Scattering and absorption coefficients measurements in silicone polymers used in fiber optics

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Abstract. Scattering and absorption coefficients of organosilicon polymers (polysiloxanes) used in fiber optics were investigated. For this purpose a new measurement technique based on relocatable integrating spheres was implemented. A computer mathematical model of light propagation in a scattering medium was created based on the Monte Carlo method. The experimental results were approximated using this model in order to obtain absorption and scattering characteristics of different polymer samples.

1. Introduction

Nowadays, fiber lasers are widely used in various fields of science, industry and medicine. Optical output power of modern continuous-wave single-mode fiber lasers exceeds 10 kW [1]. Some part of generated power inevitably converts into heat due to quantum defect – that is the energy difference between the pumping and lasing photons (about 10% in Yb-doped fiber laser).

High temperature of active fiber leads to degradation of laser radiation characteristics and, finally, to fiber destruction. According to modern researches, one of the main limiting factors of further growth of fiber lasers output power is the degradation of its protective polymer cladding [2,3]. It was proved that polymer cladding is capable to absorb some part of scattered pumping, photoluminescence and generation radiation, becoming a secondary heat source in the fiber [4]. In commercial lasers, the active fiber is usually placed into metal unit and is additionally filled by a thick layer of organosilicon polymer for laser stability and better heat sink. But it is necessary to take into account that radiation absorption in polymer can lead to additional fiber laser heating and, subsequently, to faster degradation of the polymer layer. Therefore, the study of optical properties of polymers used in fiber optics is an actual problem.

Organosilicon elastomers (polysiloxanes) are usually used as the fiber coating materials due to the good combination of its mechanical, thermal, and optical properties. The degradation temperature of these polymers lies in the range of 80 °C - 170 °C [3]. Earlier in [4], some transmission spectra measurements of different types of polymers in visual and near infrared range (400nm - 2500nm) were performed. Numerous absorption peaks in the spectral regions corresponding to the operating wavelength ranges of Yb, Er and Tm-doped fiber lasers were found. These absorption peaks are generally associated with the vibration overtones of the methyl CH₃ group in the polymer.
Also in [4], absorption coefficients in organosilicon polymers at different wavelengths of laser radiation were measured by means of laser calorimetry method: the absorption coefficient of polymer sample was determined from the dependence of polymer heating on transmitting laser radiation power. The advantage of this method was its accuracy in determining absorption coefficient in comparison with spectral measurements. However, this method did not allow to determine scattering coefficient of polymer, which is necessary to know, since the fraction of scattered radiation in some polymers can be an orders of magnitude higher than the fraction of absorbed radiation.

It's also important to mention that for a number of fiber optics applications polymers with added metal powder are used. This powder increases thermal conductivity of such type of a polymer and, consequently, enhances heat dissipation. In addition, it leads to a significant increase in scattering coefficient of optical radiation, which makes it possible to achieve uniform radiation scattering in polymer medium and, therefore, uniform heat load distribution in it. However, such polymers have several disadvantages, including an increased absorption coefficient of laser radiation.

In this paper, we present a novel method for measuring the scattering and absorption characteristics of optical radiation in polysiloxane polymers used in fiber laser optics.

2. Measurement of optical characteristics of polymer samples

2.1. Experimental setup

The method based on the use of relocatable integrating spheres was recently proposed to determine optical properties of various highly scattering biological tissues [5]. In our paper, we suggested to use this technique for measuring scattering and absorption coefficients in polymers. The block-scheme of the experimental setup is shown in Fig. 1.

![Figure 1. Block-scheme of the experimental setup for measuring the scattering and absorption characteristics of polymers.](image)

The probe source of continuous wave radiation at 1060 nm wavelength was an Yb-doped laser with output power of 4 W. To prevent thermal effects in the polymer sample, laser radiation was attenuated using a wedge-shaped mirror. Laser beam diameter was 1.3 mm and could be varied using diaphragms. Average power of radiation passing through the samples was about 100 mW. Optical chopper was used for mechanical modulation of laser radiation with frequency about 300 Hz in order to implement lock-in amplification technique.
The $S_T$ and $S_R$ integrating spheres (indices "T" and "R" corresponds to transmittance and reflectance, respectively) and polymer sample in fused silica cuvette were fixed on steel rails, so the sample was located between the spheres. Distances $L_T$ and $L_R$ between the sample and the $S_T$ outlet and $S_R$ inlet could be varied. Input hole diameters of the spheres were 7 mm and their centers were located along the direction of laser beam propagation. Averaged over the spheres scattered radiation powers were measured using $D_T$ and $D_R$ photodetectors with built-in amplifiers. Electric signals from these detectors were processed by the lock-in amplifier. Reference signal from the chopper was used for phase synchronization.

The dependences $P_T(L_T)$ and $P_R(L_R)$ of optical radiation power captured by the $S_T$ and $S_R$ spheres were measured experimentally. The signals obtained in these experiments were normalized to the value of laser radiation power passing through the sample. Experimental measurements were carried out at room temperature.

The novelty of the proposed technique in comparison with the classical method based on integrating spheres lies in the fact that the sample was moved along vertical axis during measurements. It was possible to obtain the dependence of the optical power captured by the integrating spheres on the distances $L_T$ and $L_R$ for more accurate determination of investigated parameters.

2.2. Mathematical modeling

The propagation of light in scattering media can be described in terms of the theory of radiation transfer, the basic equation of which is:

$$\frac{1}{c} \frac{\partial I(\bar{r}, \bar{s}, t)}{\partial t} = -\nabla I(\bar{r}, \bar{s}, t) - \mu_t I(\bar{r}, \bar{s}, t) + \int_{4\pi} p(\bar{s}', \bar{s}) I(\bar{r}, \bar{s}', t) d\omega' + \varepsilon(\bar{r}, \bar{s}, t)$$

(1)

where $I$ is the radiation brightness at the point with a radius vector $\bar{r}$, $\mu_t$ is the attenuation coefficient, i.e. the sum of scattering $\mu_s$ and absorption $\mu_a$ coefficients, $p$ - phase scattering function, $\varepsilon$ – function, describing radiation source, $\bar{s}$ – the direction of radiation propagation, $d\omega'$ – the solid angle with the normal vector $\bar{s}'$.

Determination of the phase scattering function $p$ can be made by means of goniometric measurements. However, for many scattering media the phase function is well described by the Henyey-Greenstein function (2):

$$p(\theta) = \frac{1}{2} \frac{1 - g^2}{(1 + g^2 - 2g\cos\theta)^{\frac{3}{2}}}$$

(2)

Where $g$ is the scattering anisotropy coefficient which has the meaning of the average cosine of the angle $\theta$ between radiation propagation direction before and after scattering (3):

$$g = \frac{\int_0^{2\pi} \cos\theta p(\theta) d\theta}{\int_0^{2\pi} p(\theta) d\theta}$$

(3)

The problem of light propagation in polymer sample was solved using Monte Carlo method based on step-by-step simulation of photons propagation. At each step, the photon flux moved a certain discrete distance inversely proportional to the attenuation coefficient $\mu_t$, while their number was decreased in proportion to the absorption coefficient $\mu_a$. During each step photons were scattered by a random angle $\theta$ in accordance with the Henyey-Greenstein phase function (2).

The code used in this work was written and verified in accordance with that described in [6].
Knowing the values of the scattering ($\mu_s$), absorption ($\mu_a$) and anisotropy ($g$) coefficients, the created model makes it possible to determine the value of the optical power captured by the integrating spheres depending on the distance to it and the diameter of its inlet. Conversely, in order to determine these coefficients from experimental data the inverse problem was solved.

3. Experimental results
We have investigated two silicone polymer samples: a relatively transparent pure polydimethylsiloxane (PDMS) polymer and the same class polymer with addition of 3%wt. of aluminum powder. These polymers are commonly used in fiber optics as a coating for active fibers and fillers for fiber blocks or fiber splices, respectively. Liquid polymers were placed into quartz silica cuvettes and then thermally polymerized. For the accuracy of measurements, it was important to prevent the appearance of air microbubbles and inhomogeneities at the quartz-polymer boundaries. The thickness of the samples was chosen in accordance with the expected optical characteristics (10 mm for the transparent polymer and 1 mm for the polymer with metal powder). Measured dependences of the transmitted radiation power on the distance to the sample ($L_T$, $L_R$) are shown in Fig. 2.

Experimental data were approximated by varying scattering, absorption, and scattering anisotropy coefficients in the created mathematical model. The difference between the experimental data and the simulation results at small distances between the sphere and the sample (Fig. 2b) can be caused by several factors: an increase in the sphere gain when the input hole is blocked by sample, and the dependence of the sphere response on the aperture of input radiation.

![Figure 2. Dependence of radiation fraction that passed through (a) transparent polymer and (b) polymer with the addition of aluminum powder on distance to the integrating sphere.](image)

It was found that addition of aluminum powder led to an increase in absorption coefficient $\mu_a$ by 3 times (from $3 \cdot 10^{-2}$ to $9 \cdot 10^{-2}$ cm$^{-1}$) and scattering coefficient $\mu_s$ by more than three orders of magnitude (from $1 \cdot 10^{-2}$ to 41 cm$^{-1}$). The value of absorption coefficient of pure PDMS polymer was $3 \cdot 10^{-2}$ cm$^{-1}$ which is consistent with the data obtained in previous work by means of laser calorimetry [4]. The scattering anisotropy coefficient $g$ was 0.98 for the transparent polymer and 0.72 for the polymer with metal powder. But it turned out that the value of backscattered radiation for investigated samples was less than the measurement error, so we do not present it here. All experimental results are shown in the Table 1.
Table 1. Experimental results of absorption, scattering and anisotropy coefficients measurements.

| Sample                        | Thickness, mm | $\mu_a$, $10^{-2}$ cm$^{-1}$ | $\mu_s$, $10^{-2}$ cm$^{-1}$ | g  |
|-------------------------------|---------------|-------------------------------|-------------------------------|----|
| Transparent polymer           | 10            | 3±1                           | $\leq$1                       | 0.96|
| Opaque polymer with Al powder | 1             | 9±3                           | 4100±100                      | 0.72|

4. Conclusions

Thus, the proposed technique was successfully implemented to determine absorption, scattering and scattering anisotropy coefficients in transparent and turbid organosilicon polymers used in fiber optics as fiber coating and laser unit filler. It was found that addition of aluminum powder leads to the threefold increase in radiation absorption, and to an increase in scattering by more than three orders of magnitude. Measured value of absorption coefficients in polymer was in a good agreement with the data obtained by means of laser calorimetry technique.

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