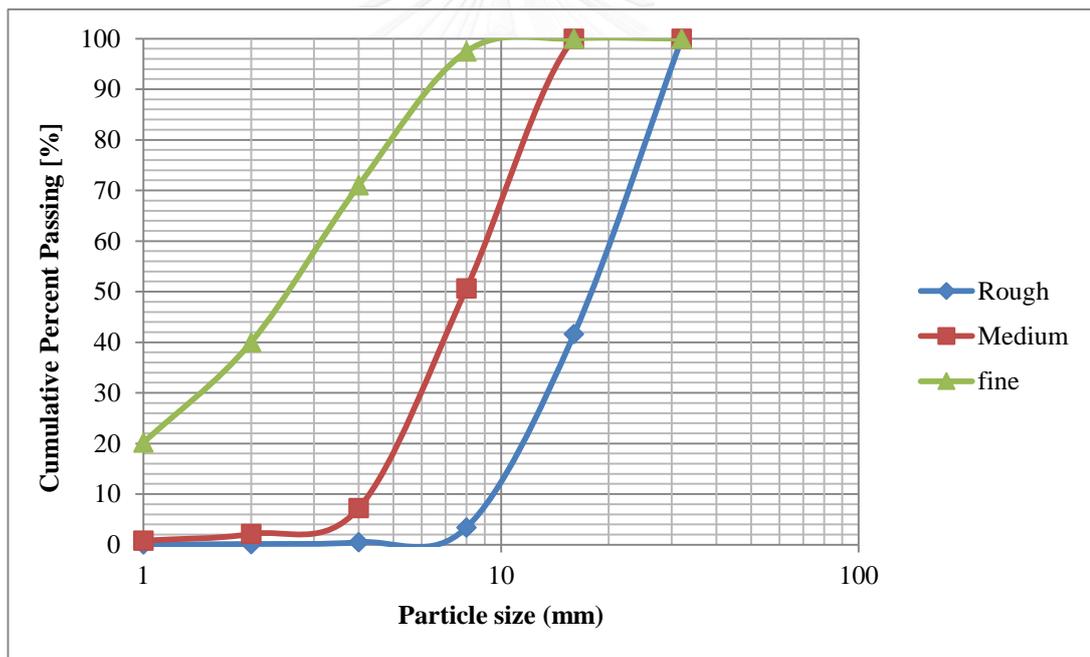


Figure 2. 17 Particle size distributions of the mixture products

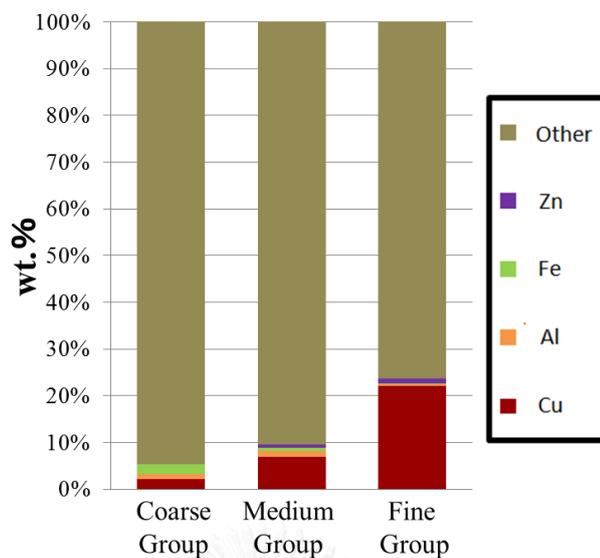


Figure 2. 18 Chemical compositions of mixture products

In the previous study, the RETAC® jig was utilized to recover valuable metals and plastics from these mixtures. The results of jig separation are shown in figure 2.19 and Table 2.2. They can recover the copper from coarse and medium group 91% and 96% in order but for a fine group is only 66%. This is because copper wires in a fine group easily get entangled in the separation chamber during jig separation, which trap plastic and PCB particles preventing their motion in the chamber.

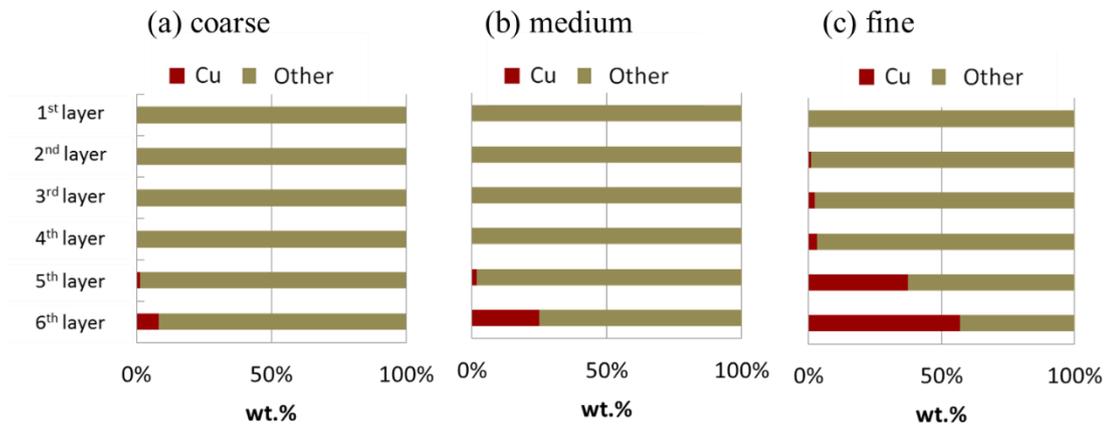


Figure 2. 19 Components of each layer after jig separation;  
(a) coarse group, (b) medium group, and (c) fine group

Table 2. 2 Recovery and purity of copper in the 6<sup>th</sup> layer of the mixture products

The mixture products	Copper recovery in 6 <sup>th</sup> layer	Copper purity in 6 <sup>th</sup> layer
Coarse group	91%	8%
Medium group	96%	25%
Fine group	66%	57%

To investigate the effects of this entanglement, this study evaluated the effects of factors like length and amount of copper wires using model samples of plastics and copper wires. To study effects of copper wires on recovery of valuable metals and plastics from crushed end-of-life small home appliances by jig separation and also evaluate the possibility to increase separation efficiency and recovery of copper wires by pre-treatment before jig separation.

## 2.4 Literature reviews

M. Tsunekawa et al., 2005 studied about jig separation of plastics from scrapped copy machines. A TACUB jig was applied to separate waste plastics used in the copy machine. The three plastics, polystyrene (PS), acrylonitrile butadiene styrene (ABS) and polyethylene terephthalate (PET), were hand-picked from copy machines and cut, then ground and sieved to obtain 3.5–10.0 mm fraction. Specific gravities of PS, ABS and PET were found to be 1.06, 1.18 and 1.71, respectively. Effect of water pulsation including amplitude and frequency on the separation performance was investigated for the feeds containing three plastics. Good results were obtained under suitable conditions (amplitude; 10 cm, frequency; 10 cycles/min, bed thickness; 6 cm, and total pulsation number; 100). Purities of 99.8% PS, 99.3% ABS, and 98.6% PET were recovered as the products in the top, middle and bottom layers, respectively (Tsunekawa 2005).

K. Hori et al., 2009 studied about optimum water pulsation of jig separation for crushed plastic particles. The authors applied jig technology by using a TACUB jig to separate plastics used in electrical appliances. The plastics used were crushed waste plastics: two types of burn resistant polyethylene (green PE and gray PE) and polyvinyl chloride (red PVC) of non-spherical random shape. The particles size was 0.5–3 mm and the specific gravities of the two kinds of PE and PVC were about 1.1, 1.3 and 1.4, respectively. The jig separation experiments were carried out under various water pulsations, at which the amplitude, frequency, and pattern of pulsation were varied. High-grade PE and PVC products over 99.8% were recovered under pulsations of small frequency and amplitude than that for coarser plastics (Hori 2009).

M. Ito et al., 2013 studied about recycling treatment of cell phones including PCBs by advanced jigging. A RETAC jig (R&E, Co., Ltd.) was applied to separate metals and plastics from mixtures of crushed plastics and PCBs. The PCBs and plastics were crushed by an Orient mill (plastic cutting mill). Samples were treated by a RETAC jig (2 cm of displacement, 30 cycles/min, separation time 3 min, and ragging material alumina balls). Products are separated into several layers from the top to bottom and the ragging layer is also recovered as a mixture of the ragging material and heavy products. All samples were dried at 40°C in a drying oven for 24 h and the products

were ground for analysis by X-ray fluorescence analysis (XRF). Low temperature grinding with liquid nitrogen was used and grinding was carried out with disc-mill (1400 rpm, for 2 min). The PCBs were recovered as bottom layer products and were separated from other plastics. Jig separation of crushed PCBs was also carried out and Cu was recovered as bottom products and base materials containing Br were recovered in the middling layers and plastic matter with low Br content could be recovered as upper layer product (Ito 2013).

M. Ito et al., 2015 studied about the recovery of valuable components from crushed mobile phones using jig separation. A RETAC jig (R&E, Co., Ltd.) was applied to separate metals and plastics from of crushed mobile phone. Discarded mobile phones were crushed by cutting mill after batteries were removed. The samples were sieved to obtain particle size +2.0 -10.0 mm. The jig separation tests were carried out with a water pulsation of 2 cm displacement and 30 cycles /min frequency for 3 min. Aerosol OT (AOT, 15 ppm) as wetting agent was added to make the surface of samples more hydrophilic. For hybrid jig experiment, bubble generator was used at aeration flow 20 ml/ min, 20 ppm of Methyl Isobutyl Carbinol (MIBC) was added as a frother for bubble stabilization, and the effects of Calcium Lignosulfonate (CaLS) were investigated. After jiggling, products were divided into 6 layers. Specific gravities were determined by ultra pycnometer and copper concentrations were measured by XRF. The results showed that hybrid jig can recover the copper better than the normal jig. Overall, copper recovery by the hybrid jig with and without CaLS both were about 90% (Ito 2015).

W. Choi, E. Park, and S. Kang, 2015 studied about status and prospects of plastics recycling of used small household appliances. Optical sorting like near-infrared ray (NIR) sensors was used for separating plastics by resin type from crushed plastic mixtures of small e-wastes at high throughputs. The ABS and PS particles with white color are effectively recovered by application NIR system from the plastic mixtures. The ratio of black particles in the final product after NIR separation was increased to 38%. The overall recycling rate of small e-waste was about 70% (Choi 2015).

## CHAPTER III

### CHARACTERIZATION OF SAMPLES

#### 3.1 Characterization of real sample

##### 3.1.1 Experimental

Real sample of small home appliances was provided by Re-Tem Corporation, Japan. This company recovers valuable matters such as metals and plastics from e-waste mixture of ATM and copy machines. Small home appliances are also treated by similar process with the e-waste mixture. After feed materials are crushed and dust removal, coarse metals are separated by each technology, iron likes matter by magnetic separation, aluminum by eddy current separation, and stainless steel by hand sorting.

Residue of the process was sieved to separate with three size fractions containing the remaining metals and plastics: course, medium, and fine. The fine fraction contains significant amounts of copper wires and used in this experiment for further separation.

Figure 3.1 shows the crushed end-of-life small home appliances sample (the fine fraction).



Figure 3. 1 Crushed end-of-life small home appliances sample  
(the fine fraction)

- **Sieve Analysis**

The sieve aperture size 1, 2, 4, 8, and 16 mm was used for particle size distribution analysis. The +1.0-16 mm fraction was used for jig tests.

- **Chemical Composition Analysis**

Hand sorting was carried out to check the component of the sample. After the hand sorting, the sample was separated into three groups: copper wires, other metals, and plastics as shown in figure 3.2. Other metals were analyzed by XRF and the plastics were ground at low temperature chilled with liquid nitrogen, and then fed to microwave digestion process using aqua regia (the mixture of nitric acid and hydrochloric acid in a volume ratio of 1:3) followed with ICP-AES analysis. The analysis flowchart of real sample, ONE TPH-02 type low temperature grinding equipment, and microwave digestion equipment (ETHOS PLUS) are shown in figure 3.3, 3.4, and 3.5.



Figure 3. 2 Three fractions of real sample after hand sorting;  
(a) copper wires, (b) others metals, and (c) plastics & PCB

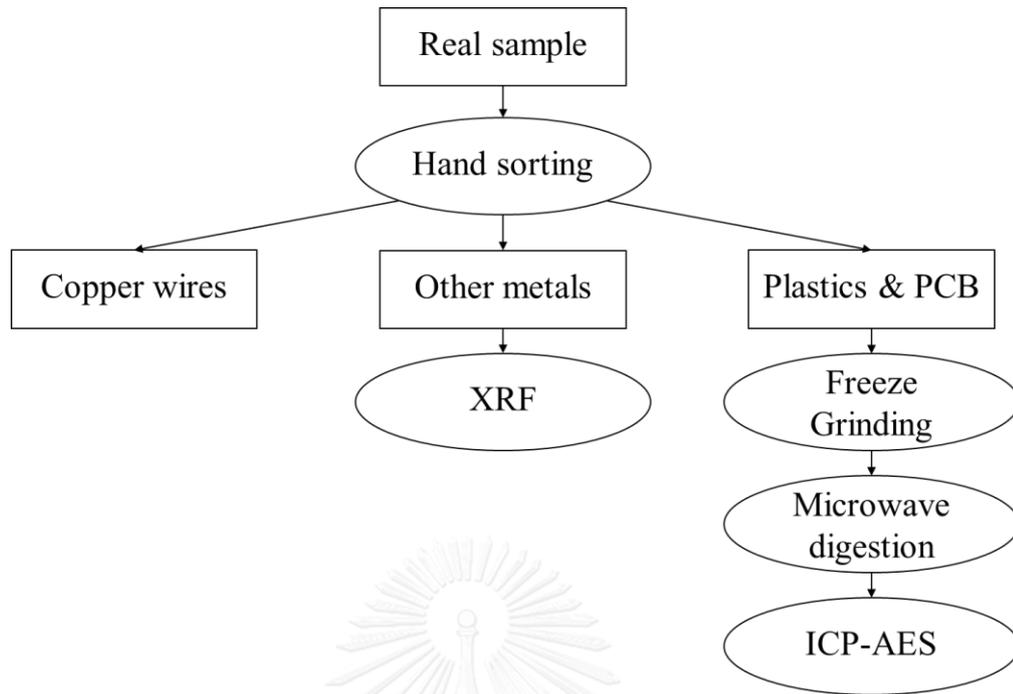


Figure 3. 3 Analysis flowchart of real sample



Figure 3. 4 ONE TPH-02 type low temperature grinding equipment



Figure 3. 5 Microwave digestion equipment (ETHOS PLUS)

### 3.1.2 Results and discussion

#### - Particle size distribution

The particle size distribution graph is shown figure 3.6. From graph, the  $d_{50}$  of this sample is about 3 mm.

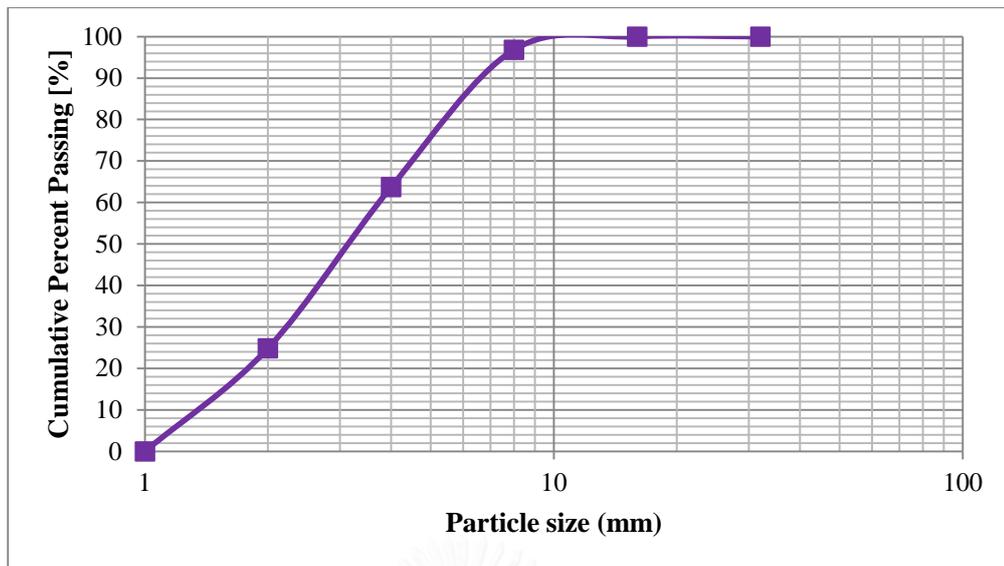


Figure 3. 6 Particle size distribution of the real sample

### - Composition of sample

The composition of the fine fraction generated from end-of-life small home appliances treatment is shown in figure 3.7. This sample contains plastics and fine metal parts, with copper as the main component.

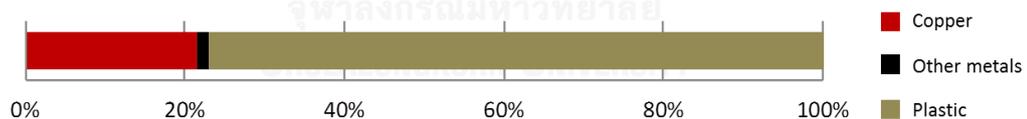


Figure 3. 7 Chemical composition of the real sample

## 3.2 Characterization of model samples

### 3.2.1 Experimental

Commonly, small home appliances have acrylonitrile butadiene styrene (ABS), polyvinyl chloride (PVC), and Polypropylene (PP) as the major plastics component. PP can be easily removed by sink-float separation by water because PP has a density less than water as known as Polyolefin. According to these reasons, model samples were made from a mixture of ABS (specific gravity 1.03) and PVC (specific gravity 1.31) at

1:1 ratio (amount 500 g), and copper wires (specific gravity 8.96). Actually, to make the model samples can be used instead of real sample, the materials contents of samples should be similar, this study plastics and entangled copper wires similar to the real sample but used plastics mixture ratio 1:1 because this condition easy to investigate the separation efficiency in jig separation.

The plastic samples were crushed by orient mill, the kind of cutting mill and then sieved to obtain the +2.0-10 mm fraction. The orient mill is shown in figure 3.8. The copper wires (diameter 0.4 mm, bent by hand) were mixed by varying mass ratios (0, 5, 10, and 15% of the total samples) and lengths (10, 30, 50 mm). The mixture of model samples are shown in figure 3.9 and 3.10.



Figure 3. 8 Orient mill



Figure 3. 9 Mixture of model samples; (a) ABS, (b) PVC, and (c) copper wires



Figure 3. 10 Model sample

- **Sieve analysis**

The sieve aperture size 2.0, 2.8, 4.0, 5.6, 6.7, 8.0, and 10 mm was used for particle size distribution analysis. The +2-10 mm fraction was used for jig tests.

- **Image analysis**

The microscope was used for image analysis. Image analysis was carried out to check the absolute maximum lengths of entangled copper wires.

### 3.2.2 Results and discussion

#### - Particle size distribution

The particle size distribution graph is shown figure 3.11. From graph, the  $d_{50}$  of the samples are also about 3 mm. Due to these results, the model samples can be used instead of real sample.

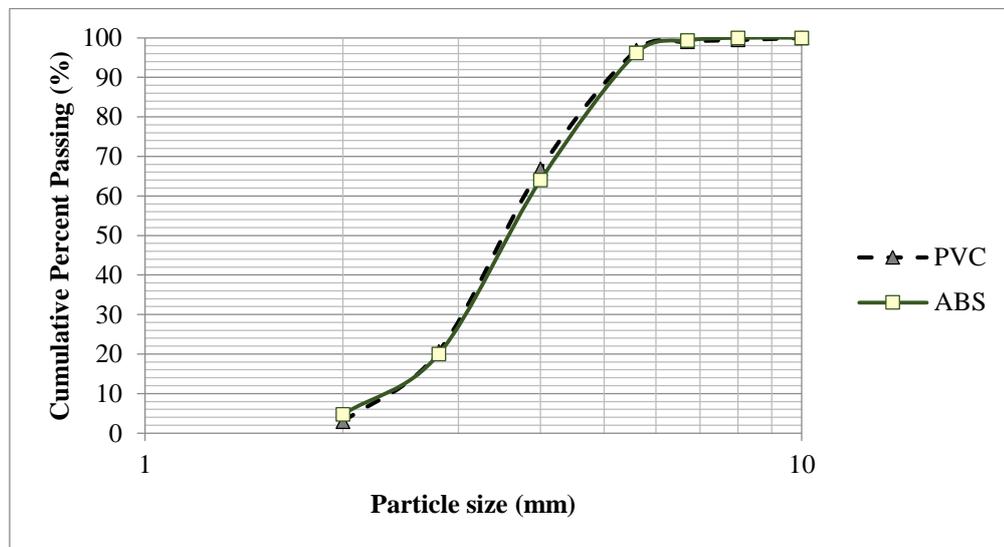


Figure 3. 11 Particle size distributions of the plastics mixture in model samples

#### - Entanglement factor of copper wires

Absolute maximum lengths of entangled copper wires were checked by Image analysis and Entangle factor of copper wires were calculated by Equation 3.1.

$$E.F. = \frac{L_e}{L_s} \quad (3.1)$$

E.F. : Entanglement factor of copper wires

$L_e$  : Absolute maximum length of entangled copper wires

$L_s$  : Real length of straight copper wires

The results are between 0 - 1, the less value of entanglement factor of copper wires is more entanglement. The pictures 10, 30, 50 mm copper wires are shown in figures 3.12 and 3.13. The absolute maximum length of entangled copper wires and entanglement factor of 10, 30, 50 mm copper wires are 9.64, 18.20, and 27.33 mm and 0.96, 0.61, and 0.55 in order. Figure 3.14 shows the relationship between entangle factor of copper wires and real length of straight copper wire. From the graph, entangle factor of copper wires decreases with longer length of straight copper wires.

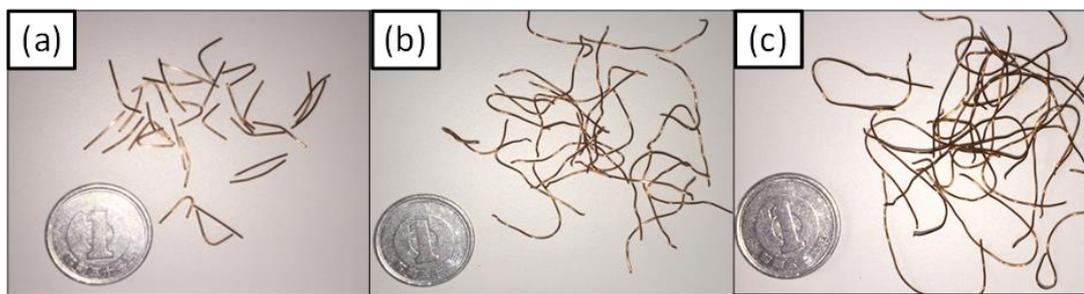


Figure 3. 12 Copper wires in the model samples; lengths  
(a) 10 mm, (b) 30 mm, and (c) 50 mm.

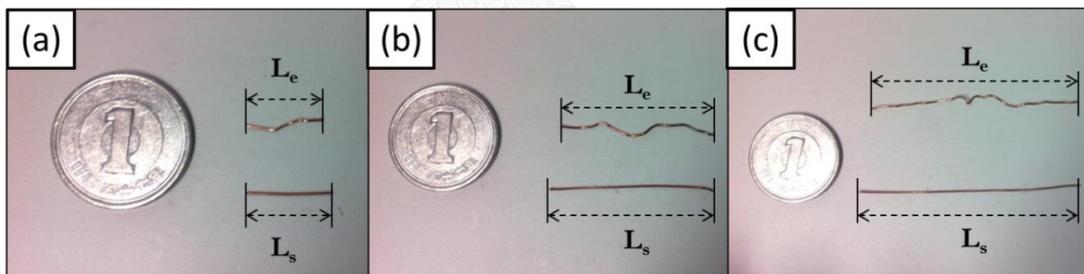


Figure 3. 13 Absolute maximum lengths of entangled copper wires:  $L_e$   
and real length of straight copper wires:  $L_s$ ; (a) 10 mm (b) 30 mm (c) 50 mm

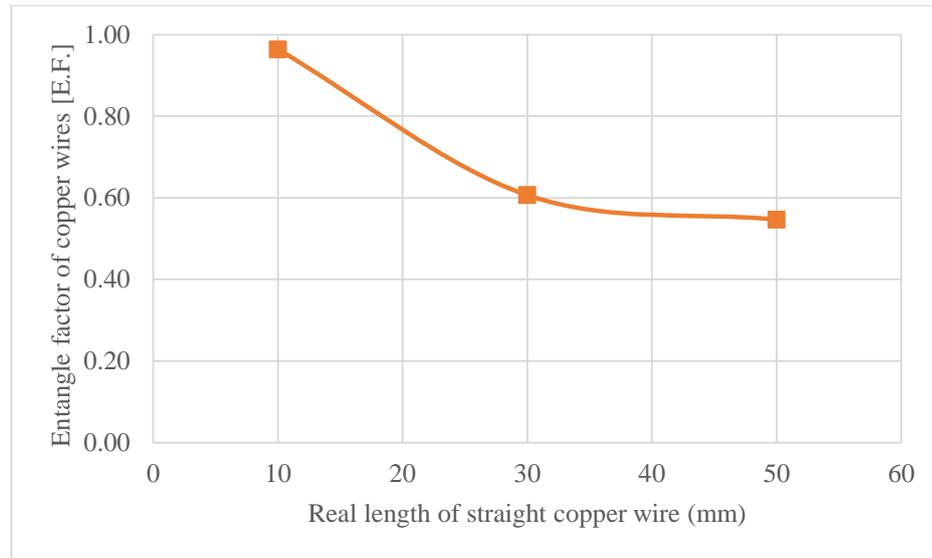


Figure 3. 14 Relationship between Entangle factor of copper wires [E.F.] and Real length of straight copper wire (mm)



## CHAPTER IV

### EFFECTS OF COPPER WIRES ON JIG SEPARATION

#### 4.1 Jig separation of real sample

##### 4.1.1 Experimental

The +1.0-16 mm fractions of the real sample described in chapter 3 was used for the jig tests. Before the jig test, sink-float separation of this sample by water was carried out to remove floating components. The jig separation was carried out under the conditions of 30 mm displacement, 30 cycles/minute frequency of water pulsation, with addition of 15 ppm of Aerosol OT or AOT (Dioctyl sodium sulfosuccinate:  $C_{20}H_{37}NaO_7S$ ) as a wetting agent, 3 minutes separation time, and alumina balls as ragging. After the jig separation, products were divided into six layers from the top as shown in figure 4.1 and hand sorting was carried out to determine the components of the jig products, the products were separated into three groups; copper wires, other metals, and plastics. The 'other metal fractions' were analyzed by X-Ray fluorescence (XRF) and the 'plastic fractions' were ground at low temperature chilled with liquid nitrogen and digested by a microwave using aqua regia (a mixture of nitric acid and hydrochloric acid in a volume ratio of 1:3) and then analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES).

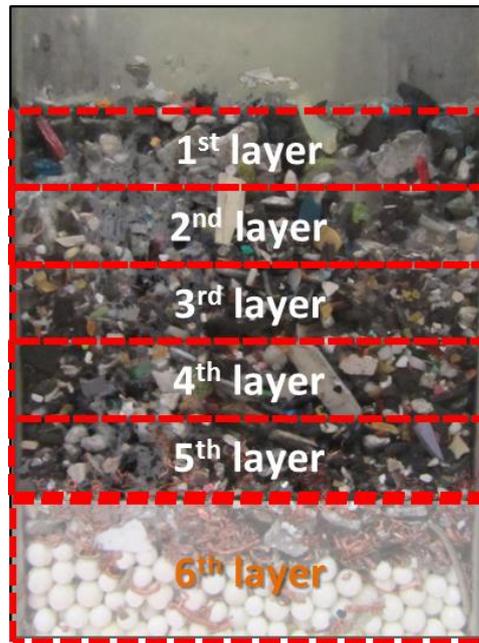


Figure 4. 1 Photo of recovered product after jig separation

#### 4.1.2 Results and discussion

The pictures of real sample in separation chamber before and after jig separation are shown in figure 4.2 and the components of each layer after jig separation are shown in figure 4.3. From the results, the copper content in the 1<sup>st</sup> to 4<sup>th</sup> layer was lower than 4%, while most of which concentrated in the 5<sup>th</sup> and 6<sup>th</sup> layers (37% in the 5<sup>th</sup> layer and 57% in the 6<sup>th</sup> layer). Jig products of the 5<sup>th</sup> and 6<sup>th</sup> layers are shown in figure 4.4. From the 5<sup>th</sup> layer product, it was visually observed that the copper wires entangled and the entangled copper wires had trapped plastic and prevented their motion in the chamber. Hence, effect of the copper wires on the jig separation was investigated using model samples.

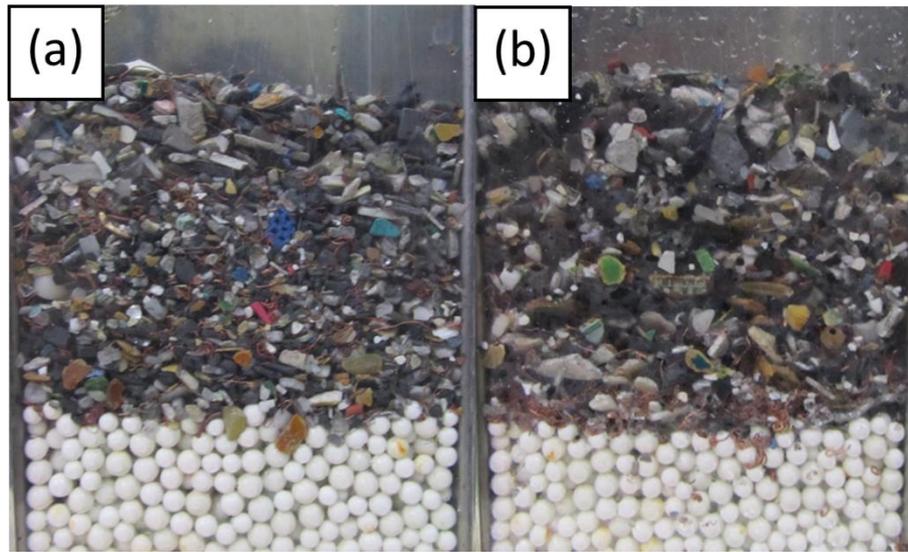


Figure 4. 2 Pictures of real sample in separation chamber;  
(a) before and (b) after jig separation

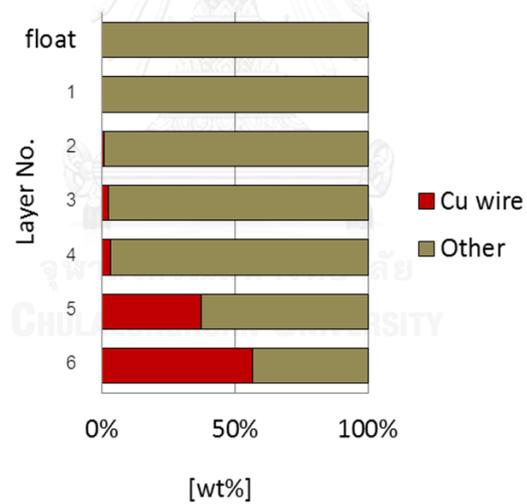


Figure 4. 3 Components of each layer after jig separation of real sample

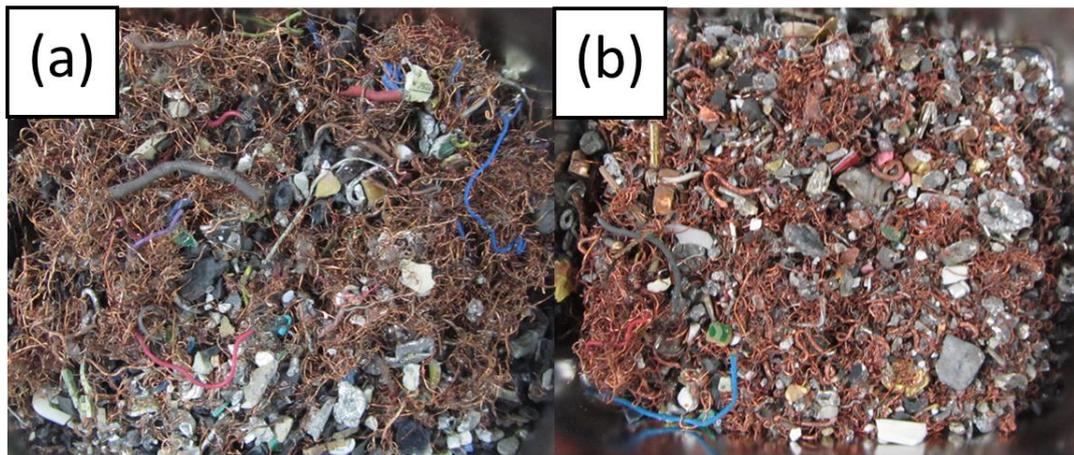


Figure 4.4 Jig products of real sample; (a) the 5<sup>th</sup> layer and (b) the 6<sup>th</sup> layer

## 4.2 Jig separation of model samples

### 4.2.1 Experimental

The mixture of ABS, PVC 500 g (ratio 1:1), and copper wires 0.4 mm diameter were used as model samples. The copper wires with specific lengths (10, 30, and 50 mm) were added at different mass ratios (0, 5, 10, and 15% of the total sample) and fed to the jig separation. The jig tests were carried out under the conditions of 30 mm displacement, 30 cycles/minute frequency of water pulsation, with 15 ppm of AOT addition, 1, 2, or 3 minutes separation time, and alumina balls as ragging. After the jig separation, products were divided into six layers from the top and hand sorting was used to determine the purities of the jig products.

### 4.2.2 Results and discussion

Example pictures of model samples in separation chamber before and after jig separation are shown in figure 4.5.

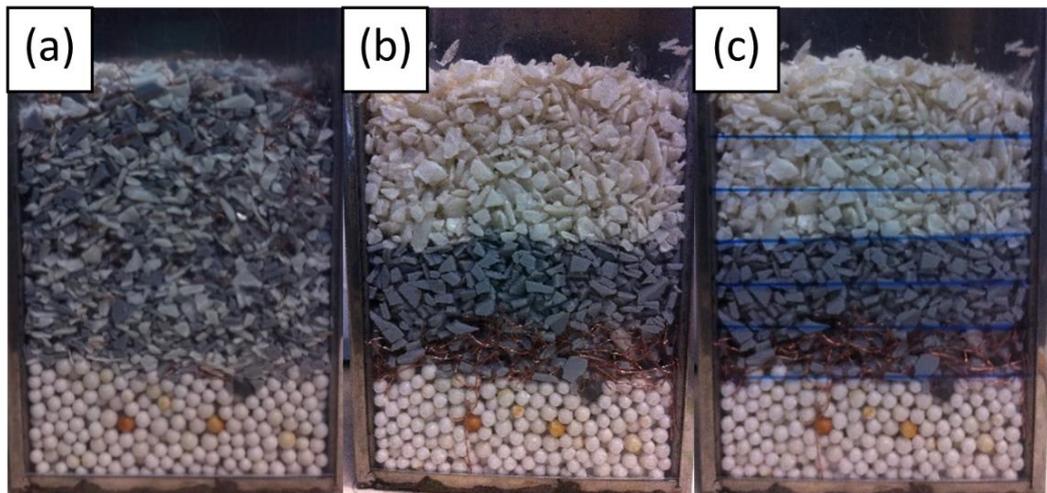


Figure 4. 5 Example pictures of model samples in separation chamber; (a) before jig separation, (b) after jig separation, and (c) upon divided into 6 layers

Figure 4.6 and 4.7 show pictures and components of the jig product of model sample (without copper wire) in each layer after variation separation time.

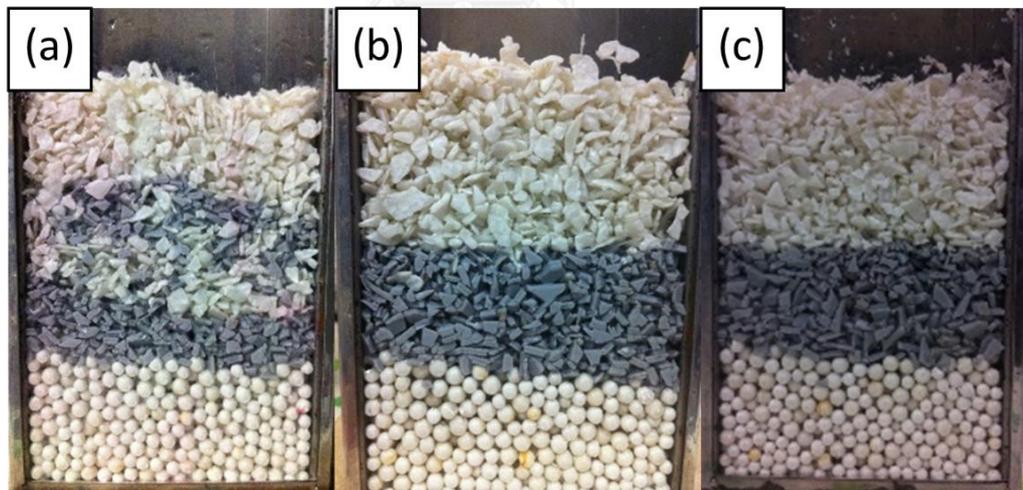


Figure 4. 6 Picture of jig product of model sample (without copper wire) at separation time variation; (a) 1 minute (b) 2 minutes (c) 3 minutes

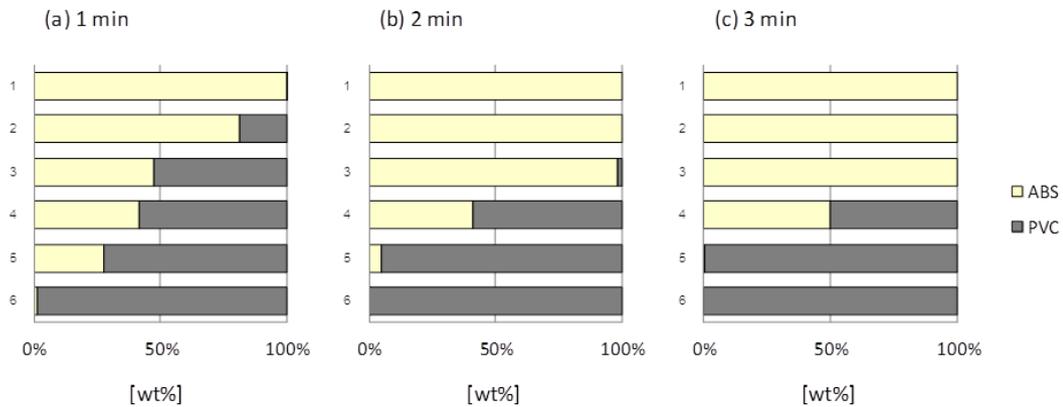


Figure 4. 7 Components of each layer after jig separation of model samples (without copper wire) at separation time variation;  
(a) 1 minute (b) 2 minutes (c) 3 minutes

The results showed that efficiency of the plastic separation increased with separation time if no copper wires. Due to ABS is lighter than PVC, the ABS was clearly concentrated in the upper layers after 3 minutes separation.

The picture of jig products and components of each layer after jig separation of model samples with copper wires (copper content 10 wt%, 30 mm copper wires length, treatment time: 1, 2, or 3 minutes) are shown in figure 4.8 and 4.9. Purity of ABS in the 1<sup>st</sup> layer and PVC in the 4<sup>th</sup> layer was close to 100% at 3 minutes of jig separation. While the 6<sup>th</sup> layer having 41% copper wires, the copper recovery and separation efficiency did not increase after 3 minutes separation. Entangled copper wires were observed and these may trap plastic particles and prevent their motion in the chamber. The results showed that efficiency of the plastic separation increased with separation time but decreased when the plastics were mixed with copper wires.

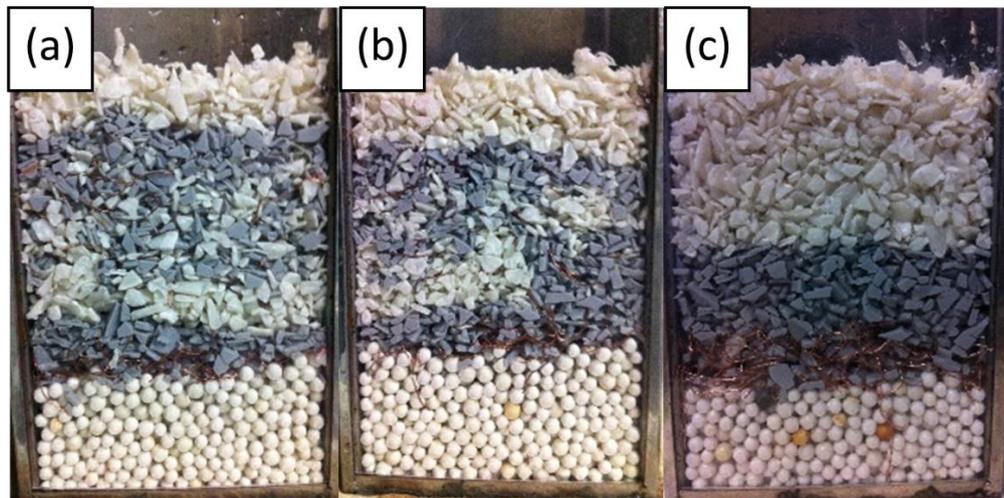


Figure 4. 8 Pictures of jig products of model samples with copper wires (10 % copper wires, 30 mm long copper wires) at separation time variation; (a) 1 minute (b) 2 minutes (c) 3 minutes

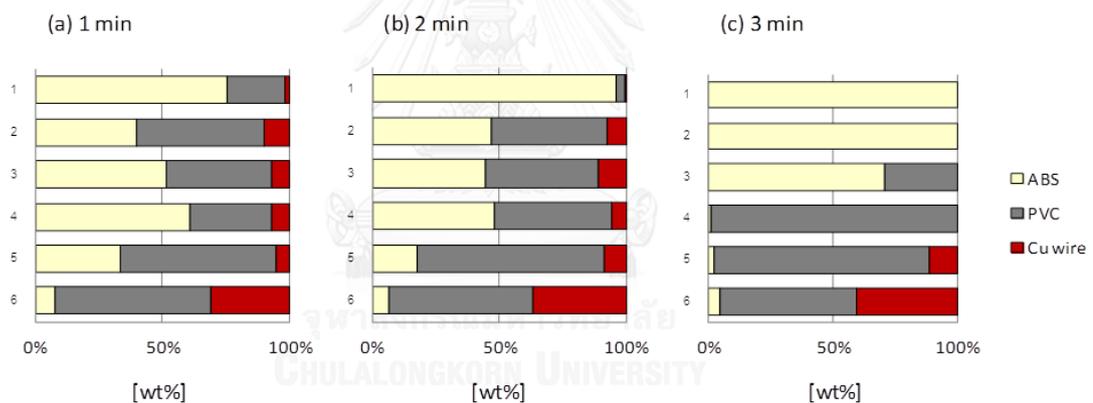


Figure 4. 9 Components of each layer after jig separation of model samples with copper wires (10 wt% copper wires, 30 mm long copper wires) at separation time variation; (a) 1 minute (b) 2 minutes (c) 3 minutes

For the effect of amount of copper wires in jig separation investigation, the picture of jig products and components of each layer after jig separation of model samples vary mass ratio (30 mm long copper wires, separation time 3 minutes) are shown in figure 4.10 and 4.11.

Table 4.1 shows the copper recovery in the bottom layer (6<sup>th</sup> layer) at different mass ratios. The results showed that recovery of the copper wires in the sixth layer and the efficiency of plastic separation decrease with increasing copper content in the feed (the recovery of copper wires in the sixth layer with 5, 10, and 15% weight of copper wires was 98, 78, and 32% respectively). It is suggested that effect of the entangled copper wires increases with copper content in the feed as the contact probability of copper wires become higher. The higher weight ratio of copper wires the increasing probability for entanglement of copper wires. From these reasons, ABS was difficult to go upward and copper wires were difficult to go downward. Hence, decrease the recovery of copper wires and the separation efficiency. However, recovery of copper at 5 wt% in the feed was 98% suggesting low effect of a small amount of copper wires if less than 5 wt%. Figure 4.12 shows the relationship between copper content in feed and copper recovery in 6<sup>th</sup> layer.

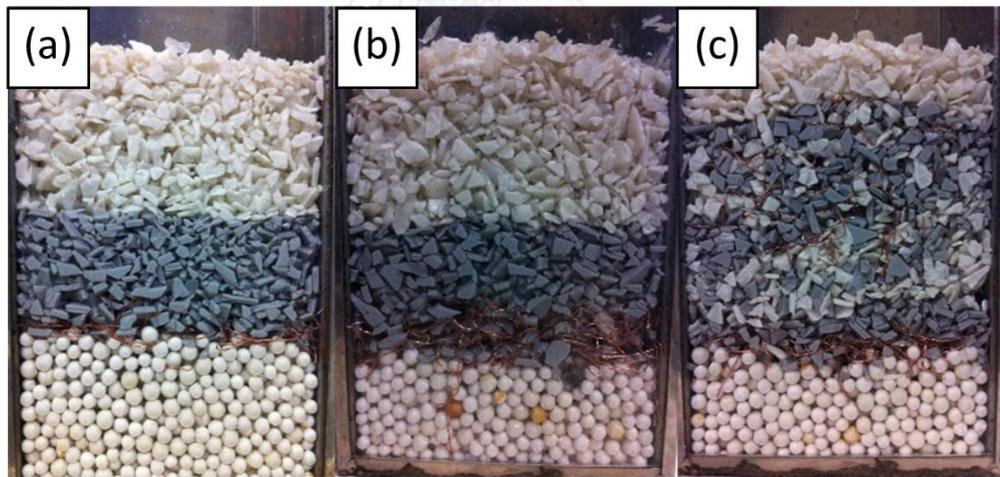


Figure 4. 10 Pictures of jig products of model samples with copper wires (30 mm long copper wires, separation time 3 minutes) at mass ratio variation;  
(a) 5 % (b) 10 % (c) 15 %

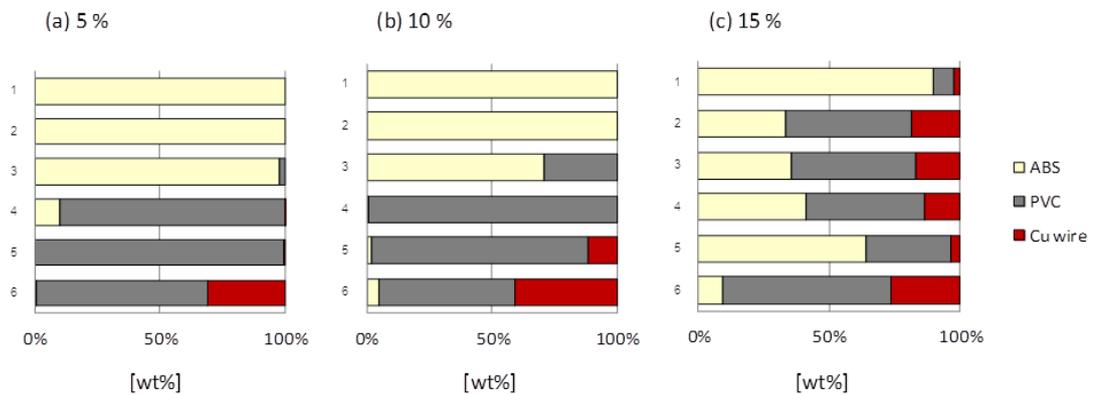


Figure 4. 11 Components of each layer after jig separation of model samples with copper wires (30 mm long copper wires, separation time 3 minutes) at mass ratio variation; (a) 5 % (b) 10 % (c) 15 %

Table 4. 1 Recovery and purity of copper in the 6<sup>th</sup> layer at mass ratios variation; 5 %, 10 %, and 15 % (30 mm long copper wires, separation time 3 minutes)

Copper content in feed	Copper recovery in 6 <sup>th</sup> layer	Copper purity in 6 <sup>th</sup> layer
5%	98%	31%
10%	78%	41%
15%	32%	26%

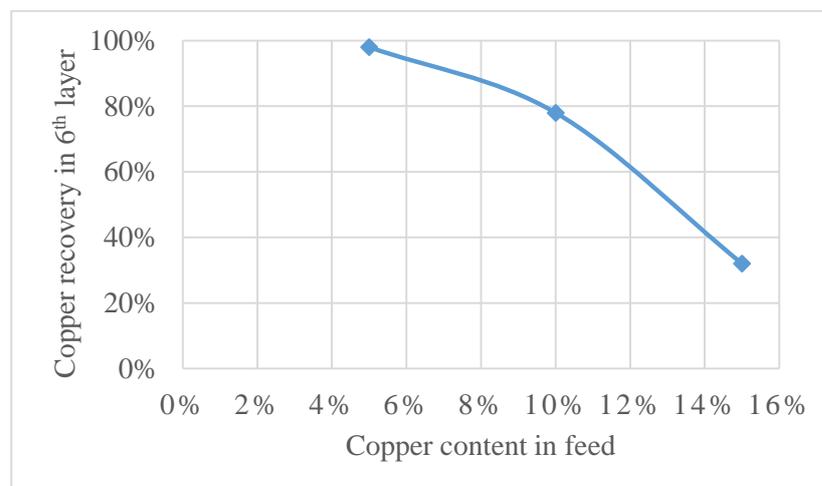


Figure 4. 12 Relationship between copper content in feed and copper recovery in the 6<sup>th</sup> layer

For the effect of length of copper wires in jig separation investigation, the picture and components of jig products at each layer of model samples with 10% copper wires, 3 min separation time, but length variation: (a) 10 mm, (b) 30 mm, (c) 50 mm are shown in figure 4.13 and 4.14.

Table 4.1 shows the copper recovery in the bottom layer (6<sup>th</sup> layer) at varying lengths. The results showed that the recovery of copper wires in the sixth layer and the efficiency of plastic separation decreased when the length of copper wires increase (the recoveries of 100, 78, and 52% respectively for the 10, 30, and 50 mm length of copper wires) since the longer copper wires the easier bending and getting entangled during the jig motion conform to the Entanglement Factor (E.F.) as described in chapter 3. Recovery of copper at 10 wt% of 10 mm long wires was 100% suggesting the low effect of copper wires shorter 50 mm long. These bring to the difficulty for moving upward of the ABS and moving downward of the copper wires. Hence, the recovery of copper wires and the separation efficiency were decreased. Figure 4.15 shows the relationship between entanglement factor (E.F.) and copper recovery in 6<sup>th</sup> layer.

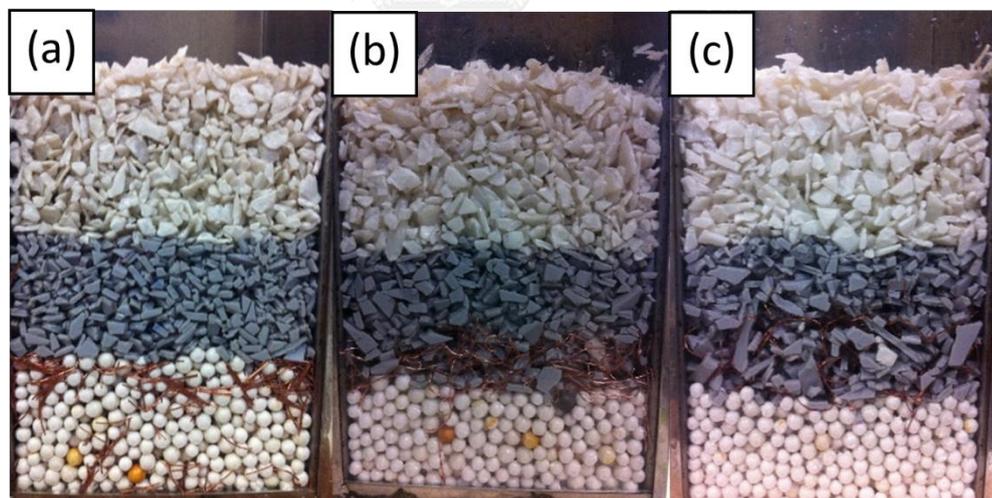


Figure 4. 13 Pictures of jig products of model samples with copper wires (10% copper wires, separation time 3 minutes) at lengths of copper wires variation; (a) 10 mm (b) 30 mm (c) 50 mm

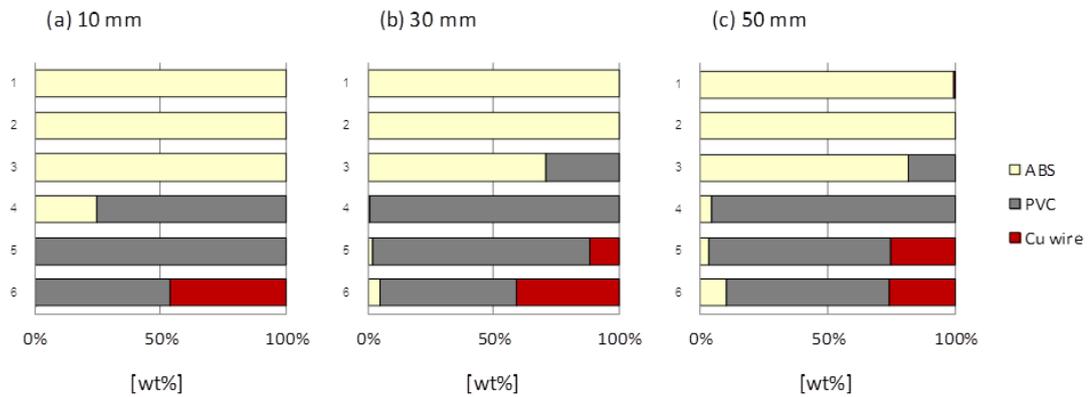


Figure 4. 14 Components of each layer after jig separation of model samples with copper wires (10 % copper wires, 3 minutes ) at lengths of copper wires variation; (a)10 mm (b) 30 mm (c) 50 mm

Table 4. 2 Recovery and purity of copper in the 6<sup>th</sup> layer at lengths of copper wires variation; 10 mm, 30 mm, and 50 mm (10 % copper content, separation time 3 minutes)

Length of copper wires	Copper recovery	Copper purity
	in 6 <sup>th</sup> layer	in 6 <sup>th</sup> layer
10 mm	100%	48%
30 mm	78%	41%
50 mm	52%	26%

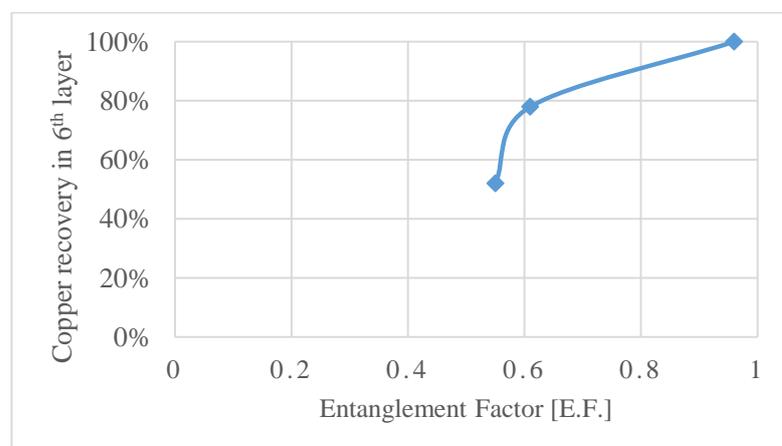


Figure 4. 15 Relationship between entanglement factor [E.F.] and copper recovery in the 6<sup>th</sup> layer

These results indicate that the shorter wires and the lower copper content in the feed exhibit the lower influence. Hence, removal of long wires and control of the copper content in the feed are effective to promote the separation of copper and plastics. Figure 4.16 shows the entanglement phenomena during jig separation. The ABS, PVC, and copper wires are represented by white circles, grey circles, and red line in order. First, the mixtures were mixed and put in separation chamber. All particles were moved up in pulsion stroke, and were moved down in suction stroke. Due to the densities difference, copper wires which have the highest density will go down fastest but when many copper wires were contacted they will entangle with other copper wires. They trap the plastic particles and prevent their motion in the chamber. ABS was difficult to go upward and copper wires were difficult to go down in next pulsion stroke and suction stroke.

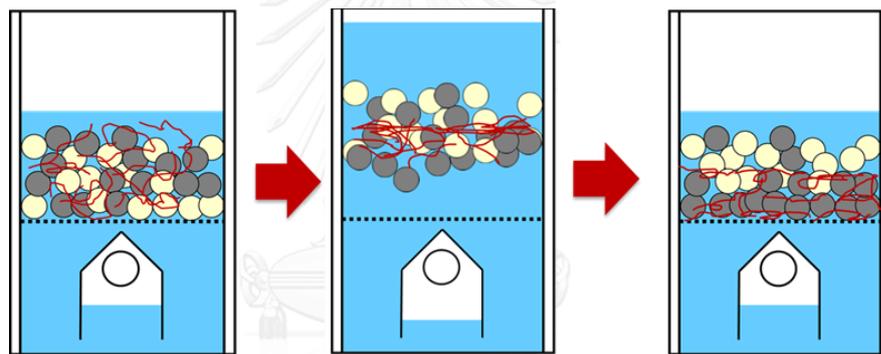


Figure 4. 16 Entanglement phenomena during jig separation  
(white circles: ABS, grey circles: PVC, and red line: copper wires)

There are two ways to increase the recovery of copper wires and the efficiency of plastic separation. The first method is reducing the mass ratio of copper wire by pre-treatment before jig separation. The second method is reducing the length of copper wires by crushing because if the length of copper wires are about 10 mm. or less, copper wires are hardly entangled but there are the problems of this method because when the samples size are reduced, it will be more difficult for separation and crushing costs also increase. Pre-treatment to remove copper wires before the jig separation was carried out next and is described in the next chapter.

## CHAPTER V

### DEVELOPMENT OF PRE-TREATMENT METHOD

#### 5.1 Pre-treatment of model samples

##### 5.1.1 Experimental

To improve the separation performance, two approaches were considered. One was reducing the mass ratio of copper wires by pre-treatment before jig separation. The second was reducing the length of the copper wires by crushing until the length of the copper wires become 10 mm or less, however all particle sizes become smaller by the crushing and this may cause decreases in separation efficiency by the jig separation.

This study tried method one, reducing the mass ratio of copper wires. A mill pot or ball mill without balls was tested to remove copper wires from the model sample by using the entanglement properties of the wires. The model samples with copper wires (the conditions were 10% weight copper wires of the total samples and vary length 10, 30, and 50 mm) were fed to mill pot for pre-treatment. The mill pot was rotated at 78 rpm (80% of the critical speed of rotation) for 5, 10, and 15 minutes. The products were sieved at 16 mm to remove the entangled copper wires as an oversize fraction.

For the speed calculation, the formula is shown in Equation 5.1.

$$N_c = \frac{42.3}{\sqrt{D-d}} \quad (5.1)$$

Where  $N_c$  is the critical speed of the mill (revolutions per minute: rpm),

$D$  is the mill diameter (meter: m),

$d$  is the ball diameter (meter: m),

In this study, a mill pot without balls was used, then the median size of samples diameter ( $d_{50}$ ) 3 mm was used instead of the ball diameter ( $d$ ) and the mill pot inner diameter ( $D$ ) was 190 mm. From these data, the critical speed of the mill was 97.8 rpm and 80% of the critical speed was 78.2 rpm.

This process is similar with the mechanism of the trommel. Trommel, also known as the rotary screen or revolving screen, is the one of the oldest screening devices which is used mainly in the mineral processing. It consists of a perforated cylindrical drum which is normally elevated at an angle at the feed end. Physical size separation is achieved as the feed material spirals down the rotating drum, where the undersized material smaller than the screen apertures passes through the screen while the oversized material exits at the other end of the drum (B.A. Will, 2006).

The mill pot, 16 mm sieve, and the outline of trommel are shown in figure 5.1, 5.2, and 5.3.

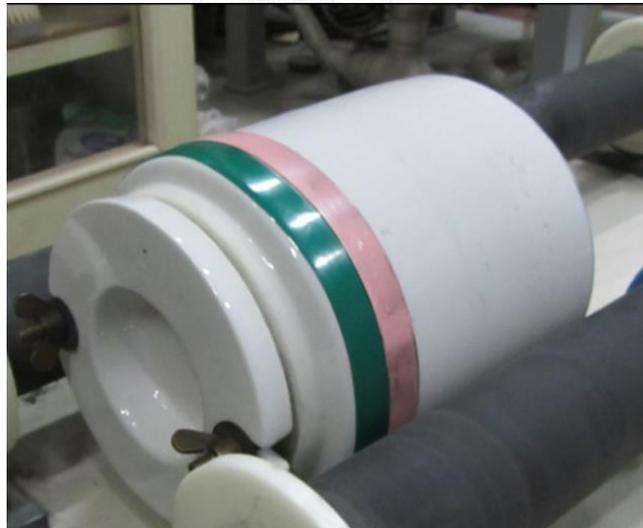


Figure 5. 1 Mill pot; inner diameter 190 mm



Figure 5. 2 Sieve; aperture size 16 mm.

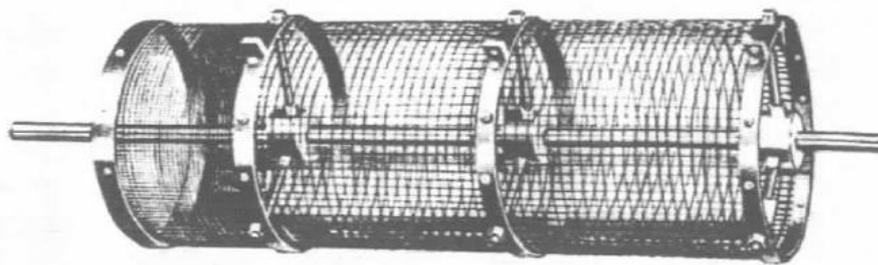


Figure 5. 3 Outline of trommel

### 5.1.2 Results and discussion

For the result of the pre-treatment by mill pot of model samples, the 10 and 30 mm copper wires were not entangled because of short length and less entanglement. For 50 mm copper wires, copper wires entangled and changed to be the ball shape when samples are stirred. The model sample before and after pre-treatment by mill pot are shown in Figure 5.4. The copper wire balls were removed by a 16 mm sieve as oversize product and the under size fraction was treated by jig separation. The pre-treatment product of model sample (+16 mm) is shown in figure 5.5. Table 5.1 shows recovery and purity of Copper in pre-treatment product of model sample (+16 mm) at varying rotating time 5, 10, and 15 minutes (10 % copper content, length of copper wires 50 mm).

The recoveries of copper wires after pre-treatment for 5, 10, and 15 minutes were 48, 79, and 75% in order (Purities of copper wires that were recovered are more than 95%). From these results, 10 minutes is enough for pre-treatment. Then, 10 minutes condition was used for pre-treatment of real sample.

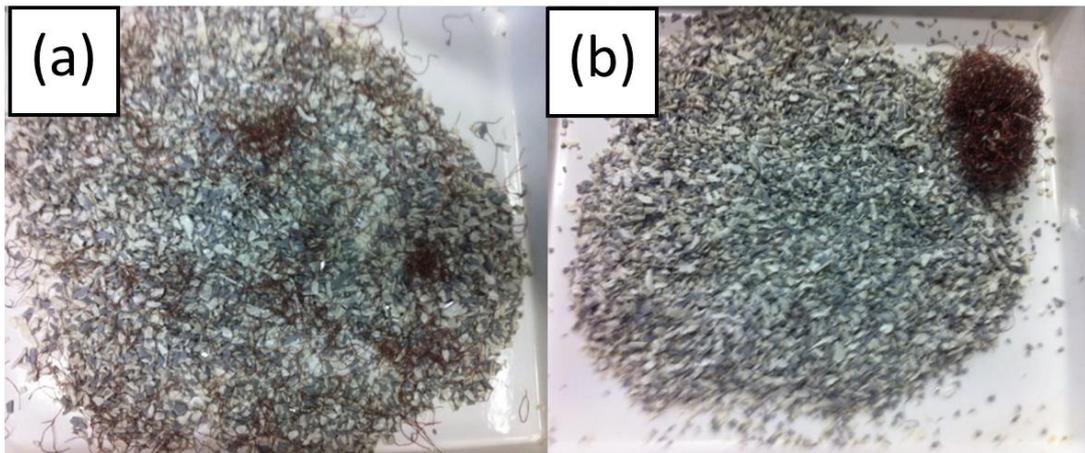


Figure 5. 4 Model sample; (a) before and (b) after pre-treatment by mill pot

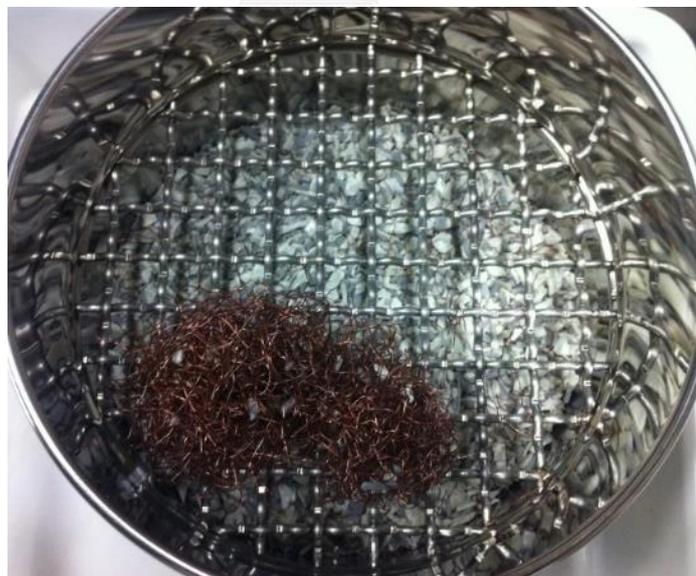


Figure 5. 5 Pre-treatment product of model sample (+16 mm)

Table 5. 1 Recovery and purity of copper in pre-treatment product of model sample (+16 mm) at rotating time variation; 5, 10, and 15 minutes (10 % copper content, 50 mm long of copper wires)

<b>Rotating time</b>	<b>Copper recovery</b>	<b>Copper purity</b>
5 min	48%	98%
10 min	79%	96%
15 min	75%	96%

## **5.2 Pre-treatment of real sample**

### **5.2.1 Experimental**

From the results and discussion of pre-treatment of model sample, 10 minutes is enough for pre-treatment. Then, a mill pot without balls was used to remove copper wires from the real sample by using the entanglement properties of the wires. The real sample was fed to a mill pot and the mill pot was rotated at 78 rpm (80% of the critical speed of rotation) for 10 minutes. The products were sieved at 16 mm to remove the entangled copper wires as an oversize fraction. The under size fraction was fed to the jig.

### **5.2.2 Results and discussion**

The real sample; before and after pre-treatment by mill pot are shown in figure 5.6. Long copper wires (about 20 mm or more) were recovered in the pre-treatment process by the mill pot and the purity of copper in the oversize product was 93% and copper recovery was 19%. The pre-treatment product of real sample (+16 mm) is shown in figure 5.7.

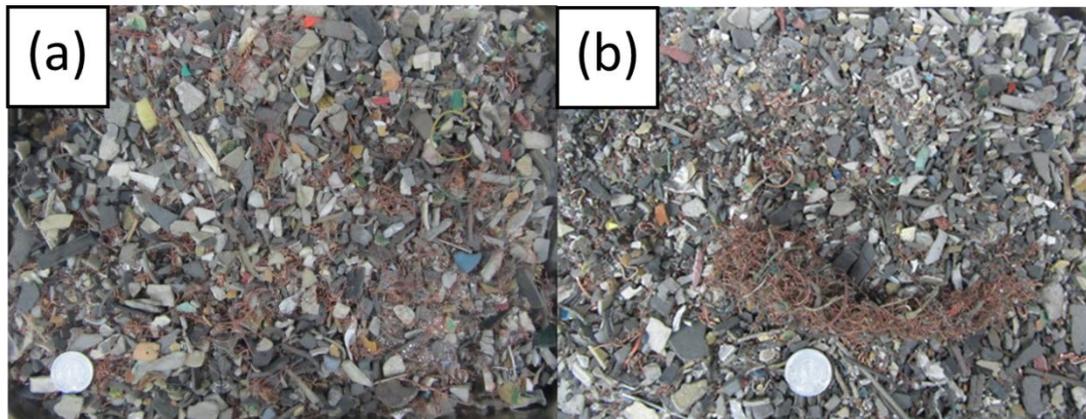


Figure 5. 6 Real sample; (a) before and (b) after pre-treatment by mill pot



Figure 5. 7 Pre-treatment product of real sample (+16 mm)

For jig separation of real sample after pre-treatment, the picture of real sample (with pre-treatment) in separation chamber (before and after jig separation) and the comparison of components of each layer after jig separation of real sample between with and without pre-treatment are shown in figure 5.8 and 5.9.

For jig separation of real sample without pre-treatment, the purity of copper in sixth layer was 57% and the copper recovery was 66%. There are copper in fifth and sixth layer about half and little bit in second, third, and fourth layer. For jig separation of real sample with pre-treatment, the results showed that the purity of copper in sixth layer was 45% and the copper recovery was 87%. Some of long copper wires (about 20 mm or more) were recovered in pre-treatment process by mill pot (Purity of copper

wires was 93% and copper recovery was 19%). For comparison with jig separation of model samples, there are only few of short copper wires after pre-treatment and the recovery in sixth layer increased same with model samples. The results showed that the recovery of copper in the sixth layer increased from 66% to 87% by using this pre-treatment and total copper recovery from pre-treatment and jig separation was 89%. It was confirmed that this pre-treatment method can improve the separation efficiency of copper and plastics. The jig products of fifth and sixth layer are shown in figure 5.10.

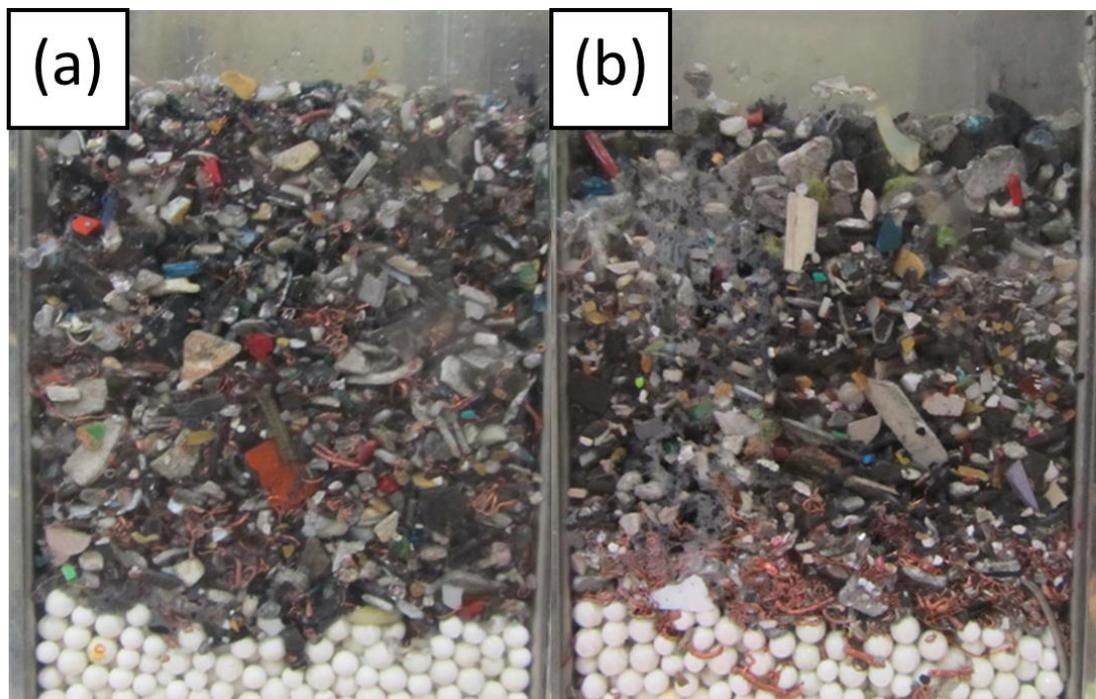


Figure 5. 8 Pictures of real sample (with pre-treatment) in separation chamber;  
(a) before and (b) after jig separation

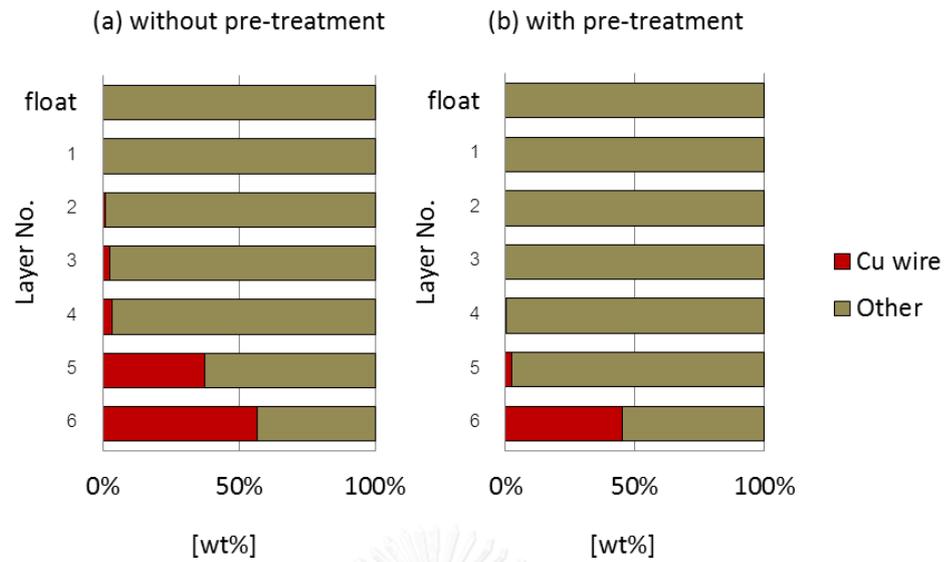


Figure 5. 9 Components of each layer after jig separation of real sample;  
 (a) without and (b) with pre-treatment

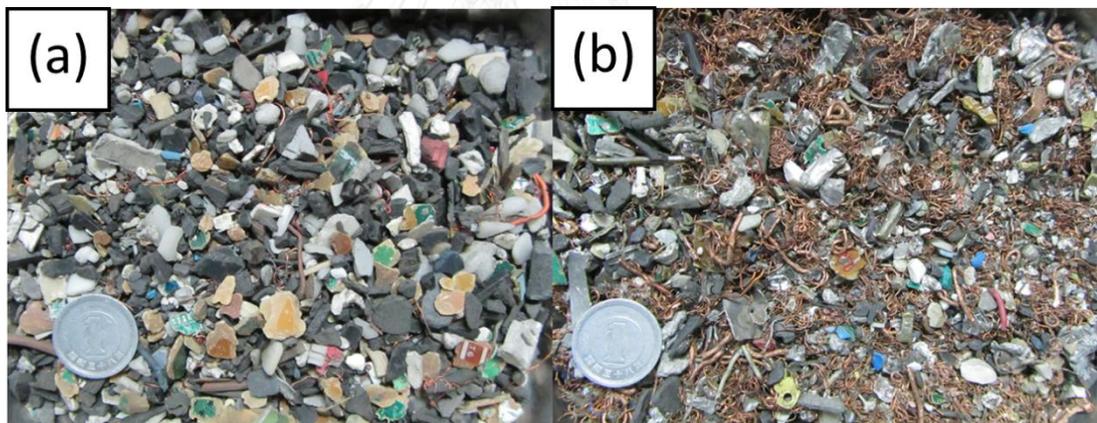


Figure 5. 10 Jig products of real sample (with pre-treatment);  
 (a) 5<sup>th</sup> layer and (b) 6<sup>th</sup> layer

## CHAPTER VI

### CONCLUSION

#### 6.1 Conclusion

After removing large pieces of metals from crushed end-of-life small appliances by magnetic and eddy current, separation of plastics and metals from the +1.0-16 mm fraction was investigated by RETAC jig separation. Entanglement of copper wires in the separation chamber was observed to prevent the progress of separation. The higher mass ratios as well as the longer copper wires tend to cause the more entanglement. Pre-treatment by a mill pot can recover the entangled copper wires and increase the recovery of copper in jig separation. Copper with 93% purity was recovered from the pre-treatment process and the copper with 45% purity was recovered from sixth layer of jig separation. Hence, the total recovery of copper increased from 66% to 89%.

The copper wires balls with 93% purity recovered from the pre-treatment process can be sold to smelting plant or recycling plant as a high grade copper and the copper with 45% purity recovered from sixth layer of jig separation can be sold to smelting plant as a low grade copper.

The recovered plastic can be sent to recycling plant or incinerator (exclude PVC) to generate electricity. In addition, special trommel was suggested to use in the pre-treatment process.

From the results above, pre-treatment using mill pot and 16 mm sieve can be used to recover the entangled copper wires and increase the recovery of copper in jig separation. In commercial scale, either mill pot or ball mill is not commonly used for the pre-treatment. Trommel or rotating screen is suggested for commercial scale of the pre-treatment process because the movement of trommel is similar with the mill pot while sizing the materials and feeding to the next process at the same time.

Special trommel should be used for the pre-treatment before jig separation. Outline of a special trommel is shown in figure 6.1. The speed should be at 80% of the critical rotational speed. First part of special trommel should have no screen because the copper wires have to be in this part for changing to copper wires balls (residence time in this part should be about 10 minutes). Then, the particle should be fed to the next parts having a screen to separate copper wires balls from the other materials. For the last section, the screen apertures should be about 16 mm to separate them. The copper wires ball will go to over screen fraction and other materials (mixture of plastics and short copper wires) will pass through the screen as the under screen fractions and were fed to the jig separation.

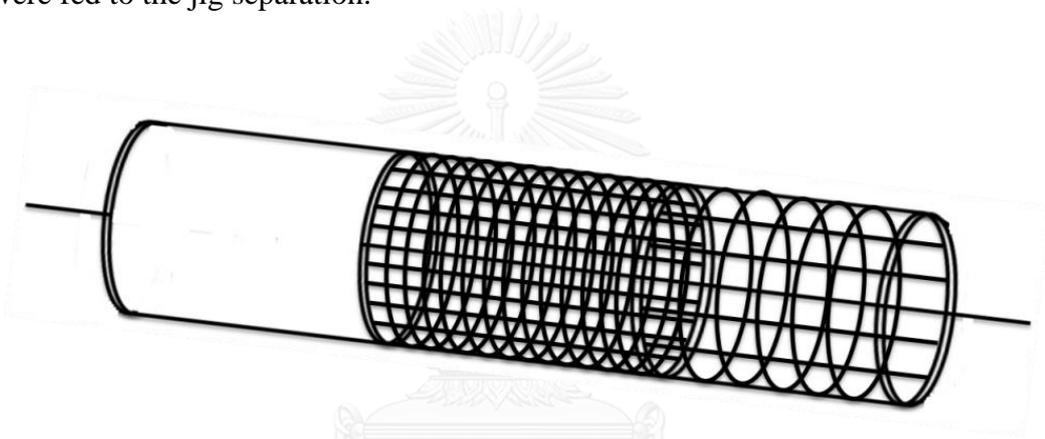


Figure 6. 1 Outline of special trommel

From the results of this study, the suggestive process for small home appliances recycling was created. The copy machine and ATM recycling process can be applied to recover valuable metals and plastics from the end-of-life small home appliances. After dust removal from the crushed end-of-life small home appliances, coarse magnetic matters were removed by magnetic separation and the coarse metals were removed by eddy current separation. There are three groups of the plastics and metals mixture products; coarse, medium, and fine. The coarse and medium groups can be fed to the RETAC jig to recover copper directly; while the fine group should be fed to special trommel for the pre-treatment to recover copper wires before RETAC jig separation. The recycling flow chart of small home appliances (suggestive process) is shown in figure 6.2.

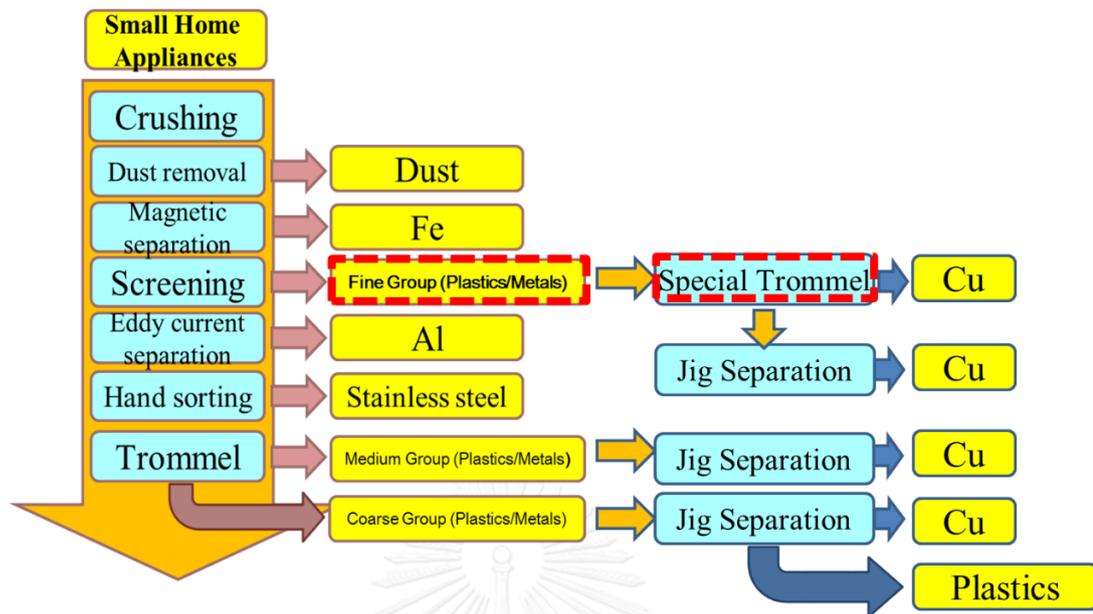


Figure 6. 2 Recycling flowchart of small home appliances (suggestive process)

## 6.2 Recommendation

- 1) Laboratory scale trommel should be investigated before applying in commercial scale to investigate suitable conditions such as feed rate, rotating speed, incline angle, residence time, screen sizes and lengths of each part of trommel, etc.
- 2) Fourier transform infrared spectroscopy (FTIR) should be used in the future work to analyze the kind of plastics found in the samples.
- 3) In this research, the RETAC jig was used to efficiently recover valuable metals and plastics from the end-of-life small home appliances with large and moderately sized particles. However, separation efficiency rapidly drops when applied to fine particles ( $< 2$  mm) because of some unclear phenomena that happen in the separation chamber. From this reason, recovery of valuable metals and plastics from fine particle fraction of the crushed end-of-life small home appliances and other e-wastes (e.g., laptops, mobile phones, etc.) should be considered in the future work. Effect of agglomeration in the recovery of valuable components from fine particles e-wastes by jig separation and comparison with other gravity separation ex. Knelson concentrator (centrifugal gravity concentrator) and hydrocyclone should be

investigated. For fine particles, separation using jiggling is very difficult. As such, agglomeration, the process of connecting fine particles to create larger agglomerates using organic “bridging” molecules [e.g., Benzohydroxamic Acid (BHA)], may improve the separation efficiency because of the formation of much larger particles. In addition, metals and plastics have distinct surface properties (i.e., hydrophobic or hydrophilic) so agglomeration and separation could be adjusted to be more selective. Several parameters such as flocculants type, flocculants dosage, mixing speeds and mixing time could affect the degree and efficiency of agglomeration, so all of these factors should be evaluated. Other gravity separation techniques like Knelson concentrator (a type of centrifugal gravity concentrator) hydrocyclone should be used to process the fine fraction of e-wastes. The effect of agglomeration should also be evaluated in these experiments under various conditions. Finally, the results from this type of gravity separation technique should be compared to that of the RETAC jig not only in terms of separation efficiency but also in terms of economic viability.

4) Development of a mathematical model, calibration and optimization should be considered in the future work to simulate the movement of particles in the RETAC jig, which could predict the optimum conditions needed for jig separation at various particle sizes and compositions. The mathematical model should be based on the different forces acting on particles during dilation and settling. The model will be calibrated first using pure plastic resins and metals of known particle sizes. After calibration, it would be used to predict the movement of particles during jig separation and the model results would be compared with the actual movement of particles captured by the high speed video camera.

## REFERENCES

- Choi, W., Park, E. and Kang, S. (2015). Status and Prospects of Plastics Recycling of Used Small Household Appliances. The 13th International Symposium on Earth Asian Resources Recycling Technology, Pattaya, Thailand.
- Exexutive, H. a. S. (2015). "Waste Electrical and Electronic Equipment recycling (WEEE)."
- Hori, K., Tsunekawa, M., Hiroyoshi, N. and Ito, M. (2009). "Optimum water pulsation of jig separation for crushed plastic particles." International Journal of Mineral Processing **92**: 103-108.
- Information, C. (2015). "Waste from electrical and electronic equipment."
- Ito, M., Akatsuka, M., Kuwayama, Y., Hori, K., Hiroyoshi, N. and Tsunekawa, M. (2008). "Immersion Behavior of Automobile Shredded Residue in Surfactant Solutions and Detachment of Particulate Matter." The Mining and Materials Processing Institute of Japan: 1-6.
- Ito, M., Khanchang, S., Saito, A., Murase, N., Hamaya, N. and Hiroyoshi, N. (2013). Recycling Treatment of Cell Phones Including PCBs by Advanced Jigging. ASEAN++2013 Moveing Forward, The 11th International Conference on Mining, Materials and Petroleum Engineering, The 7th International Conference on Earth Resources Technolgy, Chiang Mai, Thailand.
- Ito, M., Kitajima, N., Saito, A., Khanchang, S., Tabelin, C. and Hiroyoshi, N. (2015). Recovery of Valuable Components from Crushed Mobile Phones using Jig Separation. The 13th International Symposium on Earth Asian Resources Recycling Technology, Pattaya, Thailand.
- Ito, M., Saito, A., Murase, N., Hamaya, N., Takeuchi, M. and Hiroyoshi, N. (2013). Separation of Polyethylene and Cross-Linked Polyethylene Using a Reverse Hybrid Jig. ASEAN++2013 Moveing Forward, The 11th International Conference on Mining, Materials and Petroleum Engineering, The 7th International Conference on Earth Resources Technolgy, Chiang Mai, Thailand.
- Ito, M., Tsunekawa, M., Ishida, E., Kawai, K. Takahashi, T., Abe, N. and Hiroyoshi, N. (2010). "Reverse jig separation of shredded floting plastics - separation of polypropylene and high density polyethylene." International Journal of Mineral Processing **97**: 96-99.
- Khanchang, S. (2014). Recycling of mobile phones including Printed Circuit Boards (PCBs) by advanced Jig Separation. Division of Sustainable Resources Engineering, Hokkaido University.

LTD, C. M. (2015). "Category 3 Electronic WEEE Wastes And Other Waste Categories."

Tsunekawa, M., Naoi, B., Ogawa, S., Hori, K., Hiroyoshi, N., Ito, M. and Hirajima, T. (2005). "Jig separation of plastics from scrapped copy machines." International Journal of Mineral Processing **76**: 67-74.

Wills, B. A. (2006). Wills' Mineral Processing Technology, Oxford, UK: Pergamon Press.



## APPENDIX

### Analytical machines in this study

#### X-Ray Fluorescence (XRF) Spectrometry

##### *Basic theory*

XRF Spectrometry is used to identify the elements in substances and quantify the concentration of those elements to determine the chemical composition of materials. The elements are identified by their characteristic X-ray emission wavelength ( $\lambda$ ) or energy (E). The concentration of each element is quantified by the intensity (I) measurement of its characteristic emission.

All atoms have a fixed number of electrons, arranged in orbitals around the nucleus. Energy Dispersive (ED) XRF and Wavelength Dispersive (WD) XRF Spectrometry commonly use activity in the first three electron orbitals, the K, L, and M lines, where K is closest to the nucleus.

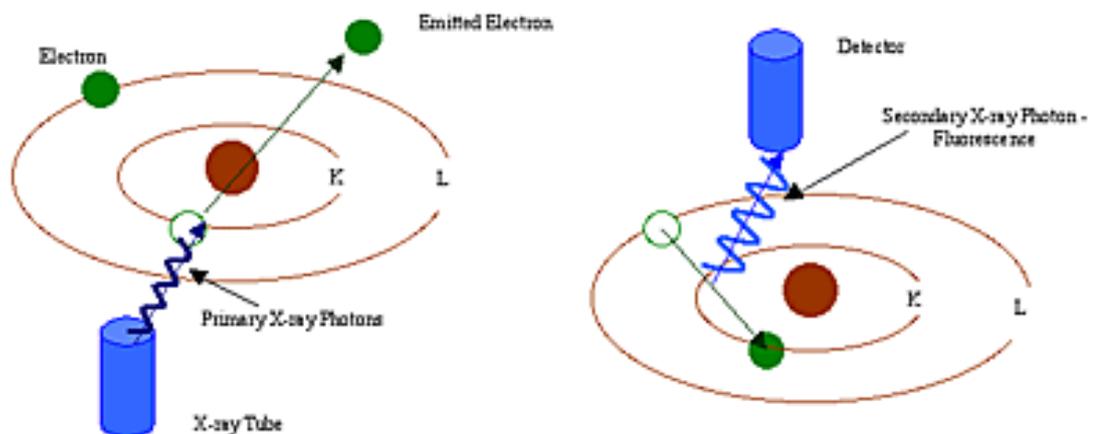


Figure A-1 Schematic of X-ray fluorescence spectrometry

In XRF Spectrometry, high-energy primary X-ray photons are emitted from an X-ray tube and strike to the sample. The primary photons from the X-ray tube have enough energy to knock electrons out of the innermost, K or L, orbitals. When this phenomena occurs, the atoms will change to be unstable ions. An electron from an outer orbital, L or M, will move to the newly vacant space at the inner orbital to regain stability.

The secondary X-ray photon is emitted when the electron from the outer orbital moves into the inner orbital space. This phenomenon is called fluorescence. The secondary X-ray produced is characteristic of a specific element. The emitted fluorescent X-ray photon energy (E) is determined by the energy difference between the initial and final orbitals of the individual transitions.

This is described by the Equation A-1

$$E = hc\lambda^{-1} \quad (\text{A-1})$$

Where h is Planck's constant, c is the velocity of light, and  $\lambda$  is the characteristic wavelength of the photon. Energies are inversely equivalent to the wavelengths, they are characteristic for each element.

### ***Elemental analysis***

XRF Spectrometry easily and quickly identifies and quantifies elements over a wide dynamic concentration range, from ppm to 100% by weight. XRF Spectrometry does not destroy the sample, requires just little sample preparation, and take only short time. These factors lead to a significant reduction in the per sample analytical cost when compared to the other elemental analysis techniques.

All elemental analysis techniques experience interferences, both chemical and physical in nature, and corrected for achieving sufficiently analytical results. In XRF Spectrometry, the primary interference is from other specific elements in a substance that can influence the analysis of the elements of interest.

However, these interferences are well-known. Documented and instrumentation advancements and mathematical corrections in the system's software easily and quickly correct for them. In certain cases, the geometry of the sample can affect XRF analysis, but this is easily compensated for by selecting the optimum sampling area, grinding or polishing the sample, or by pressing a pellet or making glass beads.

Quantitative elemental analysis for XRF Spectrometry is typically performed using Empirical Methods (calibration curves using standards similar in property to the unknown) or Fundamental Parameters (FP). FP is often preferred because it allows elemental analysis to be performed without standards or calibration curves. This enables the analyst to use the system immediately, without having to spend more time setting up individual calibration curves for the various elements and materials of interest. The capabilities of modern computers allow the use of this non-standard mathematical analysis, FP, accompanied by stored libraries of known materials, to determine not only the elemental composition of an unknown material quickly and easily but even to identify the unknown material itself.



Figure A-2 X-ray fluorescence (XRF) Spectroscopy (Rigaku Corporation, Japan, Rigaku EDXL300) at the Laboratory of Mineral Processing and Resources Recycling, Division of Sustainable Resources Engineering, Graduate School of Engineering, Hokkaido University, Japan

### **ICP-AES (Inductively Coupled Plasma-Atomic Emission Spectrometry)**

ICP-AES (Inductively Coupled Plasma-Atomic Emission Spectrometry) is an emission spectrophotometry technique, exploiting the fact that excited electrons emit energy at a given wavelength as they return to ground state. The fundamental characteristic of this process is that each element emits energy at specific wavelengths peculiar to its chemical character. Although each element emits energy at multiple wavelengths, in the ICP-AES technique it is most common to select a single or very few wavelength for a given element. The energy intensity emitted at the chosen wavelength is proportional to the element concentration in the analyzed sample. Thus, by determining which wavelengths are emitted by a sample and by determining their intensities, the analyst can quantify the elemental composition of the given sample on a reference standard.

ICP-AES analysis requires a sample in solution phase. Thus, interstitial waters can be analyzed simply, requiring only dilution in most cases. All ICP-AES systems consist of several components, namely on three main aspects: the sample introduction system, the torch assembly, and the spectrometer. The sample introduction system on the ICP-AES consists of a peristaltic pump, Teflon tubing, a nebulizer, and a spray chamber. The fluid sample is pumped into the nebulizer through the peristaltic pump. The nebulizer generates an aerosol mist and injects humidifier Ar gas into the chamber along with the sample. This mist accumulates in the spray chamber, where the largest mist particles settle out as waste and the finest particles are subsequently swept into the torch assembly. Approximately 1% of the total solution eventually enters the torch as a mist, another part will be pumped away as waste.

Ar gas humidification injected into the nebulizer is important when analyzing samples with high dissolved solids. Humidification takes place in the Ar humidifier, where Ar is bubbled through deionized water prior to its expulsion in the nebulizer.

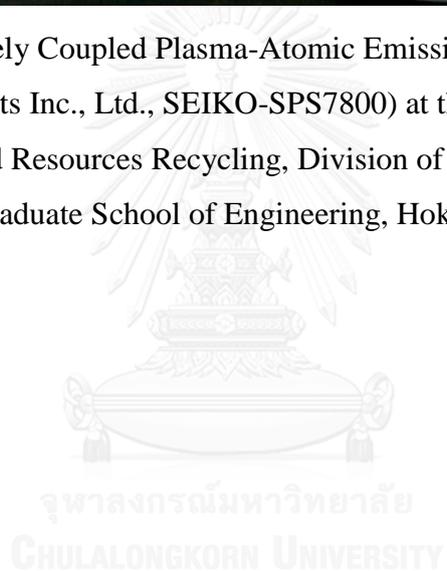
The fine aerosol mist containing Ar gas and sample is injected up to the length of the torch assembly into the plasma. There are several recommended Ar flow rates used in the torch. The radio-frequency-generated and maintained Ar plasma, portions of which are as hot as 10,000 K, excites the electrons. When the electrons return to

ground state at a certain spatial place in the plasma, they emit energy at the specific wavelengths peculiar to the sample's elemental composition.

The plasma is viewed horizontally by an optical channel. Light emitted from the plasma is focused through a lens and passed through an entrance slit into the spectrometer. There are two types of spectrometers used in ICP-AES analysis: sequential (monochromator) and simultaneous (polychromator). The SII Nano Technology Inc. - SPS 7800 Bench-top Optical Emission Spectrophotometer has a sequential spectrometer (double monochromator type). This means that the diffraction grating in the spectrometer is similar to a prism that refracts visible light into its component colors. The photomultiplier tube detector is fixed in space at the far end of the spectrometer. Rotation of the diffraction grating sequentially moves each wavelength into the detector. The computer control ensures that the detector is synchronized with the grating so that the intensity at the detector at any given time is correlated with the wavelength being diffracted by the grating. The operator enters the wavelengths that he or she wishes to detect into the computer, the grating sequentially moves to the specified wavelengths, and the energy intensity at each wavelength is measured to provide a quantitative result that can be compared to a reference standard. Using standard spectroscopic techniques (e.g., background corrections), sequential ICP-AES can provide extremely flexible and rapid analysis of a number of chemical elements. The spectrometer is flushed with N<sub>2</sub> gas to improve the detection limits of elements with emission wavelengths that are severely compromised by interference with air (e.g., P). This N<sub>2</sub> flush, which is maintained in the instrument regardless of whether such elements are being analyzed, also protects the optics from the corrosive aspects of the atmosphere, which are particularly acute at sea.



Figure A-3 Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES) (Seiko Instruments Inc., Ltd., SEIKO-SPS7800) at the Laboratory of Mineral Processing and Resources Recycling, Division of Sustainable Resources Engineering, Graduate School of Engineering, Hokkaido University, Japan



## VITA

The author of this thesis was born on 23th September 1990, in Bangkok, Thailand. After receiving his bachelor's degree of Georesources Engineering from the Department of Mining and Petroleum Engineering, Faculty of Engineering, Chulalongkorn University in 2012, he continuously studied his master's degree of Georesources Engineering at the same department.

He got the scholarship by PARE Program to exchange study and do this thesis during August 2014 – August 2015 at Laboratory of Mineral Processing and Resources Recycling, Division of Sustainable Resources Engineering, Graduate School of Engineering, Hokkaido University, Japan.

