EFFECT OF LEACHING REAGENT ON LEACHING PROCESS OF EUROPIUM, TERBIUM AND YTTRIUM FROM LIQUID CRYSTAL DISPLAY

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I hereby declare that I have checked this thesis and in my opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Master of Science.

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I hereby declare that the work in this thesis is based on my original work except for quotations and citations that have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

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ABSTRAK

Paparan kristal cecair (LCD) monitor menggunakan penapis warna untuk menghasilkan gamut warna dan penapis ini terdiri daripada elemen europium (Eu), terbium (Tb) dan yttrium (Y). Kitar semula serta pemulihan semua elemen ini sangat penting kerana kekurangan elemen nadir bumi (REEs) dan juga kapasiti kelimpahan sisa elektronik (e-sisa), terutamanya monitor LCD yang tersedia di seluruh dunia. Kitar semula serta pemulihan semua elemen ini sangat penting kerana kekurangan REEs dan juga kapasiti kelimpahan sisa elektronik (e-sisa), terutamanya monitor LCD yang tersedia di seluruh dunia. Dalam hal ini, salah satu langkah penting dalam aktiviti pemulihan melibatkan proses larut-lesap. Oleh itu, kelakuan pelarutan unsur-unsur dalam larutan berair perlu difahami secara kritikal untuk menentukan urutan keutamaan REEs yang akan diekstrak dari sisa LCD. Kajian ini memberi tumpuan kepada proses pelaurtan Eu, Tb, dan Y dengan menggunakan asid hidroklorik (HCl), sulfurik (H_2SO_4) dan nitrik (HNO_3), di mana ia secara umum dibahagikan kepada dua bahagian utama. Bahagian pertama berkaitan dengan analisis sifat termodinamik, di mana gambarajah Pourbaix telah digunakan sebagai alat yang terutama mewakili kestabilan pelarutan. Keputusan menunjukkan bahawa Eu, Tb, dan Y larut dalam asid HCl, H_2SO_4 dan HNO_3 masing-masing sebagai kation individu dan membentuk kompleks samada dengan ligan dan radikal lain. Sementara itu, fasa kedua kajian ini berkenaan dengan keberkesanan pelarutan melalui ujian analisis berdasarkan dua mod eksperimen - satu dan dua peringkat proses pelarutan. Jumlah Eu, Tb, dan Y masing-masing diukur menggunakan teknik XRF dan ICP-MS. Namun demikian, teknik XRF tidak dapat mengesan sebarang REEs elemen kerana kepekatan REEs yang kecil berbanding dengan komposisi lain. Walau bagaimanapun, keputusan analisis ICP-MS menunjukkan bahawa hanya elemen Y yang dapat dilarut dengan baik sama ada oleh satu atau dua peringkat. Dua lagi unsur yang terdiri daripada Eu dan Tb, didapati dalam kuantiti yang sangat rendah dan sangat sukar untuk di ekstak. Di samping itu, kedua-dua reagen HNO_3 dan H_2SO_4 dikenal pasti sebagai pilihan terbaik untuk digunakan sebagai reagen pelarutan berbanding dengan HCl. Analisis struktur morfologi terhadap sampel yang tersisa terutamanya selepas pelarutan menunjukkan bahawa morfologi sampel H_2SO_4-HNO_3 dan HNO_3-H_2SO_4 relatif tinggi dalam kepadatan. Ini mungkin disebabkan unsur-unsur silikon yang tidak larut semala proses pelarutan. Ia boleh dirumuskan bahawa hanya elemen Y yang boleh diekstrak daripada Eu atau Tb dari e-sisa LCD.

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ABSTRACT

Liquid crystal display (LCD) monitor uses color filter to produce color gamut and this particular filter is mainly made up from europium (Eu), terbium (Tb) and yttrium (Y) of Rare Earth Elements (REEs). Recycling as well as recovering of all these elements are of particularly important due to the scarcity of REEs and also the abundance capacity of electronic waste (e-waste), especially LCD monitors that available worldwide. In this regard, one of the crucial steps in recovering activities is involving leaching process. Thus, the dissolution behavior of the elements in the aqueous solution needs to be critically understood in order to determine the priority sequence of the REEs that to be extracted from the LCD scraps. This study focuses on the dissolution process of Eu, Tb, and Y using hydrochloric (HCl), sulfuric (H_2SO_4) and nitric (HNO_3) acids respectively, whereby it is generally divided into two main parts. The first part relates to the thermodynamic property analysis, whereby Pourbaix diagram has been utilized as the tool in mainly representing the dissolution stability. The results showed that Eu, Tb, and Y are dissolved in HCl, H_2SO_4 and HNO_3 acids respectively as an individual cations and forming complex with ligand and other radical. Meanwhile, the second phase of the study deals with assessing the dissolution effectiveness via analytical testing, particularly based on two experimental modes – single and two stages of dissolution processes. Regarding this, the amount of Eu, Tb, and Y were measured respectively using XRF and ICP-MS techniques. Unfortunately, the XRF method was unable to detect any of the elements due to their very small REEs concentration relative to other compositions. However, the results of ICP-MS analysis have shown that only Y element was dissolvable feasibly either by single or two-stage steps. This observation also suggests that the other two elements, which consist of Eu and Tb, are extremely low in quantity and complicated to be recovered. In addition, both HNO_3 and H_2SO_4 solutions were identified as the best option to be utilized as the dissolution reagents relative to HCl. The morphology structure analysis on the remaining samples particularly after the dissolution completed indicates that the morphology of H_2SO_4-HNO_3 as well as HNO_3-H_2SO_4 samples are relatively richer in the molecules compactness. This is perhaps due to the silicon elements that did not dissolve effectively during the dissolution process. It can be summarized that only Y element is practically feasible to be extracted rather than Eu or Tb from the LCD e-waste.

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LIST OF SYMBOLS

%	Percentage
°C	Celcius
µ	Micro
aq	Aqueous
Cp	Heat capacity
e	Electron
E0	Standard electrode potential
Eh	Activity of electron
g	Gas
g/cm^3	Density
g/mol	Molar mass
J/mol·K	Standard entropy
K	Kelvin
k	Reaction constant
Keq	Equilibrium constant
kPa	Kilopascal
l	Liquid
ml	Milliliter
mmHg	Pressure
pH	Potential of hydrogen
R	Gas constant
s	Solid
T	Temperature
V	Voltage
ΔG	Gibbs free energy of formation

LIST OF ABBREVIATIONS

ARE	Asian Rare Earth
BCC	Body-Centered Cubic
BISC	The Baotou Iron And Steel Company
BSE	Backscattered Secondary Electron
CdS	Cadmium Sulphide
CRT	Cathode Ray Tube
DOE	Department Of Enviroment
DVB	Divinylbenzene Bridge
EDTA	Ethylene Diamine Tetra Acetate
FE-SEM	Field Emission Scanning Electron Microscopy
HEDTA	N-Hydroxyl Ethyethylendiaminetri Acetate
HREE	Heavy Rare Earth Element
HTP	High-Temperature Process
ICP-MS	Inductively Coupled Plasma Mass Spectroscopy
IRE	Indian Rare Earth
LAMP	Lynas Advance Material Plant
LCD	Liquid Crystal Display
LREE	Light Rare Earth Element
MAREC	Malaysian Rare Earth Corporation
NiMH	Nickel-Metal Hydride
PPB	Part Per Billion
PPM	Part Per Million
ppq	Part Per Quadrillion
R&D	Research And Development
RE	Rare Earth
REE	Rare Earth Element
REO	Rare Earth Oxide
RGB	Red, Green, and Blue
SE	Secondary Electron
TFT	Thin-Film Transistor
UV	Ultra-Violet

XRF	X-Ray Fluorescent
YAG	Yttrium-Aluminum-Garnet

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