

## A STUDY OF THE EFFECT OF IONIZING AND NON IONIZING RADIATIONS ON CELLULOSE TRIACETATE POLYMERS

A Dissertation Submitted to the Faculty of Science Physics Department



In Candidacy for the Degree of Ph.D.

By

## SAMIR AHMED EL-SAYED NOUH

B.Sc., Physics, Ain Shams University, 1986 M.Sc., Physics, Ain Shams University, 1991 M. El - Snany M. S. Aldel-Wald

91643

### CONTENTS

		Page
ACKNOWL	EDGEMENT	
SUMMARY		i
CHAPTER	I: INTRODUCTION	1
CHAPTER	II: THEORITICAL ASPECTS AND PHYSICAL PROPERTIES	
	OF POLYMER	12
	II.1 Effect of ionizing radiations on polymers	12
	II.2 Changes in physical properties of polymers upon irradiation	17 17 19 21 23 27
	II.3 Effect of non ionizing radiations on polymers	28
CHAPTER	III: EXPERIMENTAL TECHNIQUE	32
	III.1 Samples	32
	III.2 Irradiation facilities	33
	III.3 Mechanical measurements	34
	III.4 Viscosity measurements	36
	<pre>III.5 X-ray diffraction measurements</pre>	39
	III.6 Refractive index measurements	4.0



III.7 Infrared measurements	41
III.8 Absorption and transmission measurements	41
CHAPTER IV: THE EFFECT OF GAMMA IONIZING RADIATION ON	
THE MECHANICAL, STRUCTURAL AND OPTICAL PRO-	
PERTIES OF CELLULOSE TRIACETATE POLYMER	45
<pre>IV.1 Statistical aspects in radioactivity     measurements</pre>	45 45 46
IV.2 Mechanical properties	47
<pre>IV.3 Intrinsic and kinematic viscosities- dose relations</pre>	51
<pre>IV.4 X-ray diffraction</pre>	55
IV.5 Optical properties	57 57 59
IV.5.2 Infrared spectra for cellulose triacetate polymer	61
IV.5.3 Absorbance & transmission dose- relations	62
<pre>IV.5.4 Colour changes in irradiated</pre>	63
CHAPTER V: THE EFFECT OF INFRARED PULSATED LASER ON THE	
MECHANICAL, STRUCTURAL AND OPTICAL PROPERTIES	
OF CELLULOSE TRIACETATE POLYMER	66
V.1 Mechanical properties	66
V.2 Viscosity-laser dose relations	69

V.3 X-ray diffraction	72
V.4 Optical properties	74
samples	74
V.4.1.B Refractive index of solid sampls V.4.2.A Absorbance and transmission of	75
liquid samples	76
solid samples	77
V.4.3 Colour changes in exposed cellu- lose triacetate polymer	79
V.5 Post gamma irradiation	81
CONCLUSIONS	84
REFERENCES	87

ARABIC SUMMARY

#### ACKNOWLEDGEMENT

The author would like to thank Prof. Dr. M.A. EL-SHARKAWI, the head of Physics Department, Faculty of Science, Ain Shams University.

The author is pleased to express his deep appreciation to Prof. Dr. M.A. EL-FIKI, National Institute for Standards and Prof. Dr. M.S. ABDEL-WAHAB, Physics Department, Faculty of Science, Ain Shams University for proposing and planning this investigation and their continuous encouragements, valuable suggestions, capable supervision and reading throughout the manuscript have rendered the realization of this work to be possible.

Deep thanks to Prof. Dr. S.U. EL-KAMEESY, Physics Department, Ain Shams University, for his faithful help.

Many thanks are indebted to Prof. Dr. H.M. EASA, National Institute for Standards and to Dr. S.A. EL-FIKI, Physics Department, Ain Shams University, for their faithful help and encouragement.

I wish to express may thanks to Prof. Dr. T. EL-DESOKI, Physics Department, Ain Shams University and Prof. Dr. M.M. RADWAN, Physics Department, Military Technical College and to Dr. N.M. EL-ENANY, Physics Department, Ain Shams University, for their beneficial help during the experimental part of the research program.

Also deep thanks to Dr. G.ABDEL HAMID, Armed Forces Technical Research Center for the providing of necessary facilities.

The author is most obliged to his colleagues of the radiation physics group of the physics department, Faculty of Science, Ain Shams University, for their cooperation. The staff members of the Armed Forces Technical Research Center and of the National Institute for Standards are greatly acknowledged for their beneficial help during the experimental part of the research program.

Finally, the author is indebted to the director and members of the Scientific Computing Center, Ain Shams University, for their beneficial help during the representation of the data.

#### SUMMARY

In this work comparative studies on the effect of ionizing radiations (gamma) and non ionizing radiations (laser beam) on cellulose triacetate (C.T.A.) polymer have been carried out. Strips of C.T.A. of thickness 0.2 and 0.25 mm were used. Many parameters were checked, such as fracture stress and strain, elasticity, yield stress, intrinsic viscosity, crystallinity, refractive index, absorbance and transmission, tristimulus values, and chromaticity coordinates.

It was found that at relatively high gamma doses ranging (14 - 32 KGy) i.e.  $(7-16 \text{ J/cm}^2)$  the young's modulus decreases with increasing the gamma dose. Also it showed a decrease with laser doses greater than 3 J/cm<sup>2</sup>.

It was also shown that higher gamma doses ranging (14 - 32 KGy) i.e.  $(7-16 \text{ J/cm}^2)$  and higher laser doses ranging (3-9  $\text{J/cm}^2$ ) increases the yield stress.

The intrinsic viscosity showed an increase at some gamma doses ranging (0-1.3 KGy), (8 -13 KGy) and (15-32 KGy) i.e. (0-0.65  $J/cm^2$ ), (4-6.5  $J/cm^2$ ) and (7.5-16  $J/cm^2$ ). While it showed a decrease at the other dose ranges. Also the

relation between the intrinsic viscosity and the laser dose showed a maximum around  $3 \text{ J/cm}^2$ .

The dependance of intrinsic viscocity on temperature indicated that the temperature coefficients of C.T.A. are negative and much greater in magnitude. Also, the C.T.A. displays coefficients which are the same within experimental error.

The X-ray diffraction measurements showed that the integral intensity decreases at higher gamma doses (15 - 32 KGy) i.e.  $(7.5 - 16 \text{ J/cm}^2)$  indicating that the samples tend to be amorphous at this dose range. Also the half height width showed an increase at this dose range indicating a decrease in the crystallite size and the tendency towards the amorphous state. Also, the X-ray diffraction study showed that the integral intensity increases with increasing the laser dose indicating that the samples tend to be crystalline when exposed to laser pulses. Besides, the half height width showed a decrease with increasing the laser dose above 4.5 and up to 9 J/cm<sup>2</sup> indicating an increase in the crystallite size and the formation of grains and grain boundaries.

The refractive index measurements showed that the refractive index increases with increasing dosage of gamma

up to 5 KGy (2.5  $J/cm^2$ ) and then decreases with increasing the gamma dose up to 32 KGy (16  $J/cm^2$ ). Also, the relation between the refractive index and the laser dose showed a maximum around 3  $J/cm^2$  indicating that laser doses above 3  $J/cm^2$  enhances the anisotropic character of C.T.A polymer.

The infrared spectra for cellulose triacetate showed that  $\log (I_O/I)$  at the maximum of the absorption band was not affected by the environment of irradiation.

The ultraviolet % transmission spectra appeared for all samples as a band with different intensities and the ultraviolet % transmission measurements showed that the absorbance of the samples decreases at higher gamma doses (14-32 KGy) i.e.  $(7-16 \text{ J/cm}^2)$ .

The variation of the absorbance with laser dose showed that at lower wavelengths the induced changes in the samples are relatively high, while at higher wavelengths the induced changes in the exposed samples are smaller. This indicated that the absorbance of the samples was proved to be wavelength dependent.

The colour difference measurements showed that irradiating the samples to gamma doses or to laser pulses causes variations in the tristimulus values and the chromaticity coordinates indicating the existence of a colour difference between irradiated samples and the non irradiated one.

Exposure to laser pulses after gamma irradiation lead to an increase in the tristimulus values X, Y, Z and a decrease in both the refractive index and the % transmission values.

# CHAPTER I

**INTRODUCTION** 

#### INTRODUCTION

Deep chemical and physical changes occur in polymers under the action of ionizing radiation, regardless of their kind (X-rays - gamma rays - fast and slow neutrons, fast electrons, alpha particles, protons and other products of nuclear reactions.). The energy of these radiations ranges from 9 to 10 ev., whereas the energy of the chemical bonds in polymers is of the order of 2.5 - 4 ev.(1,2). Such radiations can therefore break bonds in a chain, but this does not always occur because of the redistribution and dissipation of energy(1).

Ionizing radiation directly produce ionized and excited molecules and electrons. Some excited molecules may be deexcited through the emission of radiation or through non radiative transitions. Excitation energy can also be transfered from one molecule to another. Electrons are trapped at various sites, or can combine with molecules to form negative ions, or recombine with positive ions yielding excited molecules. Both ions and excited molecules may acquire considerable vibrational energy and undergo bond rupture to form a complex array of stable molecules, free radicals, ionized molecules and radical ions. High energy

radiations cause degradation, crosslinking of polymers, an increase in the unsaturation of the molecular chains and breaking up of the crystalline structures.

Irradiation causes a polymer molecule to be ionized and excited as follows:

Where P, P+ and P\* are polymer molecule, ionized molecule and excited molecule, respectively.

In fact, radiation conversions in Polymers are caused by the energy effectively transmitted to the mass of the material<sup>(2)</sup>, which takes place during the process of consecutive interactions for the incoming particles with the electrons and atoms of the medium. The results of these reactions are ionization, excitation of the polymer and the formation of high energy secondary charged particles. These changes depend on chemical composition, general morphology and free energy state of the polymer.

The effect of main chain scission can be followed by measurements of viscosity<sup>(3,4)</sup> or molecular weight and also by a reduction of the mechanical strength. In fact, this may be used as a method of measuring chain entanglements.

If a polymer is irradiated below its glass transition temperature, the life time of the free radicals is prolonged because of the low mobility of the polymers structural elements<sup>(1)</sup>. At a definite stage of irradiation, the polymer molecules are chemically bonded (crosslinked) into a common structure and loses its ability to dissolve in its customary solvents.

Numerous publications dealt with the effect of gamma radiation on polymers and fibres. Chapiro (1962)<sup>(5)</sup> reported that most polymers acquire a colour under irradiatin but the range of doses in which discoloration becomes noticeable varies widely depending on the chemical structure of the polymer. The chemical changes in irradiated polymers include crosslinking, chain scission and recombination of broken chains (Parkinson, 1969). In some polymers noticeable changes in colour are caused by very small radiation doses which can be used for dosimetry purposes.

Hamza et al., (1989b)<sup>(6)</sup> studied the change in colour of two polymer fibres by gamma irradiation. Keller (1982)<sup>(7)</sup> gave a survey of a series of studies on the influence of crystallinity on the radiation induced effects, crosslinking in particular, in polyethylene and paraffins. Lawandy (1982)<sup>(8)</sup> have shown that colour changes in a polymer can be