OPTICAL REVIEW Vol. 6, No. 1 (1999) 68–70

# Detection of Refractive Index Change of Liquids by Diffractive Element Based Sensor

Raimo SILVENNOINEN,[1] Kai-Erik PEIPONEN[1] and Jukka RÄTY[2]

[1]*Department of Physics, University of Joensuu, P.O. Box 111, FIN-80101 Joensuu, Finland,* [2]*Measurement and Sensor Laboratory, University of Oulu, Technology Park 127, FIN-87400 Kajaani, Finland*

(Received August 17, 1998; Accepted October 8, 1998)

Refractive index change of liquids was detected using a temperature compensated prism cavity together with a diffractive optical element fabricated by electron beam lithography. Small changes of refractive index could be detected from image data of the sensor.

**Key words:** refractive index change of liquids, diffractive optical element, process industry

## 1. Introduction

Data obtained by optical spectroscopy can be exploited for determination of optical constants of liquids. Traditionally reflectometers have provided various measurement modes for the detection of optical spectra from liquids. A typical system is a prism reflectometer which makes it possible to gain information about the refractive index by detecting reflectance near the critical angle of incidence.[1)] In the case of turbid liquids a linear differentiating refractometer has been applied.[2)]

Most of such devices are restricted to operating at a fixed wavelength of incident light. Recently, a reflectometer was developed[3)] which makes it possible to measure reflectance of liquids at ultra-violet visible (UV-VIS) range. Especially the UV region is often important in analysis and identification of liquids appearing in process industries. The applicability of the reflectometer of Ref. 3) was tested by investigating liquids that frequently appear in pulp and paper and in food industries. A common feature with the above mentioned devices is that they are mainly for laboratory use. In other words, they provide tools such as for off-line liquid quality assessment.

Another type of reflectometer is the one that makes use of surface plasmon resonance sensing.[4)] The prism face, which is in contact with the liquid sample, has a thin metal film layer. For certain angle of incidence, and usually for a fixed wavelength, there is a dip in reflectance due to the surface plasmon generation which results in an evanescent wave that emerges from metal to liquid. This technique is rather sensitive for the refractive index[5)] and absorption changes[6)] of liquids, including turbid liquids. However, one has to be sure that the thickness of the metal film is not reduced as a function of time, since the signal depends on the film thickness. In the process industry there frequently appear liquids that can dilute the metal whether we consider off- or on-line measurements. The on-line measurement of liquid quality in the flowing process can be especially problematic.

In this paper we describe new applications of a diffractive element for the detection of refractive index change of liquids. So far we have observed good sensitivity of diffracted light in the detection of metal surface roughness,[7–9)] surface and coating quality of pharmaceutical tablets,[10–12)] wedgeness of ferroelectric material[13)] and, finally, surface porosity of paper.[14)]

The advantage of the present sensor is that we can obtain and process image information. Then we can, for instance, get rid of intensity measurement when detecting refractive index change. The applicability of the sensor is tested using a well-known liquid composition of ethyl alcohol in water. The experiments were performed under laboratory conditions, but the sensor can be built to a robust system, which may find applications for on-line quality assessment of process liquids in industrial environments.

## 2. Experimental and Results

We fabricated a binary amplitude-type diffractive optical element (DOE), which was computed using the scalar diffraction theory in the Fresnel region as explained in Ref. 7). The output pattern of the DOE, reconstructed by a laser beam, produces an image of regular array of a $4 \times 4$ light spot matrix. Positive electron beam resist was spun on a chrome layer, sputtered on a glass substrate (thickness 120 nm), and the resist was exposed by an electron beam writer. The resist was developed and the chrome was wet etched, resulting in a good quality DOE.

Next, we constructed the sensor for the detection of the refractive index change of liquids. A schematic diagram of the setup is shown in Fig. 1. The light source was a low power HeNe laser ($\lambda = 632.8$ nm). A plane wave is incident on the DOE, and thereafter the diffracted wavefront is incident on the temperature compensated prism compartment simultaneously containing reference liquid (distilled water) and alcohol-water solution. The reference homogenous liquid was in the cell at left, the prism compartment was divided into two cells by a glass plate, and the homogenous sample liquid was in the cell at right. The deviation angle of the prism was 72.8°. Due to the small change in refractive index of the sample liquids the diffracted wavefront refracted, at the wedge

E-mail: Raimo.Silvennoinen@joensuu.fi

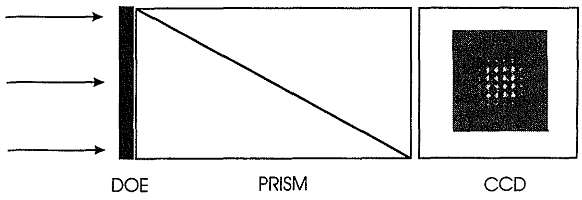


Fig. 1. Schematic diagram of liquid refractive index change sensor. Note that refracted images were incident directly on the chip of the CCD camera.

Table 1. Refractive index measured at 20°C by ABBE refractometer at Sodium $D_1$ wavelength 589.6 nm.

| Alcohol concentration in distilled water (%) | Refractive index |
|---|---|
| 0.000 | 1.3327 |
| 0.125 | 1.3328 |
| 0.250 | 1.3329 |
| 0.500 | 1.3330 |
| 1.000 | 1.3331 |
| 2.000 | 1.3336 |

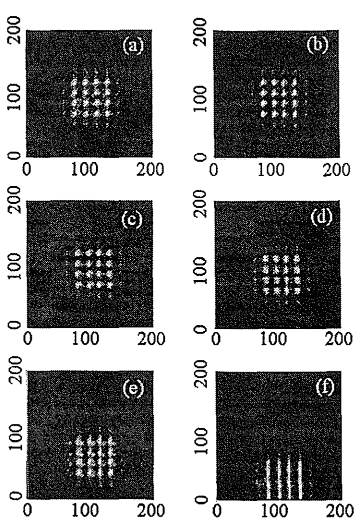


Fig. 2. The spot images for different alcohol concentrations of distilled water-alcohol mixture when distilled water was acting as a reference liquid. The angular deviation is changing in vertical direction: (a) alcohol percentage (in distilled water) $p=0\%$, (b) $p=0.125\%$, (c) 0.250%, (d) $p=0.5000\%$, (e) $p=1.000\%$, and (f) $p=2.000\%$. The numbers shown on the axes are the pixel numbers.

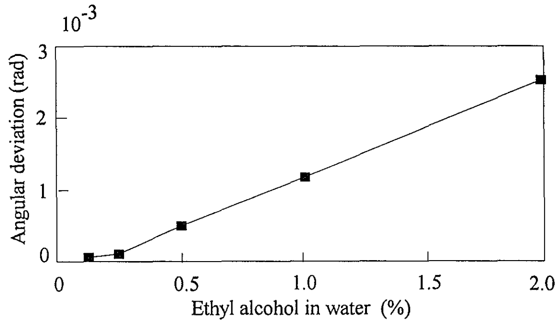


Fig. 3. Angular deviation of the sensor image as a function of alcohol concentration in water.

surface, to slightly different angles. The change of the angle of refraction was detected by a CCD camera located at the focal plane ($f=200$ mm) of the DOE. As a result we observed spatial change of the image of the light spot matrix. Due to the separation of the individual light spots (125 $\mu$m) it is, in principle, possible to observe local changes in liquid concentration (this topic was out of scope of the present study) or aberrations in prism system. Furthermore, when using 16 light spots instead of a single one, we obtain better spatial resolution of the refracted light beam. The angle resolution of the sensor, taking into account the pixel size ($8 \times 8\ \mu m^2$) of the CCD camera, can be calculated as 80 $\mu$rad.

For test solutions we prepared a series of purified ethyl alcohol-water samples with low volume concentration of alcohol. The amount of alcohol was measured using a digital pipet. For the present purpose, samples with alcohol volume percentage of 0.125%, 0.250%, 0.500%, 1.000%, and 2.000% were prepared.

First, we measured the refractive indices of the samples by ABBE refractometer (Bellingham & Stanley, Model 60/ED). The results of the measurements are presented in Table 1. Next we detected the angular deviations obtained from the samples with low alcohol concentration, using the sensor shown in Fig. 1. The spot images for different alcohol concentrations of distilled water-alcohol mixtures, when distilled water was acting as a reference liquid, are presented in Fig. 2. From Fig. 3 we can observe that the order of angular deviation, drawn out from the data of Fig. 2, was 1 mrad. In addition, relatively linear relation between angular deviation and alcohol concentration could be detected. The growth of the angular deviation as alcohol concentration increased, was consistent with the refractive index increase measured by ABBE refractometer at wavelength 589.6 nm.

## 3. Discussion

We have constructed a diffractive element based sensor for observation of small refractive index/concentration changes of transparent liquids. Such liquids are frequently used in chemical and pharmaceutical process industries. One way to detect process liquid quality is to let process liquid flow into prism sample compartments, which are located at different process stages. The use of DOE provides us freedom in designing sensors to study different types of process liquids, since the focal length of the DOE as well as the geometry of the prism can be adjusted and optimized depending on the optical properties of the liquids and measurement environments.

We wish to emphasize that although we here presented data for "good quality" liquids, the sensor can also be employed for quality assessment of "poor quality" such as turbid liquids. Indeed, we have also studied turbid liquid samples obtained from different process stages at a paper mill. We found that the refractive index and also turbidity of the light scattering liquid samples, obtained from a paper mill, could be detected by the present sensor by using a slightly different configuration for the turbidity measurement. The results of such measurements will be presented elsewhere.

**Acknowledgment**

K.-E. P. wishes to thank the Academy of Finland for financial support during this study.

**References**

1) G.H. Meeten and A.N. North: Meas. Sci. Technol. **6** (1995) 214.
2) J.E. Geake, C.S. Mill and M.S. Mohammadi: Meas. Sci. Technol. **5** (1994) 531.
3) J. Räty, E. Keränen and K.-E. Peiponen: Meas. Sci. Technol. **9** (1998) 95.
4) C. Nylander, B. Liedberg and T. Lind: Sens. Actuators **3** (1982/83) 79.
5) H. Kano and S. Kawata: Jpn. J. Appl. Phys. **34** (1995) 331.
6) H. Kano and S. Kawata: Appl. Opt. **33** (1994) 5166.
7) J.T. Räsänen, M. Savolainen, R. Silvennoinen and K.-E. Peiponen: Opt. Eng. **34** (1995) 2574.
8) R. Silvennoinen, J. Räsänen, M. Savolainen, K.-E. Peiponen, J. Uozumi and T. Asakura: Sens. Actuators A **51** (1996) 117.
9) M. Savolainen, J. T. Räsänen, K.-E. Peiponen and R. Silvennoinen: Opt. Eng. **35** (1996) 3637.
10) R. Silvennoinen, K.-E. Peiponen, P. Laakkonen, J. Ketolainen, E. Suihko, P. Paronen, J. Räsänen and K. Matsuda: Meas. Sci. Technol. **8** (1997) 550.
11) K.-E. Peiponen, R. Silvennoinen, J. Räsänen, K. Matsuda and V.P. Tanninen: Meas. Sci. Technol. **8** (1997) 815.
12) R. Silvennoinen, K.-E. Peiponen, M. Sorjonen, J. Ketolainen, E. Suihko and P. Paronen: J. Mod. Opt. **45** (1998) 1507.
13) R. Silvennoinen, K.-E. Peiponen, A. Krumins and J. Räsänen: Opt. Mater. **7** (1997) 145.
14) R. Silvennoinen, K.-E. Peiponen, J. Räsäsnen, M. Sorjonen, E.J. Keränen, T. Eiju, K. Tenjimbayashi and K. Matsuda: Opt. Eng. **37** (1998) 1482.