Probing Specific Gravity in Real-time With Graphene Oxide Plasmonics

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Supporting Information

Correlation between RI and SG of aqueous samples

For the purpose of correlation, refractive index (RI) and specific gravity (SG) are measured by the MASTER-SUR/Nα clinical refractometer (ATAGO CO., LTD, Japan) and the portable density/specific gravity/concentration meter (Anton Paar DMA 35 Ex, Japan) respectively, for solutions of NaCl, C\textsubscript{12}H\textsubscript{22}O\textsubscript{11}, and C\textsubscript{2}H\textsubscript{5}OH dissolved in 18.2 MΩ cm\textsuperscript{-1} Milli-Q water (3 mL).

<table>
<thead>
<tr>
<th></th>
<th>Mass</th>
<th>100 mg</th>
<th>200 mg</th>
<th>300 mg</th>
<th>400 mg</th>
<th>500 mg</th>
<th>600 mg</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaCl RI</td>
<td>1.3375</td>
<td>1.3417</td>
<td>1.3455</td>
<td>1.3490</td>
<td>1.3524</td>
<td>1.3555</td>
<td></td>
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<tr>
<td>NaCl SG</td>
<td>1.0220</td>
<td>1.0424</td>
<td>1.0624</td>
<td>1.0764</td>
<td>1.0941</td>
<td>1.1088</td>
<td></td>
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<tr>
<td>C\textsubscript{12}H\textsubscript{22}O\textsubscript{11} RI</td>
<td>1.3380</td>
<td>1.3426</td>
<td>1.3470</td>
<td>1.3510</td>
<td>1.3544</td>
<td>1.3554</td>
<td></td>
</tr>
<tr>
<td>C\textsubscript{12}H\textsubscript{22}O\textsubscript{11} SG</td>
<td>1.0103</td>
<td>1.0236</td>
<td>1.0345</td>
<td>1.0460</td>
<td>1.0549</td>
<td>1.0651</td>
<td></td>
</tr>
<tr>
<td>C\textsubscript{2}H\textsubscript{5}OH RI</td>
<td>1.3355</td>
<td>1.3380</td>
<td>1.3405</td>
<td>1.3430</td>
<td>1.3450</td>
<td>1.3475</td>
<td></td>
</tr>
<tr>
<td>C\textsubscript{2}H\textsubscript{5}OH SG</td>
<td>0.9926</td>
<td>0.9871</td>
<td>0.9811</td>
<td>0.9767</td>
<td>0.9719</td>
<td>0.9680</td>
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</table>

Figure S1 shows how refractive index (RI) values are correlated with specific gravities (SG) of various aqueous samples. In Figure S1a, RI values were measured by using hand-held ATAGO refractometer, and SG values by using Anton Paar density/specific gravity meter. Namely, after an aqueous sample was prepared, its RI and SG were measured using above mentioned instruments. These values were then plotted against each other. Since SG depends on the density of the analyte in the sample, it is an independent variable.

Thereafter, we tested the same solutions for SPR measurements by Biacore T200 SPR machine. From Figure S1b, it is seen that changes in SPR response units follow the same trend as changes in RI values in Figure S1a. In addition, from the plot in the Figure S1a, RI values are linearly dependent on the SG values of the tested solutions ($R^2 = 0.9997$). For these reasons, we plotted SG values acquired by Anton Paar density/specific gravity meter against SPR response units obtained from Biacore T200. Figure S1c displays that SPR responses are linearly dependent on the SG values of the analyzed samples. Based on this information, we can then create calibration curves for standard aqueous samples containing salt, sugar, and ethanol, diluted in Milli-Q water (18.2 MΩ cm\textsuperscript{-1}), as well as for real food samples containing salt and sugar dissolved in soy sauce, green tea, and apple juice.
Figure S1: Correlation of RI values (acquired by ATAGO refractometer) and SG values (acquired by Anton Paar density/specific gravity meter) for various aqueous samples.

Spectroscopic Ellipsometry measurements of real and imaginary parts of permittivity

Figure S2: Real and imaginary parts of permittivity of the (a) Au SPR and (b) Au-GO SPR chip surfaces.

Spectroscopic ellipsometry is an optical analysis technique used to measure reflection of the light from the material. Reflection of the light from the sample causes change in the polarized light which in turn is detectable by ellipsometry. Ellipsometer measures amplitude ratio $\phi$ and phase difference $\delta$ between p- and s-polarized light waves. These measurements thereafter can be converted into real and imaginary parts of the complex dielectric constants. Results from spectroscopic ellipsometry (Horiba UVISEL 2 Spectroscopic Ellipsometer, Japan) display a significant decrease in the imaginary part of the dielectric function upon
deposition of a thin GO layer. Plasmonic materials such as Au generally show high values of imaginary part of the dielectric function which is associated with high optical losses. However, adding GO on the surface of Au film decreased optical loss coefficient from 3.276 to -0.067 at 500 nm. This implies that deposition of GO decreased absorption loss and, thus, led to a higher sensitivity caused by stronger surface plasmon coupling at the sensor surface. However, it is to be noted that to obtain absolute values for real and imaginary parts of the complex dielectric constant, appropriate fitting of the model is required. Nevertheless, for the sake of this discussion, general comparison between permittivities of Au and Au-GO sensors is sufficient.