Decay time control of mass diffusion in a transient grating using a fringe-tunable electrothermal Fresnel mirror

Yuki KIUCHI*, Yoshihiro TAGUCHI** and Yuji NAGASAKA**

*School of Integrated Design Engineering, Keio University
3-14-1 Hiyoshi, Yokohama, Kanagawa 223-8522, Japan
**Department of System Design Engineering, Keio University
3-14-1 Hiyoshi, Yokohama, Kanagawa 223-8522, Japan
E-mail: tag@sd.keio.ac.jp

Received: 14 June 2017; Revised: 24 August 2017; Accepted: 18 September 2017

Abstract
This paper reports a microelectromechanical systems (MEMS) mirror with electrothermal polymer actuators for the diffusion sensor. A compact and high-speed diffusion sensor is desirable for point-of-care testing because of its real-time monitorability and portability, and because diffusion coefficient reflects the abnormality of biological samples such as proteins. Herein, a fringe-tunable electrothermal Fresnel mirror (FEFM) is analyzed to maximize the mirror’s angular shift while maintaining the repeatability of the actuator drive. The thermal-response-speed and temperature-distribution characteristics were examined. The proposed fabrication process contributed toward improving the yield and quality of the device. The diffusion coefficient was successfully measured using the fabricated FEFM. Moreover, by making the fringe spacing 7.1 times narrower than its initial value, the decay time of diffracted light became 50 times faster than that of the wider fringe, thereby showing reasonable agreement with theory. The results validated the development of a compact, high-speed diffusion sensor that realizes control of the decay time of the mass diffusion in a transient grating using an FEFM.

Keywords: Diffusion coefficient, Electrothermal actuation, Microelectromechanical systems (MEMS), Polymer actuator, Transient grating

1. Introduction
The importance of microscale devices in the healthcare field has increased owing to their advantages of small-sample-volume and high-throughput measurements of chemical and biological reactions between proteins and antibodies (Chin et al., 2012, Baret et al., 2010, and Chin et al., 2007). Li et al. developed a volumetric bar-chart chip to accurately detect the antibody–antigen reaction without any calibrations for clinical analysis such as point-of-care testing (POCT) (Li et al., 2015). Teerapanich et al. demonstrated fluorescence-microscopy monitoring of protein binding kinetic in real time with low noise for clinical diagnostic applications and drug screening (Teerapanich et al., 2017). However, these methods take a long time (several tens of minutes) or require preprocessing. Because the knowledge on diffusion coefficients is important for biological-sample analysis, Terazima et al. developed a diffusion-coefficient-sensing method that analyzes the kinetics of biomolecular interactions between proteins and DNA or between proteins and other proteins in real time using an ultra-short-pulsed laser (Terazima et al., 2011). Barrett et al. observed lipid dynamics in the presence of the amyloid-β associated with Alzheimer’s disease (Barrett et al., 2016). Torres et al. reported a phase-shifting interferometry for accurate measurement of diffusion coefficients (Torres et al., 2013). However, these state-of-the-art methods require a large sample volume and a large apparatus; thus, they are not applicable to POCT or drug-discovery applications. Although dynamic light scattering (Lehermayr et al., 2011), the Taylor-dispersion method (Chamiieh et al., 2016, Hogstedt et al., 2016, and Oukacine et al., 2015), and nuclear-magnetic-resonance spectroscopy (Latour et al., 1994) are widely used as methods for measuring the diffusion coefficient, a long measurement time is required. A transient-grating method (Terazima et al., 1993) and
fluorescence-correlation spectroscopy (Magde et al., 1972, Ruan et al., 2004, and Dertinger et al., 2007) have realized high-speed and small-sample-volume diffusion-coefficient sensing; however, these methods require surface modification or the application of additives to the sample in advance.

In this study, a compact diffusion sensor that achieves small-sample-volume and high-speed measurements without any additives and with a low driving voltage is reported. Our method observes the mass diffusion process of the interferometrically created fringe-shaped microscale concentration distribution of samples that are manipulated by a laser-induced dielectrophoretic force (LIDEP). For the miniaturization of our sensor, Oka et al. firstly developed a micro Fresnel mirror that can form a high-contrast interference fringe without any complicated imaging optics and successfully detected the mass-diffusion process of a lattice-shaped concentration distribution in the 10^8 s range (Oka et al., 2012). Matoba et al. demonstrated a comb-driven micro Fresnel mirror (CD-MFM) actuated by electrostatic force (up to 150 V) to tune the fringe spacing, thereby creating suitable measurement conditions for various samples (Matoba et al., 2015). By changing the fringe spacing of the transient grating, the decay time of the mass diffusion can be controlled, therefore the low-voltage micro Fresnel mirror could be a promising tool for a POCT and massively parallel sequential analysis of samples that have different mass diffusion coefficient.

In recent years, low-voltage and long-range microelectromechanical-systems (MEMS) mirrors have been developed using electrothermal actuators based on the bimorph configuration consisting of high and low thermal expansion materials (Sun et al., 2010, and Han et al., 2016). Although the electrothermal actuator has a disadvantage of the limitation of the actuation speed (for the application of the sequential diffusion sensing, only several Hz-order response is required), the electrothermal actuator can achieve significant low driving voltage compared with other MEMS actuators such as piezoelectric actuator (Brunne et al., 2013). In our previous work, a prototype fringe-tunable electrothermal Fresnel mirror was developed, and the feasibility of the proposed MEMS mirror was experimentally confirmed for a compact sensor with a low driving voltage (Kiuchi et al., 2017). The fringe-tunability and the reliability during 24-h period have been confirmed, however the diffusion sensing has not been performed because of the fabrication problem resulting in the limitation of the actuated angle. In this paper, the improved fabrication process is proposed, and the decay time control of the mass diffusion in the transient grating is demonstrated by tuning the fringe period of laser interference using an electrothermal Fresnel mirror.

2. Analysis of the fringe-tunable electrothermal Fresnel mirror (FEFM)

Figure 1 shows a schematic of the measurement principle of the diffusion sensor, which consists of the sample cell and the fringe-tunable electrothermal Fresnel mirror. The photoconductive layer is irradiated with the interference optical pattern, and the lattice-shaped concentration distribution is formed in the sample cell by laser-induced dielectrophoresis. Measurement of the diffusion coefficient is carried out by observing the diffusion process of the microscale transient grating formed by the concentration distribution of the sample. In our case, first-order diffracted light from the probing laser is utilized to increase the sensitivity of signal detection (more detail can be found in (Oka et al., 2012)). To control the decay time of the mass diffusion, which contributes to generating appropriate measurement conditions, the inclination of the mirror angle θ is changed because it governs the fringe spacing A. The decay time constant τ0 observed by the first-order diffracted light is described as (Oka et al., 2012),

\[ \tau_0 = \frac{A^2}{(2\pi)^2D} = \frac{\left(\frac{\lambda}{2 \sin 2\theta}\right)^2}{(2\pi)^2D} \]

where \( \lambda \) is the wavelength of the laser, D is the diffusion coefficient of the sample.

The FEFM was developed to tune the fringe spacing with a low driving voltage. The device has two movable mirror plates and four serpentine-shaped actuators comprised of NiCr alloy and SU-8 to tilt the mirrors. The temperature change attributed to Joule heating of NiCr alloy induces deformation of the actuators through different coefficients of thermal expansion, resulting in tilt of the mirror plates. The schematic of the FEFM and its dimensions are shown in Fig. 1. NiCr alloy and SU-8 were selected as materials to be used for the actuator to generate precise angular control at a low driving voltage. When SU-8 exceeded the glass-transition temperature (\( T_g = 473 \) K), deterioration in the repeatability of actuator driving was lost; therefore, the temperature should not exceed \( T_g \). The temperature rise of the actuator due to Joule heating depends on the thickness of NiCr alloy. The mirrors and the electrothermal actuators are connected by hinges made of SU-8, whose thickness is same as that of serpentine-shaped actuators. In this configuration, both spring constant of hinges...
and heat capacity of thermal actuators are depending on the thickness of SU-8 and govern the actuation angle. For a
detailed design of the FEFM, electrothermal–mechanical-coupling finite-element analysis (CoventorWare) was
conducted. The analytical conditions are summarized in Table 1. In the analysis, the heat transfer due to the convection
and the radiation was considered. Figure 2(a) shows the temperature and mirror angle as a function of the thickness of
NiCr alloy at a driving voltage of 7 V. The temperature of the actuator rises with increasing thickness of NiCr alloy
because of the increase in the electrical current resulting from the decrease of the electrical resistance of the electrodes
made of NiCr. When the thickness of NiCr alloy reaches 300 nm, the temperature of the actuator exceeds the
glass-transition temperature of SU-8. Thus, 200-nm-thick NiCr alloy was selected for the electrothermal actuator.

In addition, the maximum driving angle range at a certain voltage varies with the thickness of SU-8. To maximize the
mirror’s angular shift, the thickness of SU-8 layer was analyzed. The analytical result of the dependence of angular shift
on the thickness of SU-8 at a driving voltage of 7 V is shown in Fig. 2(b). Because the stiffness of the actuator is adjusted
by changing the thickness of the device, the maximum driving angle range depends on the thickness of SU-8. When SU-8
was too thin, the force generated by the drive of the actuator became small compared with the stiffness of the torsion
spring, resulting in small angular shift. Based on our analysis, the thicknesses of NiCr alloy and SU-8 were determined as
200 nm and 10 µm, respectively.

The temperature distribution of the FEFM with the handling layer (Si, thickness of 385 µm) was investigated using
finite-element analysis under the consideration of the heat transfer through the substrate. The analytical result is shown
in Fig. 3. The temperature in the middle part of the actuator increases because heat conduction through the mirror and
substrate was significant in the connection part. Furthermore, significant heating of the mirror was not observed due to
the thermal isolation of the polymer hinges having low thermal conductivity.

![Fig. 1 Schematic of the transient-grating diffusion sensor.](image)
Table 1 Simulation conditions.

<table>
<thead>
<tr>
<th>Mesh type</th>
<th>Manhattan bricks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Minimum mesh size</td>
<td>5 µm</td>
</tr>
<tr>
<td>Ambient temperature</td>
<td>299.68 K</td>
</tr>
</tbody>
</table>

Fig. 2(a) NiCr thickness dependences of the actuator temperature and mirror angle; (b) SU-8 thickness dependence of the mirror angle.

Fig. 3 Analytical temperature distribution at a driving voltage of 7 V.
3. Fabrication of the FEFM

The FEFM was fabricated by patterning SiO$_2$ on a silicon substrate followed by bonding a silicon wafer (as shown in Fig. 4(a)). SiO$_2$, NiCr alloy (80%Ni–20%Cr) and SU-8 were patterned on the bonded silicon-wafer (as shown in Fig. 4(b)). The silicon substrate was subjected to deep-reactive ion etching (DRIE), and the patterned SiO$_2$ was removed by vapor-hydrofluoric-acid etching (as shown in Fig. 4(c)). To improve the yield and quality of the device, the SU-8 film was placed on the flat surface in the air environment at least 10 min after the spin coating of SU-8 to eliminate bubbles in the film. Moreover, additional deposition of SiO$_2$ between the NiCr alloy and the bonded silicon-wafer was introduced to protect the actuators from overetching of the silicon substrate. This oxide thin film was removed by vapor-hydrofluoric-acid etching following deep-reactive ion etching, and the polymer residue was removed by this sacrificial etching. Figure 5 shows the scanning electron microscope (SEM) images of the fabricated FEFM. The device was successfully fabricated without any residue on its surface or cracks in the narrow torsion spring.

Fig. 4 Fabrication of the FEFM.
4. Characterization of the temperature

The thermal-response speed was acquired via infrared thermography (TVS-8500, Nippon Avionics, frame rate: 29.978 fps). Figure 6 shows the temperature-response speed of the FEFM under application of a square wave at 7 V. To evaluate heat conduction from the NiCr heater to the mirror and the other side of the actuator, the actuation voltage was only applied to the left actuator. After applying the voltage \( t = 0 \), Joule heat was immediately generated, and the steady-state temperature was attained within 134.3 ms. Although the temperature at the middle part of the actuator increased, those of the mirror and the substrate hardly changed, corresponding to the analytical result described in Fig. 3. Heat conduction through the mirror was not significant; therefore, the thermal cross-talk between the two actuators was negligibly small. The total thermal resistance of the actuator was roughly estimated to be around \( 6 \times 10^4 \text{ K/W} \) which is higher than bimetal electrothermal actuators (Zhang et al., 2015) because of low thermal diffusivity of SU-8 (0.14 mm\(^2\)/s).

Fig. 5 SEM picture of the fabricated FEFM: (a) whole device; (b) close view of the connection between the mirror and the actuator; (c) close view of the torsion spring.

Fig. 6 Response speed of the FEFM obtained by infrared thermography.
5. Decay time control of mass diffusion using the FEFM

The diffusion process of polystyrene beads of diameter 200 nm was preliminarily observed using the FEFM. The experimental setup used to generate the concentration distribution of the sample and to observe the mass-diffusion process is shown in Fig. 7. This benchtop apparatus with the imaging optics to relay the interference pattern on the sample cell was utilized because the working distance of the FEFM was short (in the order of several mm). These imaging optics were not required when the MEMS packaging of FEFM and the sample cell was accomplished. The excitation laser (wavelength 532 nm) emitted from the laser diode was concentrated onto the FEFM (the mirror surface was uncoated with any high reflectance material) by lens 1 and split via the FEFM. The split beams were reflected and collimated by a non-polarized beam splitter and lens 2, followed by interference on the photoconductive layer in the sample cell using lens 3. The sample cell consists of a photoconductive layer (a-Si:H, thickness of 500 nm), two transparent electrodes (ITO, thickness of 200 nm), a synthetic quartz substrate, and an SU-8 microchannel (channel thickness of 40 μm) modified by aminosilane. The AC voltage was applied between two transparent electrodes, and the dielectrophoretic force was generated by the nonuniform electric field when the photoconductive layer was interferometrically excited. The probe laser (wavelength 637 nm) was concentrated using lenses 3–5 and irradiated onto the formed concentration distribution of the sample, resulting in diffracted light being generated. Decay of the intensity of diffracted light caused by mass diffusion was observed by the photo detector.

Prior to detection of the diffusion phenomenon, the fringe spacing formed by the fabricated FEFM was measured. Table 2 shows the fringe spacing at each driving voltage. The fringe spacing became narrower with increasing voltage applied to the FEFM. The fringe spacing is related to the mirror angle (\( \Lambda = \lambda (2 \sin \theta) \), where \( \Lambda \) is the fringe spacing, \( \lambda \) is the wavelength of the excitation laser, and \( \theta \) is the mirror angle). The experimental and analytical values of the mirror angle obtained by converting the fringe spacing into the mirror angle are shown in Fig. 8. The initial inclination at 0 V was observed in the experiment because of the residual stress generated during the fabrication process. However, the fabricated mirror was tilted by applying the voltage according to the analytical results. Finally, the fabricated mirror achieved a mechanical angle change of 2.76° from the initial state at 0 V to the actuated state at 7V.

![Fig. 7 Schematic of the experimental benchtop setup.](image)

Table 2 Fringe spacing at each driving voltage.

<table>
<thead>
<tr>
<th>Voltage, V</th>
<th>0</th>
<th>3</th>
<th>5</th>
<th>7</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fringe spacing, μm</td>
<td>17.0</td>
<td>11.9</td>
<td>4.07</td>
<td>2.38</td>
</tr>
</tbody>
</table>

Fig. 8 Analytical and experimental mirror angles of the FEFM.

Table 3 Measurement conditions in diffusion sensing.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Sample density, vol%</th>
<th>Voltage applied to the sample cell, $V_{pp}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polystyrene beads, φ200 nm</td>
<td>0.1</td>
<td>10</td>
</tr>
<tr>
<td>Applied frequency, kHz</td>
<td>Intensity of excitation laser, mW</td>
<td>Intensity of probe laser, mW</td>
</tr>
<tr>
<td>10</td>
<td>1.41</td>
<td>1.98</td>
</tr>
</tbody>
</table>

Fig. 9 Decay-intensity signal of first-order diffracted light at each fringe spacing.

In order to confirm the feasibility of the decay time control of the mass diffusion in the transient grating, 200 nm diameter polystyrene beads suspended in DI water was utilized. Table 3 describes the measurement conditions of the diffusion coefficient, and Fig. 9 shows the optical-intensity changes of first-order diffracted light at each fringe spacing altered by the FEFM.

Decay of the diffracted light intensity became faster as the fringe spacing was reduced with increasing voltage.
applied to the FEFM. The decay constant of mass diffusion is related to the fringe spacing. The theoretical value of the mass diffusion coefficient of the dispersed sample in the liquid is calculated by the Stokes–Einstein equation,

$$D = \frac{k_B T}{6 \pi \eta_m r}$$  \hspace{1cm} (2)

where $D$ is the diffusion coefficient, $k_B$ is Boltzmann’s constant, $T$ is the temperature, $\eta_m$ is the solvent viscosity, and $r$ is the sample radius. Figure 10 shows a comparison between the experimentally obtained time constant of the diffracted-light-intensity decay and the calculation result of the time constant, both as functions of the square of the fringe spacing. The decay signals were processed by the inverse problem analysis using the following equation,

$$I = A \exp \left( -\frac{2t}{\tau_D} \right) + B \exp \left( -\frac{t}{\tau_D} \right)$$  \hspace{1cm} (3)

where $I$ is the optical intensity of the first-order diffracted light, $A$ and $B$ are the amplitude factors for the signal and the coherent noise component, respectively. For the precise measurement, the threshold value $A/B \geq 10$ was introduced (for more detail, see Niwa et al., 2009). The measured time constant agreed well with the Stokes–Einstein equation. Moreover, by making the fringe spacing 7.1 times narrower than the initial value, the decay time of diffracted light became 50 times faster (from 3.25 s to 59.2 ms), showing reasonable agreement with the calculation formula. Figure 11 shows the experimental and calculated diffusion coefficients at each fringe spacing. As the fringe spacing decreased, the

![Fig. 10 Calculated and experimental time constants of diffracted-light-intensity decay.](image)

![Fig. 11 Calculated and experimental diffusion coefficients at each fringe spacing.](image)
deviation between the measured diffusion coefficient and the value calculated by the Stokes–Einstein equation became smaller because of the suppression of undesired three-dimensional diffusion over the short mass-diffusion period. In the case of the wider fringe spacing, the distortion of the decay-intensity signal caused the over/under estimation of the time constants even when the signal satisfied the threshold value A/B ≥ 10, therefore, the scattering of the experimental result that was indicated by the error bar became significantly larger. These results indicated that the FEFM can reasonably be used for tuning the mass diffusion time in the transient grating to enhance the measurement accuracy as well as the sensing speed during the sequential sensing of different samples.

6. Conclusion

A fringe-tunable electrothermal Fresnel mirror for a compact and high-speed diffusion sensor was demonstrated. The thicknesses of the NiCr alloy and SU-8 used in the FEFM were designed to maximize the mirror’s angular shift under the condition that the temperature of the actuator does not exceed the glass-transition temperature of SU-8. The temperature distribution obtained by infrared thermography agreed well with the analytical result. By introducing a proposed fabrication process, the experimental mirror angular shift showed a reasonable agreement with the analytical result. The fabricated FEFM realized high-speed, precise measurements; that is, by reducing the fringe spacing due to voltage application to the FEFM, the decay-time constant of diffracted light intensity was successfully shortened 50-fold, which well matched the theory. The experimental results show the validity of the FEFM as a compact and high-speed diffusion sensor realizing control of the decay time in a transient grating. The Joule heating temperature of FEFM is relatively low compared with other bimetal electrothermal actuators, therefore the external ambient temperature control will be required in the future.

Acknowledgments

This research was partially supported by the JSPS KAKENHI Grant Numbers JP24226006 and JP15K13890, Kawasaki city subsidy for promoting R&D of nano–micro technology by SMEs based on academia–industry cooperation. The microfabrication was fabricated at the clean room in “Global nano micro technology business incubation center (NANOBIC), Kawasaki city, Japan” supported by the academic consortium for nano and micro fabrication of four universities (Keio University, Waseda University, Tokyo Institute of Technology, and the University of Tokyo).

References


