Real-time Sensing of NaCl Solution Concentration at Microwave Frequencies Using Novel Ag Patterns Printed on Flexible Substrates

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Abstract. A novel microwave sensor for real-time analyte composition analysis is reported. The sensing element in a form of silver pattern was printed on polyimide flexible laminate substrate to suit a broad range of applications where a sensor could be placed in water reservoirs or fluid-carrying pipes for continuous analysis. The developed system confirmed the viability of using microwaves for real-time NaCl monitoring as the reflected signals represented by $S_{11}$ parameters corresponding to deionised water and 0.1M and 0.01M of NaCl were unique with clearly observed shifts in the resonant frequencies when placed in direct contact with 20 µl of each solution.

1. Introduction
The composition of the analyte solutions must be determined with high accuracy for many applications, such as food and pharmaceutical industries, chemical processing and environmental monitoring, where the content of impurities in water is an important factor [1].

Traditional methods of aqueous solutions composition monitoring are lab-based and include standard UV-Vis measurements, mass spectrometry, ion-sensitive electrodes and amperometric sensors [2, 3]. On the other hand, a range of modern real-time monitoring approaches exist; for example, fibre-optic sensors, MEMs, lab-on-chip sensors and biosensors. It was also demonstrated that micro-Raman spectroscopy can allow the determination of the NaCl salt concentration of an aqueous solution with an error close to ±5% [4].

No single system available today can fully address the needs of the customers in its ability to determine, in real time, the composition of water solutions to the desired sensitivity level, bearing in mind the desire for system portability and cost-effectiveness. Therefore, novel real-time monitoring techniques are necessary. One technique potentially capable of meeting this current demand is based on the microwave sensing, as presented in this paper.

2. Microwave sensing
Microwave sensing is a novel but rapidly developing technology which has been successfully used as a sensing method for various industrial applications including fluid level measurements [5], material moisture content [6, 7], for continuous process monitoring of biogas plants [8], for the determination

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of moisture content in soil [9] and in the healthcare industry, for example for real-time monitoring of glucose in diabetic patients [10, 11] and for non-invasive monitoring of bodily fluids [12, 13].

Microwave sensors in a form of a cavity resonator for accurate measurements of both organic (sugar, alcohol) and inorganic (NaCl, KMnO₄) water solutions concentrations were reported [1]. Notably, the sensitivity of the sensor in determination of NaCl was 0.4 dB/(mg/ml) within 0-1 % concentration range. The sensor was able to detect the concentrations of other water solutions, but its sensitivities are strongly dependent on the type of tested chemical ingredient.

However, the microwave planar printed patterns for various sensing applications are increasingly used due to their versatility, flat profile and low weight. Their design can be tailored to suit particular applications, coupled with reliability and cost-efficiency. They are easily manufactured using common methods for printed circuit board production, and their impedance can be matched to the input line by altering the micro-strip line feed configuration.

The patch antenna represents the frequency-selective element of a phase shift transistor oscillator. The active integrated antenna frequency of operation is determined by both the patch geometry and the electrical loads connected at the two microstrip ends [14]. A convenient termination is normally represented by two open-ended stubs whose electrical length is λ/4 at the patch resonant frequency, where λ is a complete wavelength. The performance of a microstrip resonator depends on its electromagnetic field distribution, resonant frequency and quality factor (Q).

To clarify the principle behind electromagnetic wave sensing suggested in this work for water quality control, it is worth mentioning that microwave sensors in the form of planar printed patterns operate based upon the fact that an object under test, e.g. a water sample, when placed into the vicinity or in direct contact with a microwave sensor, interacts with the electromagnetic waves in a unique manner, which can be specifically correlated with the properties of this material.

By considering how reflected (S₁₁) microwave signals vary at discrete frequency intervals, the change in the signal can be linked to the composition of the object under test.

3. Experimental procedure

3.1. NaCl solutions preparation
Sodium chloride NaCl (Sigma S6150) was dissolved in deionised water at 0.1M and 0.01M concentrations (equivalent to 5800 ppm and 580 ppm respectively) and produced clear solutions.

3.2. Microwave sensor structure
The layout and dimensions of the sensor pattern is shown in Figure 1. Silver was used as the conductive metal material for both the bottom layer, which acted as a ground plane, and the top pattern to maintain chemical neutrality when the device is placed in contact with analyte solution. The thickness of the Ag layers was 35 µm. Microwave sensor was designed on 2 mil thick DuPont™ Pyralux® AP Polyimide Flexible Laminate substrate.

A distinct feature of IDE type sensors is their superior sensitivity to change close to the sensor surface, with this sensitivity decaying rapidly with distance away from the surface, as shown in Figure 2. This is advantageous as it reduces significantly the chance of undesirable factors influencing sensor response.

3.3. Measurement setup
The microwave sensor was attached to a Rohde and Schwarz ZVA24 vector network analyser (VNA) via a coaxial cable, as shown in Figure 3. An SMA type Molex connector specified for 50Ω impedance was used. The data (60,000 points for each measurement) was captured in the 0-15 GHz frequency range for the reflected (S₁₁) signals at constant temperature of 18 °C. The results are repeatable with less than 5% deviation. Notably, average sensor responses are depicted in the graphs shown in following section.
4. Results and discussion

The spectra for the microwave sensor were recorded in the 0-15 GHz frequency range when in contact with 20 µl of deionised water and NaCl analyte solutions in 0.1M and 0.01M concentrations. The ability of this sensor to distinguish the presence and concentration of NaCl is evident due to the change in the amplitude and resonance frequency of the peak occurring at frequencies below 2.5 GHz, as illustrated in Figure 4.

One may choose to monitor the change in the amplitude and frequency shift of a single resonant peak, such as that near 2 GHz, as shown in Figure 5 which depicts the spectra at 1.5-2.5 GHz range.
Figure 4. $S_{11}$ signal distribution of the microwave sensor in the 0-2.5 GHz range when in contact with deionised water and NaCl solutions in 0.1M and 0.01M concentrations.

Figure 5. $S_{11}$ signal distribution of the microwave sensor in the 1.5-2.5 GHz range when in contact with deionised water and NaCl solutions in 0.1M and 0.01M concentrations.
Figure 6. $S_{11}$ signal distribution of the microwave sensor in the 0-1.2 GHz range when in contact with deionised water and NaCl solutions in 0.1M and 0.01M concentrations.

Additionally, lower ranges below 1.2 GHz are characteristic of NaCl presence and concentration, since each spectrum, as shown in Figure 6, is different. It is possible that each spectrum pattern could be used to determine the analyte presented to the sensor. Thus, the developed sensor demonstrates a distinct ability not only to detect the presence of NaCl in aqueous solution, but to measure its concentration as well.

Figure 7. Optical image of a microwave sensor with 20 µl water sample placed over it illustrating hydrophobic nature of the flexible substrate.

Notably, the sensors’ responses returned to their original positions, namely air spectra, after each water sample measurement, confirming that the developed microwave sensors are reliable, re-usable and thus a sustainable solution for water quality monitoring. This could be explained by the hydrophobic nature of the DuPont™ Pyralux® AP Polyimide flexible laminate substrate used in the construction of these sensors, as shown in Figure 7.

**Conclusion**

This research was driven by the need for a novel, versatile, sensitive and cost-effective real-time monitoring method of NaCl presence and concentration in solution. A sensor with a silver planar
pattern was developed to verify the potential applicability of the microwave sensing approach for this task. It is believed that the sensor’s flexible structure would afford good longevity since this configuration is less prone to failure resulting from mechanical damage than typical alternatives. The sensor responses were tested in the 0-15 GHz frequency range. It was clearly shown that the resonant peaks amplitude and frequency changes once the deionised water and NaCl solutions in 0.1M and 0.01M concentrations (20 µl volumes) were placed in contact with the sensor pattern.

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