A review on aging or degradation of polymer optical fibers (POFs): polymer chemistry and mathematical approach

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Abstract

We review the chemistry of POFs degradation or aging which has been done so far to understand the transmission loss in climatically stressed fibers. Mathematical models developed so far for light propagation and influence of aging process have also been reviewed.

Introduction

Short haul optical data communication through polymer optical fibers (POFs) has got much importance over silica optical fibers because of two main reasons; they are low cost of production and have high mechanical properties. But still these fibers have a main disadvantage of high transmission loss (or attenuation) due to absorption and scattering [1, 2, 3].

Since a decade investigations have been made to reduce intrinsic loss in POFs successfully by replacing H atoms in the core polymer molecules by

- halogen atoms (F, Cl and Br),
- deuterium atoms(d),
- blending core polymer with dopants (halogen containing compounds) and
- by using co-polymerization technique [4,5,6,7].

But main disadvantages of these techniques are hazardous nature of monomers and high cost of production.

However, PMMA based POFs are widely being used in optical signal processing and many other optical applications under different environmental stress factors [8]. Some investigations have shown that degradation of macromolecules proceed via different fashions, formation of new fragments and change in their molecular network structure, when they are exposed to environmental stress factors for long time [9, 31]. Thus, in POFs transmission loss arises with different reasons.

In this article, we report a general review on three degradation processes namely

- high temperature,
- humidity, and
- mechanical degradation.

Existing mathematical models of light propagation and possible ways of incorporating aging parameters are also reviewed.

Loss factors in POF

Loss factors in a typical commercially available POF can be shown as below [9].

Loss Factors in POF			
Intrinsic loss factors		Extrinsic loss factors	
Absorption	Scattering	Absorption	Scattering
C-H overtones, Electronic transitions	Rayleigh scattering (variation in refractive index(*n*))	Transition metals, organic contaminants	Core-clad imperfection, Dust&micro-voids, Others

Intrinsic loss factors (Absorption)

A major loss contribution is from intrinsic loss factor and it is heavily due to C-H absorption overtone and partly due to Rayleigh scattering in the visible region of the spectrum. Theoretically it has been calculated that contribution of C-H absorption overtone and Rayleigh scattering loss is around 100dB/km and 90dB/km at 650nm respectively [1]. Absorption overtones appear in the lower region of the spectrum from the position of fundamental vibrations and these vibrations are anharmonic in nature. Due to the large anharmonicity constant of C-H molecular vibrations, these have been considered as anharmonic oscillators and their potential curves are approximated by a well known 'Morse Potential', given by

$$V(r) = D_e\left[1 - e^{(r-r_e)/\rho}\right]^2$$

where r_e is the equilibrium bond distance, D_e is the potential energy for bond formation and ρ a parameter controlling the width of the potential well.

The solution (Schrödinger equation) of this approximation is

$$G(v) = v_0\left(v + \frac{1}{2}\right) - v_0 x\left(v + \frac{1}{2}\right)^2$$

where $v = 0,1,2,.....$, x = anharmonicity constant, $v_0 = (2\pi c)\,(k/\mu)^{1/2}$, k is force constant, μ is reduced mass and c is velocity of light.

From the above equation it is possible to calculate potential well and the corresponding electronic transitions. With the help of infrared spectroscopy, it is also possible to calculate the overtone band positions as well as their intensities if one knows the position of fundamental vibrations and anharmonicity constant [10, 11].

On the other hand these vibrational overtones can be almost eliminated by replacing H atoms by heavier atoms (halogenation and deuteration) such as F, Cl and Br, and d. These substituted atoms molecular vibrations have a smaller anharmonicity constant as a result of very low absorption overtone loss.

Since a decade rigorous research has been made on this topic. As a result few low loss POFs have been fabricated for e.g. ARTON, ESKA, CYTOP, LUCINA etc., and exclusive work has been done by Koike et al. [12] and Kaino et al. [13]. Groh et al. [10, 11] have been extensively worked on the calculation of the position of overtone bands as well as of their intensities, which appear for several fundamental vibrations of different molecules in various core polymers.

Intrinsic loss factors (Scattering)

Rayleigh scattering is mainly due to two factors. They are minute irregularities (dust, bubble and cracks) and fluctuations in composition (density and refractive index) within the core fiber. Since core polymers are highly amorphous in nature, they can be considered as super cooled liquids. The turbidity (τ), which is the total scattering intensity with wavelength in all the angles, is frequently used as term to express the scattering in liquids [1]. It may be calculated from the equation given below.

$$\tau = \frac{8\pi^3}{3\lambda^4}\left[\rho\left(\frac{\delta\varepsilon}{\delta\rho}\right)_T\right]^2 kT_f\,\beta_T$$

Where T is the temperature, T_f is the fictive temperature, β_T is isothermal compressibility, k is Boltzmann constant, ρ is density, ε is dielectric constant and

$$\rho\left(\frac{\delta\varepsilon}{\delta\rho}\right)_T = \frac{(n^2-1)(n^2+2)}{3}$$

where n is refractive index of a polymer system.

Scattering also depends on the molecular structure and physical geometry of the molecules, for example scattering loss in

PMMA (fig.1a) (*n*=1.490) based POF is less compared to polystyrene (PS) (*n* = 1.590) or polycarbonate (PC) (*n* = 1.586) based POF. It may be explained briefly in this way: PS (fig.1b) molecules have benzene rings in their structure, this induces raise in RI and thus increases the turbidity of the whole polymer system.

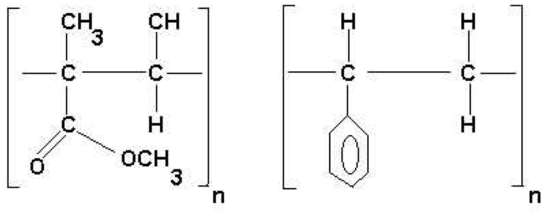
(a) (b)

Figure 1: The molecular structures of (a) PMMA and (b) PS

A detailed explanation on Rayleigh scattering in PMMA-POFs can be found in [1, 14].

Extrinsic loss factors

Extrinsic loss factor influence is very small but it cannot be neglected if one wants to achieve high optical data transmission through POFs. It could be still reduced by using well-purified monomers and having a perfectly cleaned fiber-processing laboratory.

To date, with all the molecular structural manipulations in the core polymer, attenuation has been reduced to 20dB/km or less [12].

Polymer chemistry

High temperature degradation of PMMA based POF

Temperature is one of the important parameters that considerably increases the attenuation of POF when it is used for long time. PMMA has a glass transition temperature (Tg) of about 105 ^{0}C. PS and PC have Tg of about 100 ^{0}C and 145 ^{0}C respectively. In several papers various reasons have been reported for considerable increase in attenuation when the POFs are exposed to different higher temperatures (<Tg and >Tg) for long time [9,15,16,17].

Takezawa et al. [9] have made an actual chemical analysis of high temperature (150 ^{0}C) effects on PMMA, PS and PC based POFs and found the reasons for high attenuation in the formation of carbon-carbon double bond (>C=C<) and conjugated carbonyl (>C=O) compounds. These compounds have high absorption in the visible region due to $\pi - \pi^*$ and n - π^* electronic transition respectively. They have confirmed these experimental results (formed new fragments) with the model compounds of similar structure prepared in their laboratory. But in this investigation, obviously no monomers were formed as one of the usual cases in classical high temperature oxidative degradation of PMMA [16].

Recently, Schartel et al. [17] have made a kinetic study on thermo-oxidative degradation of PMMA-POFs using chemiluminescence technique. Random chain scission reactions were observed and the degradation reaction seems to be a mixture of a first and zero order reaction. An attempt has also been made to observe the water absorption effects in the same fibers. They found decrease in chemiluminescence intensity and an increase in activation energy. However, all these investigations are still lacking in stating the actual chemical reasons for increase in attenuation.

In actual classical thermal degradation process of PMMA, three steps are involved viz., initiation, depropagation and termination reactions as in a radical polymerization reaction [16]. Initiation process in thermal degradation occurs in different modes but in pure PMMA (POF grade) it may be due to rupture of double bonds (so called weak bonds) in the main chain because generally there are no other ingredients in it. Depropagation is in favor of monomer formation as the temperature increases. Termination occurs via coupling process.

However, in high temperature degradation of PMMA-POFs, an increase in attenuation could be primarily due to formation of monomeric and dimeric compounds, which have high absorption in the UV-visible region of the spectrum (called as yellowing of color).

Water absorption or humidity effects in PMMA based POFs

Water absorption in POFs provokes increase in intrinsic loss due to appearance of O-H molecular vibrational absorption overtones in the visible region of the spectrum [18]. Loss arises not only by absorption but also by little increased scattering due to aggregation of water molecules in the core fiber [6].

However, PMMA is an hygroscopic compound which absorbs water of about 1.5%. Water absorption mechanism in PMMA have been speculated and investigated by several researchers. It occurs through a dual mechanism i.e. part of water molecules enter into the polymer network structure and induces the swelling of the structure, and part of it goes into the pre-existing microvoids [19].

Experimentally it has been shown that water absorption in PMMA can be expressed at equilibrium as

$$W_D = f \cdot W$$

where W_D indicates the weight per cent of water which is dissolved in the polymer and W is the total water percentage present in the polymer including the amount of water gone into microvoids. f represents partitioning of the water between the two sorption modes and is a constant at a given temperature [20].

Increase in attenuation due to water absorption has been experimentally found also in deuterium and halogenated POFs, particularly high loss has been noticed in deuterated POFs [10, 18]. One of the primary reasons for high loss in d-POFs could be due to low dissociation energy of C-D bonds.

Results of previous investigations have shown that temperature in combination with humidity influences the water sorption in PMMA based POFs and hence increases attenuation in a short period of exposure [18, 21]. It could be due to slight movements of polymer chains from the freezed structure which occurs during fiber processing that easily allows water molecules to move into the network structure and then cause swelling and dissolution.

This study on POFs allows us to characterize the existence of microvoids and swelling behavior, which are on the other hand strong evidence for Rayleigh scattering and mechanical degradation studies.

Mechanical degradation of POFs

Increase of attenuation in POFs when they are mechanically constrained for long time, is due to formation of new fragments by breaking off polymer chains (bond rupture)[22]. It can also be due to change in fundamental properties of polymer by natural physical aging process due to amorphous nature of core polymer. So, it can be studied in two ways viz., mechanical degradation phenomenon or classical physical aging of glassy polymers [23].

Dugas et al. [24] have speculated that increase of attenuation in POFs is due to micro cracks development in the core fiber region which are formed during fiber drawing process and are increased in their size by stress relaxation process. Also, they have stated that molecular chain orientation takes place towards the direction of fiber axis during fiber drawing process and provokes it less entanglements in the radial direction. Finally, they have concluded that PMMA as a core fiber is not fully amorphous and has weak mechanical resistance in the radial direction. It may be true because polymers undergo some percentage of crystallization due to conformational changes (rotation around the single bond) in their structure when they sufficiently get stretched to a particular direction from their melt.

Pierrejean et al. [25] have drawn a similar conclusion from their dilatometric studies. Due to chain orientation during fiber drawing process, formation of structural inhomogenities in PMMA core fiber provoke increase in attenuation (due to scattering).

Daum et al. [21, 26] have carried out mechanical (repeated bending) reliability tests on some POFs and found increase in attenuation due to reduction in core diameter (may be called it as necking) and fiber fracture by promotion of necking due to microcracks. However, in these and their subsequent investigations they still lack to produce strong evidence for the existence of microcracks as well as for the chemical reasons.

However, mechanical degradation process involves morphological changes as well as radical reactions, which are similar to thermally initiated or photo-initiated radical reactions. Mechanism of mechanical degradation process in many semicrystalline polymers has been successfully explained using Peterlin's model by many researchers [22, 27]. It is also called microfibril model. A schematic representation of bond rupture in the amorphous regions in semicrystalline polymers can be shown as in figure 2.

A brief explanation of this mechanism is: when the mechanical stress is applied to a semicrystalline polymer specimen and if stress imposed is gradually increased, initially main chain bonds are ruptured especially in the amorphous regions connecting to crystalline regions by tie molecules and shortest tie molecules are ruptured primarily.

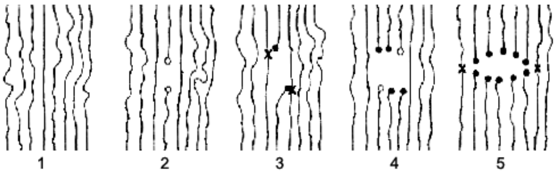

Figure 2: Schematic representation of bond rupture in the semicrystalline polymers. 1. Highly stressed chain, which rupture 2. Formation of chain ends radicals, 3. Radical reaction leading to main chain radicals, 4&5. Scission of radicalized chain and repetition of the former steps leading of the formation of a sub microcrack; o-chain end radicals, x is main chain radical, • stable end groups. [21, 27]

This model would be very useful for explaining the mechanical degradation process in POFs, too.

On the other hand, since core polymers of POFs (e.g. PMMA) are highly amorphous in nature, they have been considered as glassy polymers. According to classical physical aging phenomenon, glassy polymers (e.g. PMMA, PC, PS, etc.) undergo thermodynamic equilibrium state from non equilibrium state which is acquired when it is cooled from above Tg to a temperature T (< Tg).

Free volume change (volume available between the polymer chains) has been commonly used to characterize this behavior in glassy polymers [28]. Viscoelastic properties namely stress relaxation and creep in bulk PMMA samples have been studied by many researchers and extensive work done by Struik on different glassy polymers [23]. WLF equation (called as shift factor) has been used to generate master curve for many short-term stress-strain curves to predict long-term behavior in glassy polymers under different stress factors [29].

It has also been proved that when a solid glassy polymer undergoes physical aging under a considerable stress for long time, chemical bonds are ruptured and thereby new fragments formed due to which its mechanical strength decreases [30]. Hence, physical aging also involves the degradation of macromolecules with the structural changes.

There are only few publications available on POFs durability tests using classical physical aging idea but detail study is still lacking in those investigations [26, 31].

Mathematical approach

Existing fiber models

There are two general approaches to mathematical modeling of a multimode fiber:

- Wave optics: Models describe fiber characteristics in terms of mode-specific properties and energy transfer between modes.
- Geometric optics: Models are based on a concept of ray and often include raytracing.

Wave-optics models

The basis for wave fiber analysis is provided by solutions of Maxwell's equations, which have been obtained for a cylindrical dielectric waveguide. The exact solutions for homogenous core waveguide (i.e. step-index fiber) were obtained by Snitzer and yield the full set of *TE*, *TM*, *EH* and *HE* modes (corresponding to transverse electric, magnetic and hybrid field configurations) with different propagation characteristics [32]. However, Gloge has shown that in the typical case of small relative index difference, propagating modes may be approximately given in terms of only two linearly polarized components, *LP* modes [33].

In gradient-index case approximate solutions of Maxwell's equations are usually obtained by WKB method [34]. By its means it is also possible to accurately estimate potential bandwidth dependence on fiber index profile, as Ishigure and Koike showed [35].

Fiber imperfections and Rayleigh scattering are the main reasons for the process of energy transfer between modes [38]. Mode coupling effects are both desirable as they equate energy transfer times and thus reduce pulse dispersion and undesirable as they increase attenuation by coupling energy to higher-order unguided modes [36,37].

Geometric optics models

Mechanism of propagation in multimode fibers may be well understood by means of geometric optics only [39]. Thus, it is especially suitable for POFs, as they are large in diameter and highly multimode fibers.

Bunge et al. [40] report simulations of different types of POFs (e.g. fibers with different NA and diameter). Despite of simple assumptions (e.g. uniform mode distribution and meridional rays only), they have obtained realistic attenuation and bandwidth results.

Arrúe reports advanced mathematical formalism of raytracing through straight and bent (step-index and gradient-index) fibers [41].

Gloge [38,42] and Olshansky [44] model mode coupling by an energy diffusion process between groups of rays with different angles θ to the fiber axis (or between so called θ-modes, see [43]).

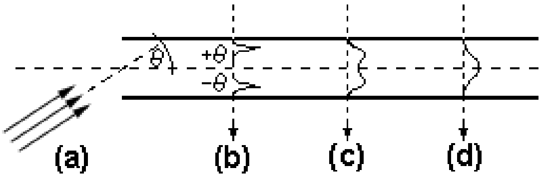

Figure 3: Pulse angular power distribution at various points along the fiber: (a) only modes with characteristic angle θ are excited; (b) near-field diagram, two sharp intensity peaks at angles of $+\theta$ and $-\theta$; (c) two peaks diffuse and begin to overlap; (d) far-field diagram, peaks are undistinguishable.

Let $P(z,\theta,t)$ be energy in group of modes with characteristic angle θ at fiber length z at time t. Gloge equates the change in energy due to space and time (left hand side) to the change due to attenuation and diffusion (right hand side). Hence, pulse propagation may be described by a partial differential equation

$$\frac{\partial P}{\partial z}+\tau(\theta)\frac{\partial P}{\partial t}=-\alpha(\theta)P+\frac{1}{\theta}\frac{\partial}{\partial \theta}\left[\theta d(\theta)\frac{\partial P}{\partial \theta}\right]$$

where τ is the transfer group delay time, α and d are attenuation and mode coupling intensity coefficient respectively. Pulse angular broadening due to diffusion is schematically shown on fig. 3.

Aging influence

There are few interconnected, aging-affected local fiber characteristics that strongly influence transmission properties. They should be taken into account in fiber aging mathematical models.

- Local energy absorption resulting in overall fiber attenuation.
- Local scattering resulting in mode mixing and leading to a steady-state energy distribution on longer fiber lengths.
- Fiber defects (e.g. microcracks).

Modeling aging

As there are no mathematical models directly dedicated to aging, it seems reasonable to modify existing models to incorporate aging effects.

Local and global mode-specific attenuation and coupling intensity seem to be parameters of wave-optics models the most affected by aging. In the case of geometric optics models, aging influence may be simulated by modifications of fiber refractive index, absorption distribution and by immersing cracks into the fiber [24]. Since analytical formulas for ray paths in non-ideal fiber are generally not available, raytracing software has to be used.

Conclusions

Loss factors have been organized in a systematic way but these are not fully explained from the chemical viewpoint in climatically (high temperature and humidity) and mechanically aged POFs. Existing mathematical models of light propagation have been reviewed. We intend to make chemical investigations on and develop detailed mathematical models for climatically and mechanically stressed POFs using spectroscopic techniques (IR, Raman, chemiluminescence) and raytracing software respectively.

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