THE THERMOLUMINESCENCE RESPONSE OF DOPED SILICON DIOXIDE OPTICAL FIBRES TO IONIZING RADIATION

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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

MARCH 2009

I dedicate this work

To my dear parents

Hjh. Sapiah binti Hasan

Hj. Hashim bin Ahmad

Hjh. Fatimah binti Arajol

Hj. Hassan bin Jalani

Whose love, kindness, patience and prayer have brought me this far

To my beloved wife

Sitti Asmah binti Hassan

For her love, understanding and support through my endeavour

To my children

Muhammad Luqmanul Hakim and Farouq Hakimi

Whose presence fills my life with joy

To my siblings

For their endless laughs and tears

ACKNOWLEDGEMENT

In the name of Allah, the Most Gracious, Most Merciful. Praise be to Allah S.W.T, Peace and blessings of Allah be upon His Messenger, Muhammad S.A.W, and all his family and companions.

I would like to express my gratitude to Prof. Dr. Ahmad Termizi Ramli, Dr. David Andrew Bradley and Prof. Dr. Husin Wagiran for their supervision, support, and encouragement throughout the course of these studies. I am sincerely grateful to the government of Malaysia and to Universiti Teknologi Malaysia for a funded PhD scholarship. I am also indebted to the Academy of Sciences Malaysia, Ministry of Science, Technology and Innovation of Malaysia and Ministry of Higher Education of Malaysia for providing research grants for this project. I am also grateful to the management and staff of the Surrey Ion Beam Centre for making available beamtime to carry out a large part of the studies featured herein. I am grateful to the Medical Physics Department of the Royal Surrey County Hospital for allowing access to the megavoltage X-ray machines situated at the hospital, in particular to Prof. Dr. Andrew Nisbet and Russel Thomas for helping out with irradiation studies. I would like to acknowledge the support of technical staff at the Physics Department, University of Surrey and Universiti Teknologi Malaysia and Ibnu Sina Institute, in particular, James Wallbank, John William Brown, Saiful, Nazri, Wani and Haliza for their assistance and cooperation.

I am similarly grateful to the Nuclear Agency Malaysia and the Department of Oncology and Radiotherapy, Hospital Sultan Ismail for giving an outstanding help and guidance in the early stage of this project, in particular, Dr. Zaharuddin, Mr. Hadi, Mr. Taiman, Mr. Hassan, Miss Nora and Mr. Izwan.

ABSTRACT

This work concerns the suitability of doped SiO_2 fibres as ionizing radiation dosimeters. The physical characters of the amorphous medium are discussed, as is the origin of the thermoluminescence (TL) signal and desirable characteristics of such a dosimeter. Facilities supporting characterization of the fibres are outlined, including an ion beam facility used for Particle Induced X-ray Emission and Rutherford Back Scattering analysis used to localize and determine the concentration of Ge and Al dopants. The dosimetric capabilities of Ge-, Al-, O_2-doped and pure SiO_2 optical fibres were investigated for low-energy X-rays, megavoltage photons, β-rays, accelerated electrons and accelerated protons, α-particles and fast neutrons. For Ge- and Al-doped fibres, linearity of dose responses were observed over useful radiotherapeutic dose range for 6 MV photons and 6 to 12 MeV electrons. TLD-100 provides a TL yield about 10 times that of Ge-doped fibre and about 30 times that of Al-doped fibres. The same order of sensitivity is displayed using a β-ray source. Linear dose response was also observed for 2.5 MeV protons irradiation. For α-particles the Bragg peak was localised to 4.5 cm in air from the point of emission. Strong TL response to fast neutrons was also found for Ge-doped fibres but was negligble for Al-doped fibres. These findings are supported by Monte Carlo simulations. Z_{eff} of between 11.9-13.4 and 11.7-13.7 were found for Ge- and Al-doped fibres respectively. The minimum detectable dose for Ge-, Al-doped fibres and TLD-100 chips were observed to be in the range of 30-50μGy, 800-1400μGy and 3-5 μGy respectively, for 6 MV photons, and 6-, 9- and 12 MeV electron irradiation. The peak of the glow curve is between 210^{o}C to 240^{o}C; the broad glow curve is characteristic of amorphous media. With oxygen forming additional defect centres in fibres, ion-implantation was used to dope pure silica with O_2 defects, implanting to the depth of 160 nm. The results show promising sensitivity on first use, although subsequent annealing leads to the loss of practically all of the dopants.

ABSTRAK

Kajian ini berkaitan dengan kesesuaian serabut SiO_2 terdop sebagai dosimeter sinaran mengion. Ciri-ciri fizikal media amorfus dibincangkan begitu juga asal usul isyarat luminesens terma dan sifat-sifat yang diperlukan pada dosimeter berkenaan. Kemudahan yang digunakan untuk mencirikan serabut dibentangkan termasuk kemudahan alur ion yang digunakan untuk teknik Pemancaran Sinar-X Aruhan Zarah (PIXE) dan analisis Serakan Balik Rutherford (RBS) untuk menentukan kedudukan dan menentukan kepekatan dopan germanium (Ge) dan aluminium (Al). Kemampuan dosimetri serabut optik SiO_2 yang didop dengan Ge, Al dan O_2 telah diselidik terhadap sinar-X tenaga rendah, foton (Megavoltan), sinar-β, elektron dan proton yang dipecutkan, zarah-α dan neutron cepat. Bagi serabut yang didop dengan Ge dan Al, sambutan luminesens terma adalah linear dengan dos radioterapeutik untuk julat tenaga foton 6 MV dan elektron 6-12 MeV dicerap. TLD-100 menunjukkan hasil luminesens terma 10 kali daripada serabut terdop Ge dan 30 kali daripada serabut terdop Al. Tertib kepekaan yang sama ditunjukkan oleh punca sinar-β. Sambutan dos yang linear juga dicerap untuk penyinaran proton 2.5 MeV. Untuk sinar-α, didapati puncak lengkung Bragg berada pada jarak 4.5 cm di udara dari titik pemancaran. Sambutan luminesens terma yang tinggi terhadap neutron cepat diperolehi untuk serabut terdop Ge tetapi boleh diabaikan untuk serabut terdop Al. Dapatan ini disokong oleh simulasi Monte Carlo. Nilai Z_{eff} (nombor atom berkesan) adalah masing-masing dalam julat 11.9-13.4 dan 11.7-13.7 bagi serabut terdop Ge dan Al. Dos minimum yang dapat dikesan untuk penyinaran 6 MV foton, electron 6-, 9- dan 12 MeV bagi serabut terdop Ge, Al dan cip TLD-100 adalah masing-masing dalam julat 30-50 μGy, 800-1400 μGy dan 3-5 μGy. Puncak lengkung berbara berada di antara 210^{o}C ke 240^{o}C, iaitu lengkung berbara lebar yang menjadi ciri bagi media amorfus. Oksigen membentuk pusat kecacatan tambahan dalam serabut, oleh itu teknik hunjaman ion telah digunakan untuk mendop silika tulen dengan cacat O_2 pada kedalaman 160 nm. Hasilnya menjanjikan kepekaan yang baik untuk penggunaan kali pertama. Namun, proses sepuhlindap seterusnya telah menyingkirkan hampir keseluruhan dopan.

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LIST OF ABBREVIATIONS

NAME	DEFINITION
TLD	Thermoluminescence dosimetry
ICRU	International Commission of Radiation Units
TL	Thermoluminescence
TLD phosphors	TLD-100 or TLD-700
STE	Self-trapped exciton
DFT	Density functional theory
ODMC	Optically detected magnetic resonance
LET	Linear energy transfer
IR	Infra-red
PMT	Photomultiplier Tube
MCVD	Modified Chemical Vapour Deposition
PTFE	Polytetrafluoroethylene
Si(Li)	Lithium drifted silicon detectors
LINAC	Linear accelerator
PIXE	Proton-induced x-ray emission
RBS	Rutherford back scattering
SEM	Scanning electron microscope
EDXRF	Energy dispersive x-ray spectroscopy
OSL	Optically stimulated luminescence
MCNP	Monte Carlo N-Particle

LIST OF SYMBOLS

ΔE	Energy of the photoelectron
E	Incident photon energy
E_o	Binding energy of the orbital electron
τ	Photoelectric mass attenuation coefficient
Z	Atomic number of the atom
θ	Small scattering angle
E_γ	Energy of the incident photon
$E_{\gamma'}$	Energy of the scattered photon
$m_o c^2$	Rest-mass energy of electron (0.511 MeV)
E_{max}	The maximum energy
m	The charged particle mass
S	The linear stopping power
dE	The differential energy loss
dx	The corresponding path length
m_o	The electron rest-mass
e	The electronic charge
v	Velocity
ze	Charge of the primary particle
I	The average excitation
N	The number density
c	Velocity of light
D	Absorbed dose
Gy	Gray
LiF	Lithium fluoride
$CaSO_4$	Calcium sulphate

E_t	A trap of depth
k	Boltzmann constant
T	Temperature
n	Number of electrons in a particular trap energy
B_{mean}	The mean TL background signal
σ	The standard deviation
F	TL system calibration factor
D_o	Threshold dose
C	Coulomb
N_2	Nitrogen gas
SiO_2	Silicon dioxide
GeO_2	Germanium dioxide
Al_2O_3	Aluminium oxide
MU	Monitor Units
Z_{eff}	The effective atomic number

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CHAPTER 1

INTRODUCTION

1.1 Overview

Thermoluminescence dosimetry (TLD) is generally acknowledged to be the most widely used and cost-effective technique for radiation dosimetry, being almost certainly the most popular technique for routine monitoring of occupational radiation exposure (Portal, 1981). TLD phosphors are widely applied in medicine to determine patient dose arising from diagnostic X-ray procedures and cancer radiotherapy treatments. The dose ranges of interest are approximately 0.01 - 1 mSv for personal dosimetry, 0.1 - 100 mSv for clinical X-ray diagnosis and 1 - 5 Sv for radiotherapy (ICRU, 1998). Rassiah *et al.* (2004) reported that there are currently 17 megavoltage radiotherapy centres to serve a population of 23.3 million in Malaysia. This large number of therapy centres has paved the way for medical therapy researchers to investigate issue of consistency on dose delivery to the patients. The International Commission of Radiation Units (ICRU) recommends that the accuracy of the dose delivered to the target volumes in radiotherapy should be within ± 5% (ICRU, 1976).

TLD phosphors most typically used in medical applications are LiF:Mg,Ti and LiF:Mg,Cu,P due to their tissue equivalence characteristics. However, these well established materials have several notable drawbacks, including being hygroscopic and having relatively poor spatial resolution, ~ up to a few mm (McKeever and Moscovitch, 2003). With these restrictions in mind, novel TLD materials are

currently identified, based on doped SiO_2 optical fibres, which offer characteristics that provide good potential for broadening the applicability of TLD.

Compared with the use of TLD phosphors, the fibres not only offer the possibility of improved positional sensitivity (fibre diameters are sub-millimeter, typically ~ 200 μm) but also, since the fibres are impervious to water (the silicates forming a glass in the fibre-preforming process)**,** this paves the way for their use in intercavitary and interstitial measurements (Abdulla *et al.*, 2001b). Such capability, in conjunction with the appreciable flexibility of the fibres (accommodating relatively small radii of curvature) also makes a fibre dosimetry system suitable for studies related to intra-coronary artery brachytherapy. Application of radiation doses to artery walls to prevent re-stenosis following balloon angioplasty is a technique that has recently gained widespread attention within the radiotherapy community. The capability of fibre TL systems also promises to be of considerable interest for dosimetry in a variety of vascular procedures involving high radiation doses to the skin, such as procedures being carried out under fluoroscopic guidance. In patients with severe medical problems for whom few, or no, alternative diagnostic techniques can be envisaged, doses have been delivered to an extent resulting in severe skin necroses (Aznar *et al.*, 2002). Geise and O'Dea (1999), cite doses of several tens of Gy to the skin in several such medical investigations, also reviewing moves towards ensuring skin dose reduction in so far as this may be possible for the particular situation. At this point it is sufficient to point out that there are many interesting dosimetric applications of optical fibres that could be imagined in such situations.

For optical communication, dopants within glass fibres are used to control the refractive index of the glassy host (Bhadra *et al.*, 2005). Fortuitously, due to the presence of these same dopants, irradiated glass fibres give rise to TL to an extent well beyond that of the silica itself. Useful intensities of TL have been observed at radiation levels familiar in high dose radiation-medicine procedures. In optical fibre systems, dopants added to the core structure provide for total internal reflection, these

defects being distributed under the high temperature and rapid quenching conditions that are required for fibre drawing.

1.2 Thermoluminescence phenomena

The presence of defects is the centre of the thermoluminescence phenomena. As an example, a negative ion vacancy is a region of excess positive charge and as such may be regarded as a potential electron trap (Furetta, 2003).

In a perfect crystal, there exists a bandgap between the highest valence band and the lowest conduction band where no electron or hole allowed states are possible. Whenever the perfect crystal structure is disturbed by the presence of a defect, one or more additional energy levels are introduced into the forbidden gap. The energy level for an electron trap can be located just below the conduction band. Likewise, a hole trap can produce an energy level just above the valence band. However, unlike the perfect crystal energy bands, which extend throughout the crystal, the additional levels are localized at the crystal defect. Figure 1.1 shows a simple representational view of energy levels in a solid containing defects.

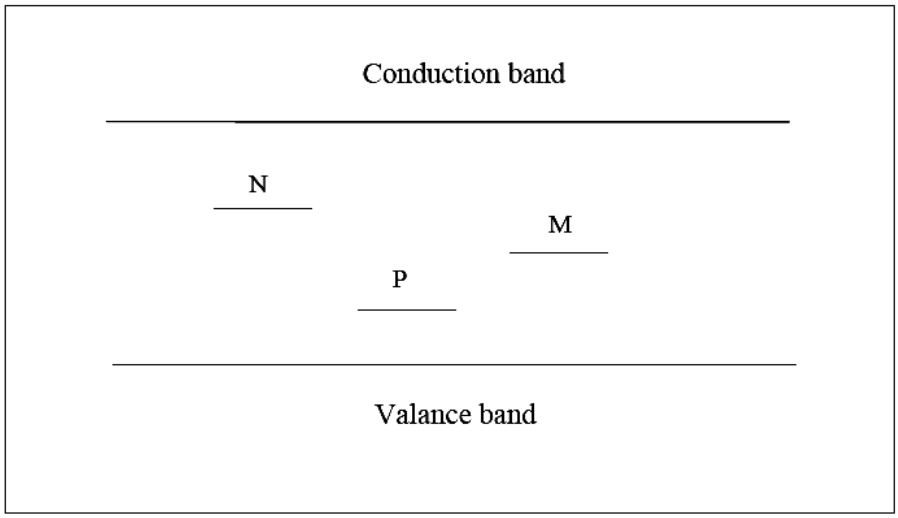

Figure 1.1: A simplified energy level diagram for the thermoluminescence phenomena. Here, N is an electron trap, P is the hole trap, and M is the recombination centre (Yusoff, 2005a).

In its ground state, the valence band is filled while the traps and the conduction band are empty. When ionizing radiation is absorbed by the material, some electrons in the valence band will be excited to the conduction band. The excited electrons are now free to move through the crystal. The presence of defects in the crystal makes it possible however, for the electrons to become trapped. Holes, which are created in the valence band during the excitation process, can also move freely in the crystal until either they are trapped in hole traps or are recombined with electrons.

The trapped charge carriers will remain in their respective traps unless sufficient energy is acquired to allow them to escape. In thermoluminescence, the two main factors that determine the extent to which the trap emptying process occurs are the depth of the trap and the local temperature around the trap. While the trap depth is determined by intrinsic properties of the defect, the temperature is an externally controllable factor (Yusoff, 2005a). If the temperature is raised, the trapped charge carriers may acquire enough energy to escape. Subsequently, the

released electrons may recombine with holes at the recombination centres with the emission of photons of commensurate energy (wavelength).

During heating, a released charge can participate in many different kinds of process. The three main kinds of processes of importance to the thermoluminescence phenomena are retrapping of charge carriers at a defect, radiationless recombination, and luminescent recombination. Only the luminescent recombination process produces a signal which is useful for thermoluminescence production (Yusoff, 2005a). Further explanation related to TL phenomena will be discussed in section 2.4.

1.3 The structures of silica (amorphous SiO_2)

The structure of silica was reported by Bakos (2003). Silica is known to be an amorphous (or glassy) material. X-ray diffraction indicates that the structural order present in crystalline forms is preserved in such glassy media over the short and intermediate range scales. The atomic coordination and the first and second neighbour distances are very similar in the amorphous and crystalline forms indicating that the basic building blocks of the two solid states of SiO_2 are similar. The structural phase diagram of crystalline SiO_2 is shown in Figure 1.2. The most common structures of SiO_2 at low pressure are α-quartz, β-quartz, and tridymite.

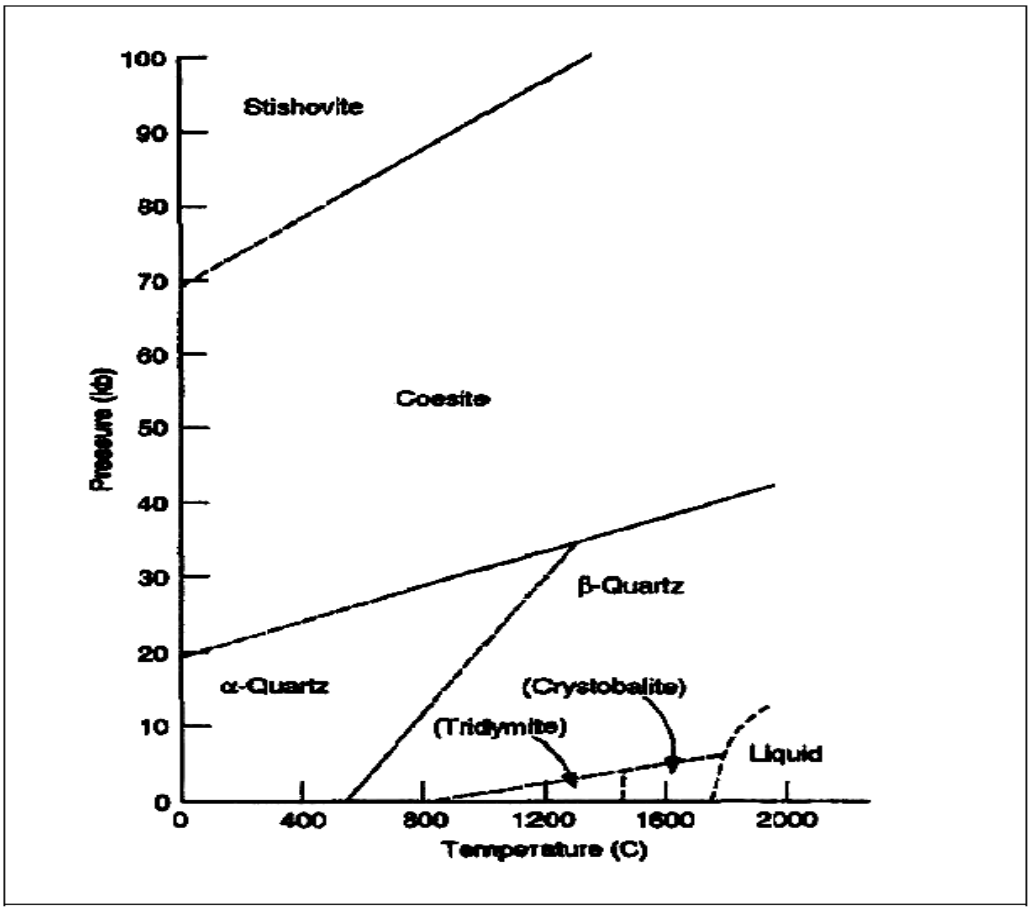

Figure 1.2: SiO_2 structural phase diagram (Bakos, 2003).

The basic bonding unit of all SiO_2 crystalline structures, except for stishovite, is a tetrahedron as shown in Figure 1.3. Each silicon atom is bonded to four oxygen atoms, with the bond lengths ranging from 1.52Å to 1.69Å. The O-Si-O bond angle is 109.18°. Each of the oxygen atoms bonds to two silicon atoms with the Si-O-Si bond angle ranging from 120° to 180°, depending on the structure. The high temperature low pressure crystabolite and tridymite possess the largest bond angles. The large bond angles reflect in the density of the material, with the larger bond angle producing less packed SiO_2 material.

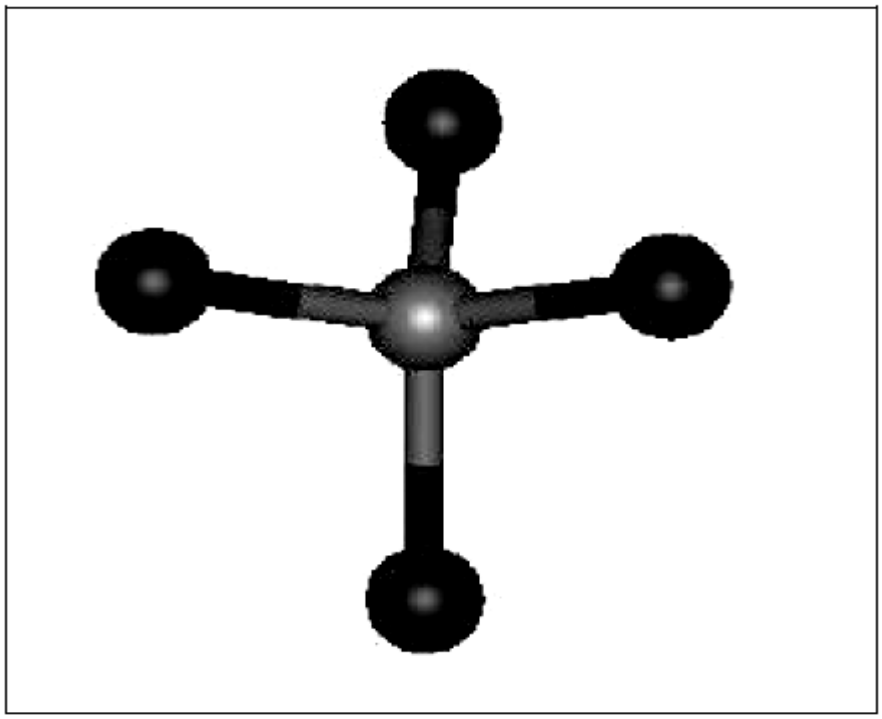

Figure 1.3: SiO_4 tetrahedral coordination is the most common structural unit for SiO_2 (Bakos, 2003).

Although the phase diagram in Figure 1.2 was derived from the case of crystalline SiO_2, it can also be used to determine which structure might be present in an amorphous sample, in the form of micro crystallites. Since silica glass is a result of fast cooling of heated SiO_2, the material is not allowed to relax to the low temperature structure. The most probable form of SiO_2 micro crystallite in silica glass produced at ambient pressure is tridymite, being stable up to 1470°C.

Two models exist for the structure of amorphous silica, namely, the continuous random network model and the microcrystalline model, both of which are consistent with experimental data. In the continuous random network model, the silica glass is modelled to be constructed from SiO_2 tetrahedrons with each of the oxygen atoms shared by two tetrahedrons. The only difference to the crystalline forms is the bond angle, which is allowed to vary from one tetrahedron corner to another. Therefore, the structure is a random network of tetrahedrons. In the

microcrystalline model, the silica is constructed from micro-crystallites of different structures, as in Figure 1.2 or from the sub units of the structures. By doing this, the distribution of bond angles falls within the types of structure included in the model. Within the limit of small crystallites, the two models converge (Karmakar, 2005).

1.4 Thermoluminescence mechanism for SiO_2-based glass

In this section, is a review of existing research into possible mechanisms for the luminescence phenomena in silica glass. Although almost all of this research concerns investigation of the photoluminescence phenomena in crystalline silica, the defects studied can also be considered to provide a link to the thermoluminescence in silica glass. While there are many possible defects in silica, discussion will only be made of those defects that have been studied in some detail, generally being those which have a greater probability of causing significant thermoluminescence. The defect modes can be divided into two major categories, according to whether one is dealing with intrinsic or extrinsic defects.

1.4.1 Intrinsic defects

It is meaningless to define defects in silica in the same way that they would be for a crystalline material, i.e. any disruption in the repeated structure is considered a defect, since amorphous silica does not enjoy long range order. Defects in silica are therefore defined as the disruption in the short or mid range order, in this case the disruption occurring in the normal SiO_2 tetrahedron coordination and the normal two fold coordination of the oxygen atoms. Therefore, the absence of oxygen atoms in the tetrahedrons are oxygen vacancy defects while the absence of silicon atoms are silicon vacancy defects.

One common type of defect in silica is the broken or dangling bond. This type of defect leaves atoms with dangling orbitals that are populated by unpaired electrons. Thus, this type of defect is paramagnetic and therefore is detectable using the electron spin resonance (ESR) technique. The main defects that fall into this category are the non-bridging-oxygen (NBO) centre ($O_3{\equiv}Si\text{-}O\cdot$), the proxy radical, and variations of E′ centre, namely the oxygen vacancy.

Diamagnetic defects (an example of which is given below) are not detectable by ESR. In this case, the defects are studied by mapping out their energy levels using absorption, luminescence, or photoionization spectroscopy. One important diamagnetic defect in silica is the neutral oxygen vacancy where the Si-Si bond is formed, being usually a precursor to one of the E′ centre type of defect.

One important feature of the intrinsic defect is that the number of these is temperature dependent. For silica, the two main defects within this category are the oxygen vacancy centre and the self-trapped exciton, a defect resulting from the interaction between an excited electron with the corresponding hole left in the valence band, to be explained further in section 1.4.1.2.

1.4.1.1 The oxygen vacancy centre

Perfect crystalline silica will comprise of single silicon atoms surrounded by four oxygen atoms. Each of these oxygen atoms forms a bridge between two silicon atoms. In this way charge neutrality is conserved within the crystal. If one oxygen atom is missing, the local charge becomes positive. As such, it becomes a possible electron trap. In silica, this trap is called the E′ centre, being analogous to the well known F centre in alkali halides. Variations of the E′ centre, i.e. an oxygen vacancy results in dangling silicon sp^3 bonds as summarized by Warren *et al.* (1992).

An analysis of the E′ centre by Silsbee (1961) has determined the existence of an unpaired spin localized at the sp^3 orbital of the silicon atom with orbital orientation towards the oxygen vacancy position. According to Fowler and Edwards (1997), if the two adjacent silicon atoms relax asymmetrically, this being the most likely occurrence in glass due to its inherit asymmetry, then one electron can stabilize itself on one of the silicon atoms. Electron spin resonance (ESR) studies in silica glass by Griscom (1979) have revealed that the same E′ structure exists in silica glass.

With an oxygen vacancy, it is also possible for the atom to rearrange itself to form a Si-Si bond, which also maintains local charge neutrality. This happens when both silicon atoms adjacent to the oxygen vacancy relax towards each other. Tsai *et al.* (1988) have suggested that this arrangement is a result of non-radiative decay of an exciton (see below for an explanation of the exciton), the Si-Si centre forming a hole trap. They have estimated that the exciton binding energy in SiO_2 is about 1.3 eV. The E′ centre can also transform into a Si-Si centre at high annealing temperature. At even higher annealing temperature, the Si-Si centres tend to transform themselves into excess silicon atoms in SiO_2 (Rebohle *et al.*, 1998).

1.4.1.2 The self-trapped exciton

When an electron is excited in a lattice it will leave a hole in the valence band. This electron will experience a coulombic interaction with the hole, screened by ions and other electrons. This electron-hole pair, termed an exciton, can propagate through the crystal. In SiO_2, the electron-hole pair is strongly bound due to the low dielectric constant of the base compound. The presence of the self-trapped exciton (STE) in SiO_2 leads to an energy level inside of the normal bandgap.

The existence of a photoluminescence band at 440 nm in SiO_2 supports the suggestion that excitons are trapped in the SiO_2 matrix, a phenomenon which will not happen in an ideal crystal. It is now accepted that the mechanism for STE in silica is the motion of oxygen atoms in the crystal. The motion, first proposed by Fisher *et al.* (1990), is an oxygen atom rotation about another Si-O bond of a neighbouring silicon atom (Figure 1.4). This motion causes a lattice distortion that significantly changes the local energy level in the crystal.

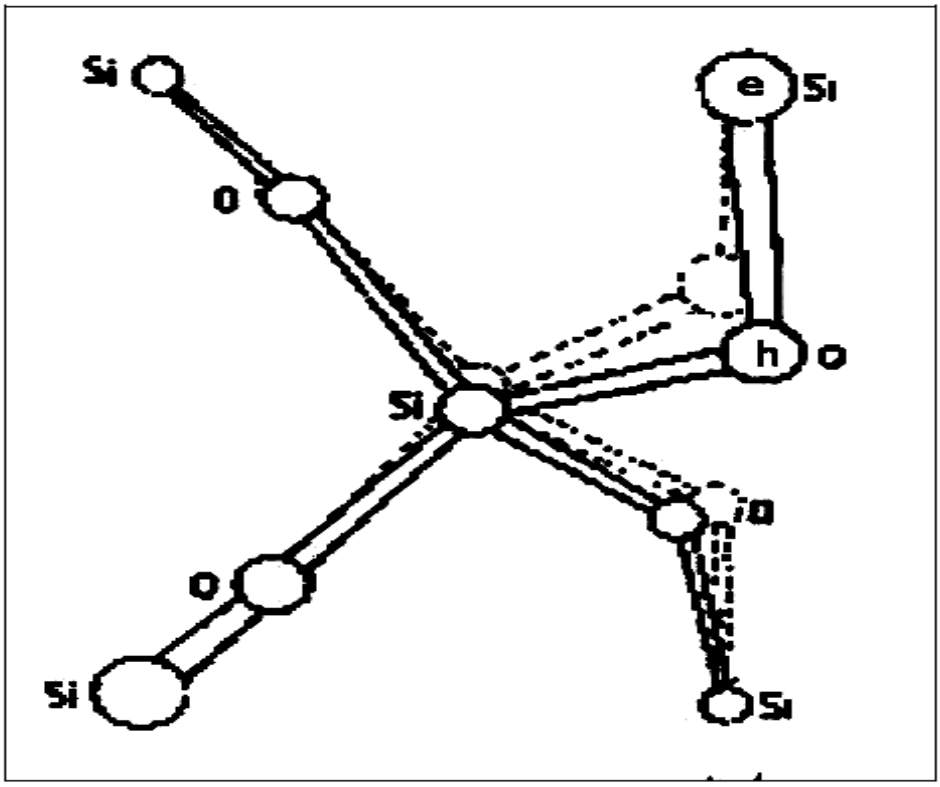

Figure 1.4: Oxygen distorted states proposed by Fisher *et al.* (1990). Dashed lines represent configuration for the case of perfect crystal geometry. The symbols e and h represent localization of the electron and hole respectively, for the trapped exciton.

A Hartree-Fock calculation on this oxygen distorted state, using the model depicted in Figure 1.4, confirmed the view that oxygen motion is the mechanism responsible for exciton trapping (Bakos, 2003). A later, more rigorous calculation using density functional theory (DFT) shows that the excited electron is localized on the antibonding molecular orbital of a silicon atom. Conversely, the hole is localized

on the molecular orbital of the three oxygen atoms. In addition to the oxygen distorted state at 2.8 eV, a silicon distorted state at 0.23 eV higher has also been found by Song *et al.* (2000).

1.4.2 Extrinsic defects or impurity centres

Analysis of luminescence when impurities are introduced into the SiO_2 substrate involves a model in which the impurity centre becomes either substitutional atoms, interstitial atoms, an impurity-intrinsic complex, or an impurity-impurity complex inside the substrate. Apart from the possibility of introducing a new luminescence band, impurity atoms might also change the number of electron or hole traps. In the following, a brief discussion is provided of a number of elemental impurity atoms, some of which have been used in commercially available telecommunication optical fibres (providing favourable optical refractive indices for total internal-reflection). Present interest is of course the thermoluminescence produced by such dopants.

1.4.2.1 Silicon rich silica

The silicon rich silica contains more oxygen defect centres than in pure silica. Accordingly, although enjoying the same spectral energy distribution, one expects more intense luminescence to result from the rich silicon silica as compared to pure silica.

1.4.2.2 Germanium impurity

Germanium atoms in SiO_2 will directly substitute for silicon atoms in the crystal. Spin resonance studies have shown that the GeO_4 unit cell and SiO_4 unit cell are similar, before and after trapping of an electron. The difference in the ionic potential of Si^{2+} and Ge^{2+} are too small to provide any impurity localized states, as reported by Hagon *et al.* (1985). However, Ge in SiO_2 is known to be a deep electron trap. An optically detected magnetic resonance (ODMC) study by Hayes and Jenkin (1988) has shown that Ge doped silica exhibits similar exciton occurrence to that existing in pure silica.

1.4.2.3 Aluminium impurity

Aluminium doped silica yields a luminescence spectra that shows the existence of an E' centre type defect (Trukhin *et al.*, 2004). In this particular case, the aluminium atom in SiO_2 is surrounded by three oxygen atoms, $AlO_{3/2}$. This arrangement is diamagnetic and also electrically neutral. At temperatures greater than 260K, the oxygen vacancy traps electrons, and the local charge around aluminium becomes negative, i.e. $AlO_{3/2^-}$.

An aluminium impurity in SiO_2 can also become a hole trap. Some aluminium atoms in silica are four-fold oxygen coordinated. For this type of coordination, the local charge is negative, i.e., $AlO_{4/2^-}$. Near room temperature and above, $AlO_{4/2^-}$ is not compensated and therefore it can trap a hole. At ground state, the trapped hole is localized on an oxygen long bond to aluminium. For excited states, the hole is localized on the short oxygen bond to aluminium (Nuttal and Weil, (1981a) and Nuttal and Weil, (1981b)). The same hole trap centre also exists in Ge doped SiO_2, occurring when a Ge atom traps an electron to become Ge^{3+}. However, this centre is not deep enough to survive at room temperature (Hayes and Jenkin, 1988).

1.5 Thermoluminescence studies on optical fibres

Early studies involving photon irradiation effects of silica based optical fibres were carried out by, among others, Friebele (1979), Friebele *et al.* (1980, 1984), Kirsh *et al.* (1989), Ellis *et al.* (1989), Khanlary *et al.* (1993) and Abdulla *et al.* (2001a). These investigations have determined that the TL performance of an irradiated optical fibre is influenced by the type of fibre and by the radiation parameters. Presented herein are a few of the studies that suggest development of TL dosimeter materials from doped silica fibres.

1.5.1 Germanium doped optical fibre

Abdulla *et al.* (2001b) has carried out a TL study on commercially available Ge-doped silica based optical fibres. The fibre was prepared in the form of 1 cm length rods (~ 0.3 mg each), and the samples were irradiated using a gamma source. The radiation dose range investigated was 1-1230 Gy. The TL glow curve parameters are summarized in Table 1.1.

Table 1.1: Values for activation energy *E*, and frequency factors *s*, for Ge-doped optical fibres obtained by Abdulla *et al.* (2001b).

Peak	**Temperature (°C)**	***E* (eV)**	***s* (s^{-1})**
1	100	0.80	1.20×10^{12}
2	140	0.99	2.30×10^{13}
3	325	1.42	1.22×10^{13}
4	399	1.56	5.48×10^{12}

Good reproducibility of TL readout through five repeat cycle of annealing-irradiation-readout was found (correlation coefficient = 0.95). In addition, the dose response for the fibre was found to be linear from 1 to 120 Gy. The TL signal from Ge doped fibre has a fast fading characteristic of about 2% within 6 hours

and a slow fading of 7% within 30 days. It was claimed that the lower detectable limit of radiation dose for this type of optical fibre is 0.02 Gy (Abdulla, 2003).

1.5.2 Erbium doped optical fibre

Work on this dopant also has been carried out by Abdulla *et al.* (2001c) using commercially available Er doped silica based optical fibre. The main feature of the TL glow curve is that it produces a single glow peak at about 150^{o}C. The activation energy (*E)* and the frequency factor (*s)* were calculated using four methods: the peak shape method, Grossweriner, Lushchik and the initial rise method. The first three methods gives value of 0.3 eV to 0.4 eV and the last method gives a value of 0.67 eV. The dose range investigated was 2-400 Gy, the dose response being found to be linear up to about 250 Gy. Significant fading of the TL signal from Er doped fibre has been found, nearly 30% of TL signal being lost after the first 24 hours. After 20 days of storage at room temperature, a total loss of 58.6% of TL signal was reported.

1.5.3 Neodymium doped optical fibre

Safitri *et al.* (2006) has observed that the Nd-doped silica fibres provide high dose response subjected to photon and electron irradiations. The Nd-doped fibres within the energy of 0.06 - 20 MeV has an effective atomic number of about 13.32 – 21.41. The TL glow curve shows a broad peak characteristic at ~ 192 $^{\circ}$C. A complete annealing process was found at 400 $^{\circ}$C for one hour. The TL fading up to 12.5% over 30 days was recorded following 202.2 mGy X-rays irradiation.

1.6 Statements of hypotheses

The hypotheses used are as follows:

1. The optical fibres investigated are typical single-mode fibres. The irradiations on the core of the optical fibre have been conducted at various doses for different types of ionizing radiation sources (photons, electrons and protons which are accelerated and neutrons, alpha and beta particles).

2. It is known that the pure silica will give rise to a degree of thermoluminescence (TL) following irradiation by ionizing radiation, the TL signal is considerably enhanced by the presence of certain dopants.

3. The exact amount of dopant added to these fibres is not specified by the manufacturers. The dopant acts as the defect centres that provide the TL signal. Of possible concern is the non-uniformities in the distribution of added dopants and impurity concentration in the core of the optical fibres which may contribute to variation in TL yield.

1.7 Objectives of the study

The objectives of this study are:

1. To provide fundamental dosimetric properties of doped SiO_2 optical fibres i.e. effective atomic number, linearity and sensitivity with respect to dose response, TL glow curve, fading, residual studies and reproducibility.

2. To understand the TL dependency with dopant and dopant concentration, so that the sensitivity of the measurement is acceptably high and the dose response is linear within the dose range of interest for medical applications.

1.8 Scope of the thesis

In regard to this doctoral thesis, an important characteristic of dosimeters such as fading, TL glow curve, residual signal studies, reproducibility and sensitivity characteristic of these fibres will be explored, as well as their dose dependency using photon, neutrons and charged particle ionizing radiations.

The methods of analyzing the TL glow curves will be described theoretically in Chapter 2. No glow curve analysis will be conducted to obtain the values of kinetics parameters (activation energy *E* and frequency factor *s*). The study on TL dosimetric properties such as the annealing condition, glow curves parameters, energy dependence, relative energy response, dose rate effect, heating rate effect and optical bleaching have also been omitted. These have been studied in detail by Abdulla, Y.A (2003) and Safitri *et al.* (2006). However, their studies only used accelerated photons and electrons from linear accelerator (linac). It is important to try different approach with the aim to understand the dependency of TL signal on variety of dopants, dopant concentrations and different types of radiation sources. This study may provide a basis for exploiting TL phenomena for various dosimetric situations.

Although producing optical fibre dosimeters appears to be of great interest, the scientific problem involved in producing such a dosimeter lies within the development of suitable silica based material that is sufficiently sensitive to radiation dose. In the studies presented herein, results from research on variously doped SiO_2 (Ge-, Al-, O_2-doped and pure silica optical fibres) will be compared against commercially available TLD phosphors (TLD-100 and TLD-700). Findings from these studies may pave the way to conduct more comprehensive investigation of TL from tailor-made doped SiO_2 optical fibres.

The present chapter, has provided introduction to the problems associated with TL and a review of the existing literature regarding the subject. Chapter 2 addresses the TL models, radiation interactions, principle of TLD and their important characteristics as a radiation dosimeter and brief introduction to the technique related in this study. Chapter 3 describes the methodology and equipment used. In chapter 4, a range of thermoluminescence studies and the results obtained are presented and discussed in detail. Chapter 5 summarizes the findings of this investigation, and provides an outlook for future study in this area.

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