MIGRATION AND SEPARATION OF PHOTO-ABSORBING MICRO-PARTICLES USING LASER-PHOTOPHORESIS IN AQUEOUS SOLUTION

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ABSTRACT

The laser photophoretic migration behavior of photo-absorbing polystyrene micro-particles suspended in water was examined by irradiating a cw Nd:YAG laser (532 nm). The photophoretic velocities of the particles were compared with those calculated by using a Mie scattering model. The observed photophoretic velocity increases dramatically with increasing the absorption coefficient of the particle. The velocity of the highly photo-absorbing particle is much larger than that calculated due to the temperature rise of surrounding medium by photothermal effect. The obtained results are important for better understanding of laser-photophoresis that can be applied to the separation of colored micro-particles in a liquid medium.

KEYWORDS: Laser-Photophoresis, Particle Separation, Photothermal Effect

INTRODUCTION

Gradient force of laser beam has been widely studied and utilized for trapping or manipulation of micrometer-sized particles or biological cells in liquid media. On the other hand, scattering force of laser is less developed as a force for the migration or the manipulation of microparticles. We have proposed a laser-photophoresis of particles[1], which is a unique technique to migrate and to characterize particles in liquid and developed fundamental studies on the photophoresis of various transparent particles in liquid media. For non-photo-absorbing particles, it has been elucidated that the laser photophoretic velocity is governed by both the refractive index and the radius of the particle, and is predictable by using Mie scattering theory[2]. However, in the case of photo-absorbing particles, anomalous photophoretic behaviors, such as inverse photophoresis[3] and expansion-contraction motion in organic droplet[4], have been observed. For highly absorbing particles, the photophoretic velocity is much higher than that of transparent particle[5], but the exact relation between photo-absorption and photophoretic velocity of micro-particles has not been clarified yet. In this study we elucidated the effect of photo-absorption on the laser-photophoretic migration of micro-particles in aqueous solution.

THEORY

Photophoretic force and photophoretic velocity. Assuming the Gaussian profile of the irradiated laser beam, the photophoretic force, \( F_{ph} \), which acts on a spherical particle at the center of the beam, is defined by the relationship,

\[
\frac{F_{ph}}{c_0^2} = \frac{2Pn_r^2}{c^2} Q_{ph}
\]

where \( P \) is the incident laser power, \( n \) is the refractive index of the suspending medium, \( r \) is the radius of the particle, \( c \) is the velocity of light in vacuum, \( \omega \) is the radius of the laser beam at the sample position, and \( Q_{ph} \) is a fraction of the light power utilized to exert force on the particle and is termed photophoretic efficiency. If plane waves incident on a perfectly reflecting or on a perfectly absorbing spherical particle, \( Q_{ph} \) is equal to unity.

When a small spherical particle is exerted by photophoretic force and migrates with a velocity of \( v \) in the medium, the sphere undergoes a resistant force, \( F_R \), from the medium. The resistant force for the solid spherical particle is expressed by

\[
F_R = 6\pi\eta rv
\]

where \( \eta \) is the viscosity of the medium. At a steady state, the velocity of the sphere is determined by the balance of \( F_{ph} \) and \( F_R \). Therefore, the photophoretic velocity of the particle in the medium can be written as

\[
v = \frac{nPrQ_{ph}}{3\pi\omega^2\eta}
\]

This is the general equation for the photophoretic migration of spherical solid particles in a viscous medium.

Calculation of Photophoretic Efficiency. In order to understand the photophoretic behavior of photo-absorbing microparticles, we have used Mie scattering (MS) theory to evaluate the photophoretic efficiency \( Q_{ph} \). The origin of the radiation force is considered to be the momentum transfer of the light to the particle due to the scattering and the absorption of irradiated light by the particle. In the MS theory, the incident laser beam is treated as electromagnetic wave, and the interaction between the beam and the spherical particle is evaluated by Maxwell's equations. To calculate the photophoretic force on the particle, we have used the Mie scattering calculation program "ScatLab ver. 2.1"[6]. By using the program, the angular distribution of the scattered light intensity by a spherical particle with any optical properties and radius can be calculated. The photophoretic efficiency was evaluated by summing the momentum vector of the scattered light, and by estimating the momentum change of the incident light in the forward direction due to the scattering and the absorption.
\[ Q_{\text{ph}} = Q_{\text{sca}} \left[ 1 - \frac{1}{\pi} \int_0^\pi F(\alpha)I(\alpha) \cos \alpha \, d\alpha \right] + Q_{\text{abs}} \]  

(4)

where \( Q_{\text{sca}} \) is the scattering cross-section, \( Q_{\text{abs}} \) is the absorption cross-section, \( F(\alpha) \) is the solid angular fraction for an angle \( \alpha \), and \( I(\alpha) \) is the intensity fraction of the scattered light in the direction of an angle \( \alpha \). The first term in square brackets of Eq.(4) corresponds to the momentum of the incident light and the second term (angular integral) corresponds to the momentum of the scattered light in the forward direction. All the parameters, \( Q_{\text{sca}}, Q_{\text{abs}} \) and \( I(\alpha) \) can be calculated by the Mie scattering program.

**EXPERIMENTAL**

The photophoretic behaviors of colored polystyrene particles were observed by an apparatus shown in Fig. 1. A cw Nd:YAG laser (Spectra Physics, Millenia, 532nm) irradiated the particles at a cross-flow configuration in a quartz square micro-cell of 400 \( \mu \)m \( \times \) 400 \( \mu \)m inner dimensions. The power of the laser was set at the range of 0.2 to 2.0 W and the diameter of the laser was 190 \( \mu \)m at the sample position. The photophoretic behavior of the particles in the cell was observed using a microscope with a CCD video-system. Five kinds of colored polystyrene particles (PolySciences, \( r = 3 \) \( \mu \)m) were used as samples and suspended in pure water. The number concentration of the sample solutions was about \( 2 \times 10^7 \) mL\(^{-1}\). Absorption spectra of the particles were measured by dissolving the dried particles to tetrahydrofuran and are shown in Fig. 2.

![Figure 1: Experimental setup for the observation of laser-photophoretic migration of micro-particles.](image1)

![Figure 2: Absorption spectra of colored polystyrene particles.](image2)

**RESULTS AND DISCUSSION**

Figure 3 shows the composite picture of the photophoretic migration of the red-colored polystyrene particles in the cross-flow irradiation configuration shown in Fig. 1. It is clear that the particles exert the scattering force of the laser and migrate to the laser propagation direction, \( y \), at the laser irradiation area. The photophoretic velocities in \( y \) direction, \( v_y \), of the particles were analyzed form the video images. As shown in Fig. 4, the photophoretic velocities distribution was a Gaussian shape and well agree with the laser intensity distribution in the micro-cell.

In order to evaluate the exact photophoretic migration behavior of the particles, the maximum photophoretic velocity at the center of the laser beam was obtained by a Gaussian fitting of the velocity distribution. In Fig. 5, the maximum photophoretic velocities for the five kinds of particles were plotted against the absorption coefficient at 532 nm of the particle. The solid line shown in Fig. 5 is the photophoretic velocity calculated by using the MS theory. The observed photophoretic velocity increases as the absorption coefficient of the particle increases as predicted by the theory. But the velocity of the highly photo-absorbing particle (red particle) is much higher than that predicted by the MS theory. This discrepancy is thought to be due to the photothermal effect. Because laser irradiation on the photo-absorbing particle results in temperature rise of the medium around the particle. And the temperature causes the lowering of the viscosity of the medium and this lead to the increase in photophoretic velocity as expressed in Eq. (3). In the case of the red-colored particles, the observed velocity was about 1.4 times higher than that predicted by the MS theory, and the temperature of the medium around the particle during photophoretic migration is estimated to be about 40 °C from the decrease in the viscosity.

The velocity increase by the photothermal effect enhances the photophoretic velocity dependence on the absorption coefficient of colored particles. If the wavelength of the laser is tuned to the characteristic absorption band of the target particles, we can separate the particles by difference in laser-photophoretic velocity. This fact indicates that laser-photophoresis is advantageous to the color-based separation of micro-particles in a liquid medium.
CONCLUSION

In this study, we have elucidated that the photophoretic velocity increases dramatically with increasing the absorption coefficient of the particle. The dependence of photophoretic velocity on absorption coefficient can be explained by applying the MS theory to the photophoretic efficiency calculation. In the case of highly photo-absorbing particles, photophoretic velocity is much higher than that predicted by the MS model calculation due to the lowering of the viscosity of the medium by photothermal effect. These results are important in the evaluation of the photophoretic forces that can be applied to the color-based separation of micro-particles in a liquid medium.

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REFERENCES


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