FEASIBILITY OF USING CARBON DOPED ALUMINIUM OXIDE CO-DOPED WITH MAGNESIUM AS A THERMOLUMINESCENCE DOSIMETER

AINUN JARIAH AWANG

UNIVERSITI TEKNOLOGI MALAYSIA

FEASIBILITY OF USING CARBON DOPED ALUMINIUM OXIDE CO-DOPED WITH MAGNESIUM AS A THERMOLUMINESCENCE DOSIMETER

AINUN JARIAH BINTI AWANG

A thesis submitted in fulfilment of the
requirements for the award of the degree of
Doctor of Philosophy (Physics)

Faculty of Science
Universiti Teknologi Malaysia

APRIL 2017

DEDICATION

To my beloved mom

My late father

My supportive in laws

My loving husband

My precious daughter

My greatest family

&

My loyal friends

ACKNOWLEDGEMENT

In the name of Allah, the Most Gracious and the Most Merciful, Alhamdulillah, all praises to Allah s.w.t. for the strengths and His blessings upon completing this thesis. First and foremost, I would like to express my greatest gratitude to my supervisor and ex-supervisor, Dr. Abd Khamim bin Ismail and Prof. Dr. Husin bin Wagiran for their unending support, supervision, guidance and advice throughout this study. My deepest appreciation to my co-supervisor, Dr. Mohamad Alam Saeed and to all Health Physics Research Group members for being very helpful, informative and supportive all the way in my postgraduate years. Thank you to En. Saiful, En. Abdul Rahman, En. Jaafar, and Pn. Anisah for their assistance in my laboratory works. I am doubtly grateful to Nuclear Science Department, Universiti Kebangsaan Malaysia, Radiotherapy and Oncology Department of National Cancer Institute in Putrajaya and Oncology Treatment Center of Hospital Sultan Ismail in Johor Bahru for giving an outstanding guidance and help for sample irradiation process in this study, in particular, En. Ahmad Takim Saring, Pn Mahzom Pawanchek, Pn Fiza, En Hassan Ali, dan Cik Zati Hani. Finally, to my parents and best friends, thank you for your support, help and consideration throughout this project.

ABSTRACT

Nano-polycrystalline samples of aluminum oxide doped carbon (Al_2O_3:C) and aluminum oxide doped carbon co-doped with magnesium (Al_2O_3:C, Mg) were investigated in search for the most efficient thermoluminescence (TL) dosimeter with low sensitivity to light and applicable for various radiation energy. Six samples were prepared and the samples were found to be nano-polycrystalline powder with particle size in the range of 30 to 250 nm through X-ray Diffraction (XRD) and Transmission Electron Microscope (TEM) analysis. Those samples were exposed to Cobalt-60 gamma rays to determine the optimum concentration of dopant. The optimum concentration of dopant in atomic weight percentage (at%) is 0.2 at% of carbon dopant for doped sample (C2) and 0.2 at% of magnesium dopant of co-doped sample (C2MG2). Samples with optimum dopant were irradiated by 10 and 12 MeV electrons, 6 and 10 MV high energy x-rays over a lower dose range 0.5 to 4.0 Gy from linear accelerator (LINAC) and by Cobalt-60 gamma rays over the dose ranges of 0.5 to 4.0 Gy, and a higher dose range 10 to 100 Gy. Sample C2 and C2MG2 exhibit two TL peaks at higher dose while single peak at lower dose. Sample C2MG2 shows a linear dose response over a dose range of 0.5 to 4.0 Gy subjected to 10 and 12 MeV electrons and 6 and 10 MV high energy x-rays. Sample C2MG2 also shows a linear dose response over a dose range of 0.5 to 4.0 Gy and 10 to 100 Gy when exposed to Cobalt-60 gamma rays. TL sensitivity of sample C2MG2 is 14.22 higher than that of sample C2 when exposed to Cobalt-60 gamma rays. TL sensitivity of sample C2MG2 subjected to electron and high energy x-rays irradiation is still higher than that of sample C2 but lower in value compared to TL sensitivity of sample C2MG2 subjected to Cobalt-60 gamma rays. Addition of Mg has also minimized thermal fading of sample C2MG2 and improved reproducibility significantly for all delivered energy. However, sample C2MG2 is more sensitive to sunlight compared to sample C2, and both samples were not sensitive to fluorescence light even after 24 h of exposure. The minimum detectable dose of sample C2MG2 subjected to Cobalt-60 gamma rays, electron and high energy x-rays are 2.76, 12.49 and 13.83 mGy, respectively. TL properties of sample C2MG2 are influenced by the energy of electrons and photons. Sample C2MG2 also shows good TL properties as TL dosimeter. The measured effective atomic number, Z_{eff} of sample C2MG2 is 11.14 and can be considered as bone-equivalence material. Kinetic analysis revealed that the glow curve of sample C2MG2 followed general kinetic order. Using four different methods, the activation energies were calculated to be in the range of 1.08 to 1.70 eV and frequency factor is between 10^{12} to 10^{19} s^{-1}. The findings of the study show that nano-polycrystalline Al_2O_3:C, Mg is less affected by the light and has a capability to be used in radiation dose monitoring.

ABSTRAK

Sampel nano-polihablur aluminium oksida berdopkan karbon (Al_2O_3:C) dan aluminium oksida berdopkan karbon dan diko-dopkan bersama magnesium (Al_2O_3:C, Mg) telah diselidiki untuk memperoleh dosimeter termopendarcahaya (TL) yang paling cekap dengan rendah peka cahaya dan berguna untuk pelbagai tenaga sinaran. Enam sampel telah disediakan dan sampel ini telah ditemui sebagai serbuk nano-polihablur dengan zarah bersaiz dalam julat 30 hingga 250 nm melalui analisis belauan sinar-X (XRD) dan mikroskopi penghantaran elektron (TEM). Sampel tersebut didedahkan kepada sinar gama Cobalt-60 untuk menentukan kepekatan dopan optimum. Kepekatan dopan optimum dalam peratusan jisim atom (at%) ialah 0.2 at% dopan karbon bagi dop sample (C2) dan 0.2 at% dopan magnesium bagi ko-dop sampel (C2MG2). Sampel ini disinari elektron bertenaga 10 dan 12 MeV, sinar-x bertenaga tinggi 6 dan 10 MV pada julat dos rendah 0.5 hingga 4.0 Gy dari mesin pemecut linear (LINAC) dan sinar gama Cobalt-60 pada julat dos 0.5 hingga 4.0 Gy dan julat dos tinggi dari 10 hingga 100 Gy. Sampel C2 dan C2MG2 mempamerkan dua puncak TL pada julat dos tinggi dan satu puncak tunggal pada julat dos rendah. Sampel C2MG2 menunjukkan sambutan dos yang linear pada julat dos 0.5 hingga 4.0 Gy terhadap penyinaran elektron bertenaga 10 dan 12 MeV dan sinar-x bertenaga tinggi 6 dan 10 MV. Sampel C2MG2 juga menunjukkan sambutan dos yang linear pada julat dos 0.5 hingga 4.0 Gy dan 10 hingga 100 Gy setelah didedahkan pada sinar gama Cobalt-60. Kepekaan TL bagi sampel C2MG2 adalah 14.22 lebih tinggi daripada kepekaan TL sampel C2 setelah didedahkan pada sinar gama Cobalt-60. Kepekaan TL sampel C2MG2 yang dikenakan elektron dan sinar-x bertenaga tinggi, masih lebih tinggi daripada kepekaan TL sampel C2 tetapi lebih rendah nilainya berbanding kepekaan TL apabila disinari sinar gama Cobalt-60. Penambahan Mg turut meminimumkan kepudaran haba bagi sampel C2MG2 dan menambah baik kebolehgunaan semula dengan ketara bagi semua tenaga yang diberi. Walau bagaimanapun, sampel C2MG2 ditemui lebih peka terhadap cahaya matahari berbanding sampel C2 dan kedua-dua sampel tidak peka terhadap cahaya pendarfluor walaupun selepas 24 jam pendedahan. Dos boleh kesan minimum oleh sampel C2MG2 setelah disinari dengan sinar gama Cobalt-60, elektron dan sinar-x bertenaga tinggi, masing-masing ialah 2.76, 12.49 dan 13.83 mGy. Sifat TL bagi sampel C2MG2 adalah dipengaruhi oleh tenaga elektron dan foton. Sampel C2MG2 juga menunjukkan sifar TL yang baik sebagai dosimeter TL. Nombor atom berkesan, Z_{eff} yang diukur bagi C2MG2 ialah 11.14 dan ini merupakan bahan setara tulang. Analisis kinetik mengesahkan bahawa lengkung berbara C2MG2 mengikut tertib kinetik umum. Menggunakan empat kaedah berbeza, tenaga pengaktifan dihitung berada pada julat 1.08 hingga 1.70 eV dan faktor frekuensi antara 10^{12} hingga 10^{19} s^{-1}. Keputusan kajian menunjukkan bahawa nano-polihablur Al_2O_3:C, Mg kurang peka terhadap cahaya dan mempunyai keupayaan sebagai pemantau dos sinaran.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATION

CF	-	Calibration factor
CS	-	Combustion synthesis
D_{LDL}	-	Lower detection limit
DREO	-	Defense Research Establishment Ottawa
EDX	-	Energy Dispersive X-ray
FESEM	-	Field Emission Scanning Electron Microscope
FOM	-	Figure of merit
FWHM	-	Full width at half maximum
HSI	-	Hospital Sultan Ismail
ICDD	-	International Centre for Diffraction Data
ICSD	-	Inorganic Crystal Structure Database
IEC	-	International Electrotechnical Commission
IKN	-	Institut Kanser Negara
LED	-	Light-emitting diode
LINAC	-	Linear accelerator
LLD	-	Lowest level of detection
MDD	-	Minimum detectable dose
MEACR	-	Mass energy absorption coefficient ratio
NIST	-	National Institute of Standards and Technology
OSL	-	Optically Stimulated Luminescence
PC	-	Personal computer
PDD	-	Percentage depth dose
PMT	-	Photomultiplier tube
REG	-	Radiation Effect Group
RER	-	Relative energy response
RL	-	Radioluminescence
SDD	-	Source detector distance

TEM	-	Transmission Electron Microscope
TL	-	Thermoluminescence
TLD	-	Thermoluminescence dosimeter
TSPs	-	Thermally Stimulated Processes
TTP	-	Time temperature profile
UK	-	United Kingdom
UKM	-	Universiti Kebangsaan Malaysia
USA	-	United States of America
UV	-	Ultraviolet
WinREMS	-	Windows Radiation Evaluation and Management system
WF	-	Wedge factor
XRD	-	X-ray Diffraction

LIST OF SYMBOLS

a_i	-	Fraction electron content of element *i*
$\overline{B}$	-	Mean TL background signal from annealed and un-irradiated dosimeter
b	-	Order of kinetics
D	-	Absorbed dose
D_{TLD}	-	Absorbed dose of the TLD
D_o	-	Minimum detectable dose
E	-	Activation energy
F	-	Calibration factor in Gy/TL unit
F_o	-	Intercept-y ftom graph TL response against absorbed dose
$F(D)$	-	Dose response at dose D
$F(D_1)$	-	Dose response at the lowest dose D_1
$F(D)_{material}$	-	Sensitivity of a material
$F(D)_{TLD-100}$	-	Sensitivity of TLD-100
$f(D)$	-	Linearity index
I	-	TL intensity
I_M	-	Maximum TL intensity
k	-	Boltzmann's constant
$\overline{M}_i$	-	Average TL signal of each dose level
M_j	-	Reading of the *j*th dosimeter
m	-	Slope from dose response graph
N	-	Trap concentration (cm^{-1})
N_A	-	Avogadro's number
n	-	Number of trapped electron
n_o	-	Initial number of trapped electron at time $t = 0$
p	-	Probability escape of an electron

R^2	-	Regression coefficient
S	-	Sensitivity
$S(D)$	-	Relative sensitivity
$S_E(E)$	-	Photon energy response
s	-	Frequency factor
s'	-	Pre-exponential factor
T	-	Absolute temperature
T_M	-	Maximum temperature
T_1	-	Temperature of half intensity on the rising side of glow curve
T_2	-	Temperature of half intensity on the falling side of glow curve
t	-	Time
W_i	-	Weight fraction of the element i
Z_{eff}	-	Effective atomic number
α	-	Alpha-phase
θ	-	Bragg angle
ρ	-	Density
ϕ	-	Fluence
μ	-	Geometric factor
$\tau_{1/2}$	-	Half-life of a trap
β	-	Heating rate
τ	-	Mean lifetime
σ_B	-	Standard deviation from mean background signal
δ	-	High temperature half width
τ	-	Low temperature half width
ω	-	Total half intensity width
λ	-	Incident wavelength

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Overview

Ionizing radiation dosimetry plays a very important role in several fields such as radiotherapy, nuclear medicine diagnosis, nuclear medicine, earth science, food irradiation, geological and archaeological dating methods, etc. Nowadays, one of the most useful dosimetry is thermoluminescence dosimetry where it is generally acknowledged to be the most widely used and cost-effective technique for radiation dosimetry (ionizing and non-ionizing), being almost certainly the most popular technique for routine monitoring of occupational and medical radiation exposure (Portal, 1981). In medicine, thermoluminescence dosimeter (TLD) is used to measure radiotherapy absorbed dose and surface entrance due to diagnostic imaging procedure. In industrial field, it is used for environmental monitoring and personal monitoring equipment for radiation workers (Abdullah, 2011).

The investigation on dosimetric technique has shown promising future of thermoluminescence (TL) and has expanded enormously by different materials, design and variable after the discovery of Daniels and his co-workers in 1940s. When new materials of TL are introduced, they come with advantages and disadvantages in a package. Hence, it boosts more researchers to work more deeply in this TL area. The development of this research is important because each small finding either positive or negative could be the small step for a bigger success. The materials used in TLD also extended into various forms such as solid pellet, chips, nanoparticle, optical fibers, powders and thin films. In early years, TL materials also widespread from lithium fluoride (LiF) to calcium sulphate ($CaSO_4$), calcium fluoride (CaF_2), aluminum oxide (Al_2O_3) and beryllium oxide (BeO). These materials were also doped and co-doped with various dopants such as, magnesium (Mg), titanium (Ti), cuprum (Cu), phosphorus (P), manganese (Mn), dysprosium (Dy), carbon (C), europium (Eu), yttrium (Y), terbium (Tb) and thulium (Tm).

1.2 Research Background

As mentioned in section 1.1, there are many materials that have been introduced and examined as a good TL candidate. Al_2O_3 is one of the materials that was first experienced as a TL dosimeter. Later, it received much less attention because of higher competition with other sensitive phosphors, and apart from several isolated studies, it was forgotten (Portal, 1986). However, from time to time, nowadays particularly, favorable properties of certain Al_2O_3 material have been demonstrated. In 1990, Akselrod M S and his group has introduced highly sensitive TL α-Al_2O_3:C (TLD-500) single crystal detectors that was found to be 40-60 times higher than LiF:Mg, Ti (TLD-100) (Akselrod et al., 1990a). This finding encouraged other researchers to believe in Al_2O_3 material and recognize that dopants in Al_2O_3 played a very important role in producing enhanced TL dosimetry.

In α-Al_2O_3:C, carbon impurities play important role to produce a highly sensitive TLD. It was understood that, there are many factors that influence TL properties in production of TLDs such as type of used material, the amount and the type of impurities and intentional dopants, their chemical bond and method of introduction into the lattice, the thermal, optical and mechanical treatment of the material. These parameters decide that how many electrons could trap in the forbidden region and how deep the electron have been trapped during a TL process (McKinlay, 1981). From earlier studies, it is known that Al_2O_3 has many useful properties as a dosimeter, such as, linearity in a wide dose range, mechanical resistance, easy handling and cheapness (Osvay and Biro, 1980). From the useful properties of Al_2O_3 material, one can make the most use of it, if one can overcome any of it limitation.

In addition to the kind of TL material, radiation type also has a significant role in TL properties. There are many type of radiation that have been used and studied in TL area, including X-rays and radiation particles such as an electron, photon, and neutron (James and Farrington, 1957, Mehta and Sengupta, 1976, Hashim et al., 2009, Azziz et al., 1997, Nikiforov et al., 2014, Leong et al., 2015). This type of radiation exposure is used in medical field, environmental monitoring, personnel monitoring, and food industries that incoherence with increasing TLD usage. Upon this reason, TL dosimeter should have the ability to measure all type of radiation energy with a wide range of dose independently. Moreover, none of the manufactured TLDs can measure all types of radiation energy independently. So, the high sensitivity of TLD-500 should be used wisely and its ability should be improvised in measuring radiation dose.

1.3 Research Problem

It is widely known that, commercialized α-Al_2O_3:C has higher sensitivity than pioneer LiF: Mg, Ti dosimeter. However, higher sensitivity does not mean popular choices, since LiF: Mg, Ti still the favorite dosimeter that is used and studied because it has a lot more information available after 75 years of its study. While α-Al_2O_3:C has been studied for about 25 years, yet there are still lacks of knowledgeable information. Hence there are still lots of rooms for the improvement in α-Al_2O_3:C dosimeter. Despite being an unpopular choice, there are still workplaces or laboratories that favored TLD-500 over TLD-100H like Defense Research Establishment Ottawa (DREO). Radiation Effect Group (REG) at DREO chooses TLD-500 because it is better suited to DREO’s need (Erhardt et al., 2001). This situation exactly fits the need of producing a better TLD because different places or people would acquire different needs. Therefore, different types of dosimeters are needed.

It is also well-known that TLD-500 experiences significant light-induced effects (Bos, 2001, Erhardt et al., 2001, McKeever et al., 1995, McKeever and Moscovitch, 2003, Moscovitch et al., 1993). This is one of its disadvantages despite being a phenomenal sensitive dosimeter. The light induced effects can be explained in two ways. The light-induced TL increases the TL intensity, while light-induced fading decreases the TL intensity. So, most of researchers or users of TLD-500 have to take a great care towards this dosimeter to avoid any visible light exposure that stimulates light-induced effects. As in DREO, a careful lab practice is implemented upon the using of TLD-500 to avoid light-induced background or fading under laboratory light. This is also the reason to improve this dosimeter light sensitivity by using a special case or apply some changes in its material. The light-induced effect is also a fading characteristic that could depend upon impurity concentration, annealing temperature and times, cooling rates and storage temperatures (Chen and McKeever, 1997). Hence, changing and/or addition of new impurity, improve the temperature and time of annealing or cooling could be the solution for minimizing the light-induced effect or even better to make it disappear. It may not be a perfect success but it can be a progressive way toward success.

Hence, in order to produce new progression for a better dosimeter, Mg element is chosen to co-doped in alumina material where Mg shows prominent features in heightened host material sensitivity and small changes in its composition give strong effects to main peak intensity of LiF:Mg, Cu, P dosimeter (Knezevic´ et al., 2010, Chen and Stoebe, 2002). In addition, Al_2O_3:C, Mg luminescence detector does not require light protection when studied for radiation-induced fluorescence (Akselrod and Akselrod, 2006). Adding Mg could be the solution of light sensitivity characteristic for Al_2O_3 family for TL material. Rodriguez (2010) also stated in his thesis that annealing of single crystal Al_2O_3:C, Mg would result in increasing of recombination centers that would improve its sensitivity comparable to Al_2O_3:C or even greater than Al_2O_3:C in certain dosimetric applications (Rodriguez, 2010). In Saharin et al. (2014) study of TL in Al_2O_3:C, Mg under 5 – 70 Gy gamma irradiation prove that, adding Mg as a co-dopant enhanced TL response. Hence, Mg element is a great choice of impurity, as it has shown remarkable features of increasing TL sensitivity and can avoid unnecessary light protection.

Most of the TL dosimeters are in the form of crystal because TL sensitivity of crystal is higher than other phase due to TL mechanism involved. TLD-500 which is in the form of single crystal that can be prepared using Czochralski method, precipitation and evaporation method, chemical vapor deposition, etc. These are complex preparation methods to achieve single crystal material. Thus, study aims to prepare new material without disregarding the crystal form but with some modification in preparation method by choosing polycrystalline form. The polycrystalline preparation method is easier than crystalline growth method, where it can be produced using hot pressed method. The hot pressed method is a technique where desire composition is mixed. Then, the mixture is put in a graphite mold, heat is provided and the sample is pressed. Hence, it is simpler, cheaper and easier method to employ with well-equipped laboratories. In addition, nanostructured TL material has shown prominent enhancement in TL response of high dose in many study (Bitencourt and Tatumi, 2009, de Azevedo et al., 2006, Prathibha et al., 2014, Salah et al., 2011). However, they were less studied for lower dose measurement and light induced effect occurs mainly in single microcrystalline form. Therefore, nano-polycrystalline sample with nanoparticle size is preferred for this study.

Considering these above said factors, carbon doped alumina co-doped with magnesium (Al_2O_3:C, Mg) nano-polycrystalline powder was carefully chosen for TL study as an alternative to single crystal TLD-500. To the best of our knowledge, no other study had synthesized a nano-polycrystalline Al_2O_3:C, Mg for TL properties measurement. Furthermore, TL performance of this newly nano-polycrystalline Al_2O_3:C, Mg comprised of various ionizing radiation such as ^{60}Co gamma rays (Co-60), high energy x-rays (6 and 10 MV) and high energy electron (10 and 12 MeV) with a wide range of doses assessed at room temperature. This study also emphasize on the enhancement of light-sensitivity of the newly proposed TLD.

1.4 Research Objectives

This research embarks on the following objectives:

1. To characterize crystalline state and material composition of Al_2O_3:C, Mg.
2. To determine TL dosimetric properties of Al_2O_3:C, Mg irradiated with various x-rays (1.25 MeV Co-60, 6 and 10 MV) and electron energies (10 and 12MeV).
3. To evaluate the TL kinetic parameter of the Al_2O_3:C, Mg irradiated to various ionizing radiation (Co-60 gamma, high energy x-rays and electron).

1.5 Research Scope

This research emphasis on the Al_2O_3:C, Mg powder as a TL material. The selected samples of Al_2O_3:C and Al_2O_3:C, Mg powder undergo structural and composition elements measurement using; X-ray diffraction (XRD) analysis, Transmission Electron Microscopy (TEM) analysis and Field Emission Scanning Electron Microscope – Energy Dispersive X-ray (FESEM-EDX) elemental analysis. Al_2O_3:C, Mg and Al_2O_3:C were then exposed to various photons (Cobalt-60 at Universiti Kebangsaan Malaysia, 6 and 10 MV at Institut Kanser Negara and Hospital Sultan Ismail) and high energy electron (10 and 12 MeV at Institut Kanser Negara and Hospital Sultan Ismail) at the same time. This experiment is designed to investigate the relationship of annealing temperature and time with TL properties, TL glow curve, linearity, sensitivity, fading, minimum detectable dose and reproducibility according to the exposed radiation. Both materials are compared in order to analyze the TL properties of Al_2O_3:C, Mg powder as new developed TL material from Al_2O_3:C family. Photon energy response for Al_2O_3:C, Mg powder is calculated using the mass energy absorption coefficient ratio (MEACR) method. The kinetic parameters also obtained using initial rise method, peak shape method, whole glow peak method, and curve fitting method to understand TL phenomenon and trap nature of the chosen material.

1.6 Research Significances

The relevance of the present study relates to the solving of problems and the needs of technology to prepare better and efficient TL material for various applications. However, this outstanding TL material cannot be achieved without great research and deep understanding in this area. Thus, detailed study of the enhanced TL characteristic by adding co-dopant into host material becomes essential to determine the effect and thus contributes to further material development. The effect could be explained by TL properties in favor of this study that reckoned to be useful in TL study which provides the knowledge on the suitability of Al_2O_3:C, Mg powder as a radiation measurement tool. In addition, this study would also offer valuable information on characterizing and handling Al_2O_3:C, Mg powder as TL material.

1.7 Outline

This thesis describes the TL characterization of carbon doped alumina co-doped with magnesium (Al_2O_3:C, Mg) irradiated by various type of ionizing radiation. The thesis is divided into five chapters. Chapter 1 is the introduction of the study, which explains the purpose of this study, its importance, and the justification of material selection. Chapter 2 describes some background knowledge on TL phenomenon, review on the previous works done on alumina material in TL area, and basic principles of TL properties engaged in this study. Details of the sample preparation, design of the experimental and the measurement techniques employed are outlined in Chapter 3. In Chapter 4, all the experimental results and discussion are presented. Finally, Chapter 5 presents the conclusions of the research and future outlook of the study. Additional information associated with this thesis is given in the Appendices.

REFERENCES

Abdullah, N. A. F. 2011. *The Study on thermoluminescence Response of Ge-doped Optical Fibre using X-Ray Irradiation.* Bachelor of Science (Health Physics), University Technology Malaysia.

Agersnap Larsen, N., Bøtter-Jensen, L. & McKeever, S. W. S. 1999. Thermally stimulated conductivity and thermoluminescence from Al_2O_3:C. *Radiat Prot Dosimetry,* 84**,** 87-90.

Akselrod, A. E., Larsen, N. A., Whitley, V. H. & McKeever, S. W. S. 1999. Thermal quenching of F centre luminescence in Al_2O_3:C. *Radiat Prot Dosimetry,* 84**,** 39-42.

Akselrod, M. S. & Akselrod, A. E. 2006. New Al_2O_3:C,Mg crystals for radiophotoluminescent dosimetry and optical imaging. *Radiat Prot Dosimetry,* 119**,** 218-21.

Akselrod, M. S. & Kortov, V. 1990. Thermoluminescent and Exoemission Properties of New High-Sensitivity TLD α-Al_2O_3:C Crystals. *Radiat Prot Dosimetry,* 33**,** 123-126.

Akselrod, M. S., Kortov, V. & Gorelova, E. A. 1993. Preparation and Properties of α-Al_2O_3:C. *Radiat Prot Dosimetry,* 47**,** 159-164.

Akselrod, M. S., Kortov, V., Kravetsky, D. J. & Gotlib, V. I. 1990a. Highly sensitive thermoluminescent anion-defect α-Al_2O_3:C single crystal detectors. *Radiat Prot Dosimetry,* 33**,** 119-122.

Akselrod, M. S., Kortov, V., Kravetsky, D. J. & Gotlib, V. I. 1990b. Highly sensitive thermoluminescent anion-defective α-Al_2O_3:C single crystal detectors. *Radiat Prot Dosimetry,* 32**,** 15-20.

Arrieta, A., Escobar-Alarcón, L., Camps, E. & González, P. R. 2007. Thermoluminescent response of aluminium oxide thin films subject to gamma irradiation. *Radiation Effects and Defects in Solids,* 162**,** 745-749.

Attix, F. H. 2008. *Introduction to Radiological Physics and Radiation Dosimetry*, Wiley.

Azorin, J., Esparza, A., Falcony, C., Rivera , T., Garcia, M. & Martinez, E. 2002. Preparation and Thermoluminescence Properties of Aluminium Oxide Doped with Europium. *Radiation Protection Dosimetry,* 100**,** 277-279.

Azziz, N., Ranogajec-Komor, M., Marino, S. A., Klemic, G. A. & Osvay, M. Fast neutron detection using aluminum oxide TLDs. *In:* Sabol, J., ed. IRPA Regional Symposium on Radiation Protection in Neighbouring Countries of Central Europe, 1997 Prague, Central Europe. International Radiation Protection Association, 1997, 495-497.

Barros, V. S. M., Azevedo, W. M., Khoury, H. J. & Filho, P. L. 2010a. Preparation and Characterization of Thermoluminescent Aluminium Oxide Doped with $Tb^{3+}$and Tb^{3+}-Mg^{2+}. *Journal of Physics: Conference Series,* 249**,** 012025.

Barros, V. S. M., de Azevedo, W. M., Khoury, H. J., Andrade, M. E. A. & Filho, P. L. 2010b. Thermoluminescence study of aluminum oxide doped with therbium and thulium. *Radiation Measurements,* 45**,** 435-437.

Bitencourt, J. F. S. & Tatumi, S. H. 2009. Synthesis and Thermoluminescence Properties of Mg^{2+} Doped of Nanostructured Aluminium Oxide. *Physics Procedia,* 2**,** 501-514.

Bos, A. J. J. 2001. High Sensitivity Thermoluminescence Dosimetry. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms,* 184**,** 3-28.

Bos, A. J. J., Vijverberg, R. N. M., Piters, T. M. & McKeever, S. W. S. 1992. Effects of cooling and heating rate on trapping parameters in LiF:Mg, Ti crystals. *J. Phys. D: Appl. Phys.,* 25**,** 1249-1257.

Busuoli, G. 1981. General characteristics of TL materials. *In:* Oberhofer, M. & Scharmaan, A. (eds.) *Applied Thermoluminescence Dosimetry.* Great Britain: Adam Hilger Ltd.

Cava, S., Tebcherani, S. M., Souza, I. A., Pianaro, S. A., Paskocimas, C. A., Longo, E. & Varela, J. A. 2007. Structural characterization of phase transition of Al_2O_3 nanopowders obtained by polymeric precursor method. *Materials Chemistry and Physics,* 103**,** 394-399.

Chen, R. 1969a. Glow Curve With General Order Kinetics *Journal of Electrochemical Society,* 116**,** 4.

Chen, R. 1969b. On the Calculation of Activation Energies and Frequency Factors from Glow Curves. *Journal of Applied Physics,* 40**,** 570-584.

Chen, R. 1969c. On the calculation of activation energy and frequency factors from glow curves *Journal of Applied Physics,* 40**,** 16.

Chen, R. & McKeever, S. W. S. 1994. Characterization Of Non Linearities In The Dose Dependence Of Thermoluminescence. *Radiation Measurements,* 23**,** 7.

Chen, R. & McKeever, S. W. S. 1997. *Theory of Thermoluminescence and Related Phenomena*, World Scientific.

Chen, R. & Pagonis, V. 2011. Advanced Methods for Evaluating Trapping Parameters. *Thermally and Optically Stimulated Luminescence.* John Wiley & Sons, Ltd.

Chen, T. C. & Stoebe, T. G. 2002. Role of impurities in the thermoluminescence of LiF:Mg, Cu, P. *Radiation Protection Dosimetry,* 100**,** 243-246.

Chithambo, M. L., Sendezera, E. J. & Davidson, T. 2002. A preliminary thermoluminescence and positron annihilation study of α-Al_2O_3:C. *Radiat Prot Dosimetry,* 100**,** 269-72.

Daniels, F., Boyd, C. A. & Saunders, D. F. 1953. Thermoluminescence as a Research Tool. *Science,* 117**,** 343-349.

Daniels, F. & Saunders, D. F. 1950. The National Academy of Sciences: Abstracts of Papers Presented at the Annual Meeting April 24-26, 1950, Washington, D. C. *Science,* 111**,** 453-463.

de Azevedo, W. M., de Oliveira, G. B., da Silva, E. F., Jr., Khoury, H. J. & Oliveira de Jesus, E. F. 2006. Highly sensitive thermoluminescent carbon doped nanoporous aluminium oxide detectors. *Radiat Prot Dosimetry,* 119**,** 201-5.

de Barros, V. S. M., de Azevedo, W. M., Khoury, H. J. & Linhares Filho, P. 2008. Combustion synthesis: A suitable method to prepare doped materials for thermoluminescent dosimetry. *Radiation Measurements,* 43**,** 345-348.

de Barros, V. S. M., Khoury, H. J., Azevedo, W. M. & da Silva, E. F. 2007. Characterization of nanoporous Al_2O_3:C for thermoluminescent radiation dosimetry. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment,* 580**,** 180-182.

Driscoll, C. M. H., Barthe, J. R., Oberhofer, M., Busuoli, G. & Hickman, C. 1986. Annealing procedures for commonly used radiothermoluminescent materials. *Radiat Prot Dosimetry,* 14**,** 16.

Erhardt, L. S., Prud'homme-Lalonde, L., Cousins, T., Estan, D. & Hoffarth, B. 2001. Comparison of Radiation Response of LiF:Mg,Cu,P and Al_2O_3:C TL Dosimeter. Canada: Defence Research Establishment Ottawa.

Escobar-Alarcón, L., Villagrán, E., Camps, E., Romero, S., Villarreal-Barajas, J. E. & González, P. R. 2003. Thermoluminescence of aluminum oxide thin films subject to ultraviolet irradiation. *Thin Solid Films,* 433**,** 126-130.

Furetta, C. 2003. *Handbook of Thermoluminescence,* Singapore, World Scientific Publishing.

Furetta, C. 2008. *Questions and answers on thermoluminescence (TL) and optically stimulated luminescence (OSL),* Singapore, World Scientific.

Furetta, C., Prokic, M., Salamon, R. & Kitis, G. 2000. Dosimetric characterisation of a new production of MgB_4O_7:Dy, Na thermoluminescent material. *Appl Radiat Isot,* 52**,** 243-250.

Furetta, C. & Weng, P. S. 1998. *Operational Thermoluminescence Dosimetry,* Singapore, World Scientific.

Garlick, G. F. J. & Gibson, A. F. 1948. The Electron Trap Mechanism of Luminescence in Sulphide and Silicate Phosphors. *Proceedings of Physical Soceity,* 60**,** 574-589.

Gimadova, T. I., Bessonova, T. S., Tale, I. A., Avvakumova, L. A. & Bodyachevsky, S. V. 1990. On The Thermoluminescence Mechanism of Non-Doped Corundum Monocrystals with Defect Structure. *Radiat Prot Dosimetry,* 33**,** 47-50.

Göksu, H. Y., Bulur, E. & Wahl, W. 1999. Beta Dosimetry Using Thin-Layer α–Al_2O_3:C TL Detectors. *Radiat Prot Dosimetry,* 84**,** 451-455.

Grossweiner, L. I. 1953. A Note on the Analysis of First-Order Glow Curves. *Journal of Applied Physics,* 24**,** 24.

Gundu Rao, T. K., Bhatt, B. C. & Page, P. S. 2008. Defect centers and thermoluminescence in Al_2O_3:Si,Ti. *Radiation Measurements,* 43**,** 295-299.

Hashim, S., Al-Ahbabi, S., Bradley, D. A., Webb, M., Jeynes, C., Ramli, A. T. & Wagiran, H. 2009. The thermoluminescence response of doped SiO_2 optical fibres subjected to photon and electron irradiations. *Applied Radiation and Isotopes,* 67**,** 423-427.

Hendee, W. R. & Ritenour, E. R. 2003. *Medical Imaging Physics,* Wiley.

Horowitz, Y. S. 1981. The Theoretical and Microdosimetric Basis of Thermoluminescence and Applications to Dosimetry. *Physics in Medicine and Biology,* 26**,** 765-824.

Hubbell, J. H. & Seltzer, S. M. 1995. Tables of X-ray Mass Attenuation Coefficients and Mass Energy-Absorption Coefficients. *In:* Gaithersbug, M. D. (ed.). National Institute of Standards and Technology.

James, K. R. & Farrington, D. 1957. Thermoluminescence studies of aluminum oxide. *J. Phys. Chem.,* 61**,** 629-633.

Kitis, G., Gomez-Ros, J. M. & Tuyn, J. W. N. 1998. Thermoluminescence glow-curve deconvolution functions for first, second and general orders of kinetics. *Journal of Physics D: Applied Physics,* 31**,** 2636-2641.

Kitis, G., Papadopoulos, J. G., Charalambous, S. & Tuyn, J. W. N. 1994. The influence of heating rate on the response and trapping parameters of a-Al_2O_3:C. *Radiat Prot Dosimetry,* 55**,** 8.

Knezevic´, Z., Ranogajec-Komor, M. & Miljanic, S. 2010. Effect of dopants on TL characteristics of LiF:Mg,Cu,P detectors. *Radiation Measurements,* 45**,** 573-575.

Kortov, V. 2007. Materials for thermoluminescent dosimetry: Current status and future trends. *Radiation Measurements,* 42**,** 576-581.

Kortov, V. & Milman, I. I. 1996. Thermally Stimulated Luminescence of Dosimetric α-Al_2O_3 Crystals *Russian Physics Journal,* 39**,** 1122-1134.

Kortov, V., Milman, I. I., Kirpa, V. I. & Lesz, J. 1994. Some features of α-Al_2O_3 dosimetric thermoluminescent crystals. *Radiat Prot Dosimetry,* 55**,** 279-283.

Kortov, V. S. 2010. Nanophosphors and outlooks for their use in ionizing radiation detection. *Radiation Measurements,* 45**,** 512-515.

Kreyling, W. G., Semmler-Behnke, M. & Chaudhry, Q. 2010. A complementary definition of nanomaterial. *Nano Today,* 5**,** 165-168.

Kristianpoller, N., Chen, R. & Israeli, M. 1974. Dose dependence of thermoluminescence peaks. *Journal Of Physics D: Applied Physics,* 7**,** 1063-1072.

Leong, C. Y., Wagiran, H., Ismail, A. K. & Ali, H. 2015. Thermoluminescence characteristics of aluminium oxide doped with carbon and titanium co-doped subjected to 6 and 10 MV photon irradiations. *J Radioanal Nucl Chem*, 1-8.

Lim, T. Y. 2015. *Evaluation of Dysprosium doped Strontium Tetraborate Glass as a Thermoluminescence Dosimeter*. Doctor of Philosophy (Physics), Universiti Teknologi Malaysia.

May, C. E. & Partridge, J. A. 1964. Thermoluminescent Kinetics of Alpha-Irradiated Alkali Halides. *The Journal of Chemical Physics,* 40**,** 1401-1408.

McKeever, S. W. S. 1985. *Thermoluminescence of solids,* United States of America, Cambridge University Press, New York.

McKeever, S. W. S., Akselrod, M. S., Colyott, L. E., Agersnap Larsen, N., Polf, J. C. & Whitley, V. H. 1999. Characterisation Of Al_2O_3 For Use In Thermally And Optically Stimulated Luminescence Dosimetry. *Radiat Prot Dosimetry,* 84**,** 163-168.

McKeever, S. W. S. & Moscovitch, M. 2003. On The Advantages And Disadvantages Of Optically Stimulated Luminescence Dosimetry and Thermoluminescence Dosimetry. *Radiat Prot Dosimetry,* 104**,** 263-270.

McKeever, S. W. S., Moscovitch, M. & Townsend, P. D. 1995. *Thermoluminescence Dosimetry Materials: Properties and Uses,* England, Nuclear Technology Publishing.

McKinlay, A. F. 1981. *Thermo-luminescence Dosimetry,* Adam Hilger Ltd.

Mehta, S. K. & Sengupta, S. 1976. Gamma dosimetry with Al_2O_3 thermoluminescent phosphor. *Physics in medicine and biology,* 21**,** 955-964.

Milman, I. I., Moiseykin, E. V., Nikiforov, S. V., Mikhailov, S. G. & Solomonov, V. I. 2004. Luminescence properties of α-Al2O3 dosimetric crystals exposed to a high-current electron beam. *Radiation Measurements,* 38**,** 443-446.

Mische, E. F. & McKeever, S. W. S. 1989. Mechanisms of Supralinearity in Lithium Fluoride Thermoluminescence Dosemeters. *Radiation Protection Dosimetry,* 29**,** 159-175.

Moscovitch, M., Tawil, R. A. & Svinkin, M. 1993. Light induced fading in α - Al_2O_3:C. *Radiat Prot Dosimetry,* 47**,** 251-253.

Musk, J. H. 1993. Time-dependant and light induced fading in VICTOREEN model 2600-80 aluminium oxide thermoluminescence dosimeters. *Radiat Prot Dosimetry,* 47**,** 247-249.

Nagabushana, K. R., Lakshminarasappa, B. N., Revannasiddaiah, D. & Fouran, S. 2008. Thermally Stimulated Luminescence Studies in Combustion Synthesized Polycrystalline Aluminum Oxide. *Bull Mater Sci,* 31**,** 669-672.

Nelson, V., McLean, D. & Holloway, L. 2010. Thermoluminescent dosimetry (TLD) for megavoltage electron beam energy determination. *Radiation Measurements,* 45**,** 698-700.

Nikiforov, S. V., Kortov, V. & Milman, I. I. 2002. Thermoluminescent dosemeters based on tld-500 detectors for control of neutron fields. *Radiat Prot Dosimetry,* 101**,** 125-128.

Nikiforov, S. V. & Kortov, V. S. 2010. Effect of deep traps on sensitivity of TLD-500 thermoluminescent detectors. *Radiation Measurements,* 45**,** 527-529.

Nikiforov, S. V. & Kortov, V. S. 2014. Dosimetric response for crystalline and nanostructured aluminium oxide to a high-current pulse electron beam. *Radiat Prot Dosimetry,* 162**,** 92-5.

Nikiforov, S. V., Kortov, V. S., Zvonarev, S. V., Moiseykin, E. V. & Kazantseva, M. G. 2014. Basic thermoluminescent and dosimetric properties of Al_2O_3:C irradiated by pulse intensive electron beam. *Radiation Measurements,* 71**,** 74-77.

Ogundare, F. O., Ogundele, S. A., Chithambo, M. L. & Fasasi, M. K. 2013. Thermoluminescence characteristics of the main glow peak in α-Al_2O_3:C exposed to low environmental-like radiation doses. *Journal of Luminescence,* 139**,** 143-148.

Ortega, F., Marcazzo, J., Molina, P., Santiago, M., Lester, M., Henniger, J. & Caselli, E. 2013. Analysis of the main dosimetric peak of Al_2O_3:C compounds with a model of interacting traps. *Appl Radiat Isot,* 78**,** 33-37.

Osvay, M. & Biro, T. 1980. Aluminium Oxide in TL Dosimetry *Nuclear Instruments and Methods,* 175**,** 60-61.

Pagonis, V., Chen, R. & Lawless, J. L. 2007a. A quantitative kinetic model for Al_2O_3:C: TL response to ionizing radiation. *Radiation Measurements,* 42**,** 198-204.

Pagonis, V., Kitis, G. & Furetta, C. 2007b. *Numerical and Practical Exercises in Thermoluminescence,* New York, Springer Science+Business Media, Inc.

Pekpak, E., Yılmaz, A. & Özbayoglu, G. 2010. An Overview on Preparation and TL Characterization of Lithium Borates for Dosimetric Use. *The Open Mineral Processing Journal,* 3**,** 14-24.

Polf, J. C. 2000. *The Role of Oxygen Vacancies in Thermoluminescence Processes* Al_2O_3*:C.* Master of Science, Oklahoma State University.

Portal, G. 1981. Preparation and Properties of Principal TL Products. *In:* Oberhofer, M. & Scharmann, A. (eds.) *Applied Thermoluminescence Dosimetry.* Bristol: Adam Hilger Ltd.

Portal, G. 1986. Review of The Principle Material Available for Thermoluminescent Dosimetry. *Radiat Prot Dosimetry,* 17**,** 351-357.

Pradhan, A. S. 1997. Requirements of Thermoluminescence Dosimeters for Personnel Monitoring. *In:* Bhushan, S. & Dewangan, P. K. (eds.) *Luminescence and Its Applications 97: Proceedings of the National Conference on Luminescence and Its Applications 97 (NCLA-97), October 13 - 15, 1997.* New Delhi: Allied Publisher Limited.

Prathibha, B. S., Chandrashekara, M. S., Nagabhushana, H., Ramesh, K. P., Naghabhushana, B. M. & Chikkahanumanthrayappa 2014. Thermoluminescence Studies on Sr2SiO4 Nano Powder. *Procedia Materials Science,* 5**,** 944-952.

Priharti, W., Samat, B. S. & Kadir, A. B. A. 2013. Uncertainty Analysis of $H_p(10)_{meas}/H_p(10)_{del}$ Ratio for TLD–100H at Energy 24–1250 keV. *Jurnal Teknologi,* 62**,** 115-118.

Randall, J. T. & Wilkins, M. H. F. 1945a. Phosphorescence and Electron Traps. I. The Study of Trap Distributions. *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences,* 184**,** 365-389.

Randall, J. T. & Wilkins, M. H. F. 1945b. Phosphorescence and Electron Traps. II. The Interpretation of Long-Period Phosphorescence. *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences,* 184**,** 390-407.

Randall, J. T. & Wilkins, M. H. F. 1945c. The Phosphorescence of Various Solids. *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences,* 184**,** 347-364.

Ranogajec-Komor, M. 2008. Thermoluminescence Dosimetry-Application in Environmental Monitoring. *Radiation Safety Management,* 2**,** 2-16.

Ranogajec-Komor, M., Vinceković, Ž., Knežević, S. & Miljanić, S. 2002. The Influence Of Thermal Annealing On The Characteristics Of Different Al_2O_3 Thermoluminescence Dosimeters. *Proceeding of European IRPA Congress: Towards Harmonization of Radiation Protection in Europe.* Florence, Italy.

Rathbone, B. A., Endres, A. W. & Antonio, E. J. Evaluation of new and conventional thermoluminescent phosphors for environmental monitoring using automated thermoluminescent dosimeter readers. *In:* Casson, W. H., Thein, C. M. & Bogard, J. S., eds. 4th Conference on Radiation Protection and Dosimetry, October 23-27, 1994 1994 Orlando, Florida. Pacific Nothwest Laboratory, 371-380.

Refaei, A. 2014. *Dosimetric Properties of Neodymium doped Optical Fibers subjected to X-rays and Gamma rays.* Doctor of Philosophy, Universiti Teknologi Malaysia.

Refaei, A., Wagiran, H., Saeed, M. A. & Hosssain, I. 2014. Thermoluminescence characteristics of Nd-doped SiO_2 optical fibers irradiated with the ^{60}Co gamma rays. *Appl Radiat Isot,* 94**,** 89-92.

Rieke, J. K. & Daniels, F. 1957. Thermoluminescence Studies of Aluminum Oxide. *The Journal of Physical Chemistry,* 61**,** 629-633.

Rodriguez, M. G. 2010. *Characterization of Aluminum Oxide Doped with Carbon and Magnesium for Radiation Detection.* Degree of Master of Science, Oklahoma State University.

Rodriguez, M. G., Denis, G., Akselrod, M. S., Underwood, T. H. & Yukihara, E. G. 2011. Thermoluminescence, optically stimulated luminescence and radioluminescence properties of Al_2O_3:C,Mg. *Radiation Measurements,* 46**,** 1469-1473.

Rogojan, R., Andronescu, E., Ghiţulică, C. & Vasile, B. S. 2011. Synthesis and characterization of alumina nano-powder obtained by sol-gel method. *U.P.B. Sci. Bull., Series B: Chemistry and Material Science,* 73**,** 67-76.

Saharin, N. S., Wagiran, H. & Tamuri, A. R. 2014. Thermoluminescence (TL) properties of Al_2O_3: C, Mg exposed to cobalt-60 gamma radiation doses. *Radiation Measurements,* 70**,** 11-14.

Saidu, A. 2015. *Dosimetric Properties of Copper and Terbium doped Zinc Borate glass co-doped Sodium.* Doctor of Philosophy (Physics), Universiti Teknologi Malaysia.

Salah, N., Khan, Z. H. & Habib, S. S. 2011. Nanoparticles of Al_2O_3:Cr as a sensitive thermoluminescent material for high exposures of gamma rays irradiations. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms,* 269**,** 401-404.

Santos, S. P., Santos, S. H. & Toledo, S. P. 2000. Standard Transition Aluminas. ElectronMicroscopy Studies. *Materials Research,* 3**,** 104-114.

Shonka, F. R., Rose, J. E. & Failla, G. 1958. Conducting plastic equivalent to tissue, air and polysterene. *United Nation of International Conference of Peaceful Uses Atomic Energy*. Geneva.

Wagiran, H., Hossain, I., Bradley, D., Yaakob, A. N. H. & Ramli, T. 2012. Thermoluminescence Responses of Photon and Electron Irradiated Ge- and Al-Doped SiO_2 Optical Fibres. *Chinese Physics Letters,* 29.

Walker, F. D., Colyott, L. E., Agersnap Larsen, N. & McKeever, S. W. S. 1996. The Wavelength Dependence of Light-Induced Fading of Thermoluminescence From a-Al_2O_3:C. *Radiation Measurements,* 26**,** 711-718.

Yoshimura, E. M. 2007. Correlation of optically stimulated luminescence and thermoluminescence of Al_2O_3:Fe, Mg, Cr crystals. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment,* 580**,** 606-609.

Yukihara, E. G., Whitley, V. H., Polf, J. C., Klein, D. M., McKeever, S. W. S., Akselrod, A. E. & Akselrod, M. S. 2003. The effects of deep trap population on the thermoluminescence of Al_2O_3:C. *Radiation Measurements,* 37**,** 627-638.

Zahedifar, M., Eshraghi, L. & Sadeghi, E. 2012. Thermoluminescence kinetics analysis of α-Al_2O_3:C at different dose levels and populations of trapping states and a model for its dose response. *Radiation Measurements,* 47**,** 957-964.