Abstract—An optimized microfluidic sensor for extracting volume ratio of binary mixture comprising of ethanol and methanol using electrical resonance technique has been presented in this work. In order to detect small changes in composition of binary mixture, a split-ring resonator structure with enhanced sensitivity was designed to operate around 2.5 GHz. A resonator was designed using HFSS, which possessed enhanced sensitivity. A novel algorithm for optimization was devised for binary mixture of the two liquids. The resonator was fabricated and tested for validation of results. Samples of ethanol and methanol mixture in different volume ratios were prepared and filled in micro-capillary tubes. These tubes were placed inside the resonant structure to perturb electric field. Variations in resonant properties due to change in volume ratio of liquid mixtures were analyzed. Resonant frequency, $s$-parameters, and quality factor of structure were measured. It was observed that change in volume fraction as small as 1/100 resulted a shift of 0.25 MHz in resonant frequency (relatively high level of sensitivity). Measured results were utilized by mathematical model to compute volume fraction of liquid in these mixtures.

1. INTRODUCTION

Volume fraction of liquids in mixtures is strictly maintained in various applications to ensure requisite standard and quality. These fields include chemistry, biology, petroleum, petrochemistry, agriculture, etc. [1–3]. Variation in liquid composition during mixing may lead to failure or poor performance. In such applications, composition is determined at various stages to ensure conformance to the laid down ratios. Numerous techniques like liquid chromatography, boiling point distillation, and capillary electrophoresis are available to determine volume fraction of liquids in mixture [4]. These methods are time consuming, require expensive equipment, and differ in techniques depending upon nature of liquid(s). Microwave techniques for determining volume fraction of constituents in a mixture have several advantages, like small sample volume [2], instantaneous result [4], small infrastructure, ease in handling, high sensitivity [4], etc.

Microwave technique for determining volume fraction of constituents in a sample under test is based on permittivity sensing capability of certain devices [2, 4, 5]. Resonant techniques are preferred over other microwave techniques (e.g., transmission and reflection methods) due to high accuracy [4]. In resonant techniques, small volume of material sample is placed inside a resonator. The resonator is then energized with electromagnetic waves. Placement of sample perturbs the resonator volume and electromagnetic field due to which this method is referred as perturbation method [2, 4]. Resonant parameters are measured before and after placement of the sample [2–4, 6]. $S$-parameters are measured and shift in resonant frequency, and quality ($Q$) factor is obtained [5, 6]. Analysis of resonant frequency and $Q$ factor determines composition of constituents in a sample.
Numerous works have been reported for permittivity sensing [7–14]. Different devices have been used in these works; however, technique remains almost the same. Compositional analysis has been done with liquid constituents placed in micro-capillary tubes [2, 3, 15]. It has been established in these works that high-resolution results are achieved with resonators having better sensitivity for permittivity. This observation motivated us to undertake this work. In earlier works, efforts have been made to design a resonator with enhanced sensitivity. However, devising a resonant structure using an algorithm is a novel idea which is reported in this work. Initially, a split-ring resonator structure, to operate around 2.5 GHz, was designed using Ansoft’s 3D full wave field solver High Frequency Structure Simulator (HFSS) [16]. Sensitivity of structure was then optimized [17], using an algorithm devised for binary mixture of ethanol and methanol. Samples of liquid solvents contained in micro-capillary glass tubes were placed inside the resonator structure for measurement. A mathematical model was developed for determining volume fraction of liquid in mixture using measured values.

2. MICROWAVE TECHNIQUE FOR DETECTION OF MATERIAL PERMITTIVITY USING SPLIT-RING RESONATOR

2.1. Permittivity and Its Association with Resonant Behavior

Dielectric materials have an inherent capability of storing energy when being subjected to an electrical field [9, 18, 19] known as permittivity. When this quantity is compared with free space, it is referred as relative permittivity [18]. It is a complex quantity ($\varepsilon^*_{r}$) and described as [19, 20]:

$$\varepsilon^*_{r} = \varepsilon'_{r} - j\varepsilon''_{r}$$  \hspace{1cm} (1)

where real part ($\varepsilon'_{r}$) signifies charge storage capability of material, and imaginary part ($\varepsilon''_{r}$) determines rate at which stored energy is dissipated (relative to free space). Each dielectric material has a unique set of real and imaginary parts of permittivity.

Microwave techniques for determining complex relative permittivity are grouped into three major categories namely reflection, transmission, and resonance [21, 22]. Present work is based upon resonance method for obtaining results with high accuracy. In resonance perturbation method, resonant frequency and $Q$ factor are two parameters which are used to determine permittivity of material. In this method, complex permittivity of a material is sensed when a small sample under test (SUT) is placed within a resonator. Shift in resonant frequency ($f_0$) and quality ($Q$) factor of resonator are observed for cases before and after insertion of sample. These effects are analyzed by first order perturbation theory [21, 23, 24]. Real and imaginary parts of materials’ permittivity are related with these shifts as under [4]:

$$\varepsilon'_{r} \propto \Delta f_0$$  \hspace{1cm} (2)

$$\varepsilon''_{r} \propto \Delta \left( \frac{1}{Q} \right)$$  \hspace{1cm} (3)

Any change in resonant behavior of a sample loaded resonator is indicative of change in volume fraction. Systems with high sensitivity yield larger shifts in these quantities for the same change in volume fraction.

2.2. Split-Ring Resonator Technique for Permittivity Sensing of Liquid Mixtures

Theory of split-ring resonator (SRR) [25] was established in early 1980’s [25–27]. Single gap SRR enclosed in a metallic cavity is shown in Figure 1 [27–29]. Resonator can be considered as a single-turn inductor connected with a gap capacitor [30] which is surrounded by cavity/shield wall. A high $Q$ factor is achieved due to shielding. Numerous mathematical models have been formulated to predict resonant frequency and $Q$ factor [25–28, 31, 32].

Utilization of SRR for permittivity sensing of materials has been reported in various works [2, 3, 15]. Liquid samples contained in small containers, micro-capillary tube or micro-fluidic channel are placed in resonator gap [2–4, 15, 33] which perturbs electric field in the gap [34]. This perturbation affects resonant behavior [2, 15] and can be measured using Vector Network Analyzer (VNA). Shifts in resonant frequency and $Q$ factor are used for permittivity sensing/measurement [22].
3. DESIGNING OF IMPROVED SPLIT-RING RESONATOR

It was observed while studying earlier works [3, 15, 21] that better resolution in permittivity sensing was achieved when SRR gap was longitudinal in shape, and micro-capillary tube containing liquid was placed parallel to electrical field. It was also observed that resonator with higher $Q$ factor yielded better resolution while performing compositional analysis of liquid solvents. These observations were used in this work and sensitivity of a predesigned SRR [17, 29]. This was achieved with simulations designed in HFSS, using a novel algorithm. Improved SRR structure, with enhanced sensitivity, was then fabricated and tested for verification.

A predesigned SRR structure (initial design), accommodating micro-capillary tube parallel to electric field plane, was initially designed with HFSS. This structure had a square cross-sectional area. Copper was used for the resonator while shield was designed using aluminum [35, 36]. Variation analysis [36] was performed, using the algorithm, to study the effect on output parameters with changes in SRR geometry. For this analysis geometrical parameters were varied within allowable ranges. Algorithm required observance of shift in resonant frequency and $Q$ factor, when $\varepsilon_r$ was varied from 1 to 85. These were noted for each change in the geometry. A design in this way was obtained which could yield larger variation in resonant frequency and $Q$ factor. Design and output parameters with and without a micro-capillary tube are given in Table 1. Shifts in resonant frequency and $Q$ factor, with $\varepsilon_r$ for final design, are shown in Table 2, Figures 2 and 3 respectively.

4. FABRICATION, EXPERIMENTATION AND MATHEMATICAL MODELLING FOR VOLUME FRACTION EXTRACTION

An SRR structure was fabricated with dimensions as mentioned in Table 1. SRR was fabricated with copper while aluminum was used for shield [35, 36]. Resonator gap was kept at 2 mm to accommodate Marienfeld cat. no. 2940202 capillary tube [37] which was used to store liquid samples. An identical pair of magnetic loop coupling was prepared, with RG 402 semi-rigid coaxial cable. These couplings were used for connecting SRR structure with VNA. At opposite sides of shield wall, a pair of holes was made at the center of its height to facilitate insertion of magnetic loop coupling. A hole was also made for placing micro-capillary tube within resonator gap. A low loss base was prepared with polystyrene to place resonator inside shield.

Agilent E8362B VNA was used for two-port $S$-parameters measurements. Figure 4 shows experimental setup. Resonant frequency and loaded $Q$ factor of SRR structure without micro-capillary
Table 1. SRR design parameters and output results.

| Parameters                          | Values |
|-------------------------------------|--------|
| Inner Radius of Shield ‘R₀’ (mm)    | 22     |
| Inner Radius of Resonator ‘r₀’ (mm) | 5      |
| Width of Resonator ‘W’ (mm)         | 6      |
| Length of Resonator ‘Z’ (mm)        | 3      |
| Gap of Resonator ‘t’ (mm)           | 2      |
| Height of Shield ‘h’ (mm)           | 32     |
| Thickness of Shield ‘T’ (mm)        | 7      |

Output parameters

| Without tube inside SRR gap | Resonant frequency ‘f₀’ (GHz) | 2.55 |
|                           | Q factor                      | 2818 |
| With tube inside SRR gap  | Resonant frequency ‘f₀’ (GHz) | 2.571 |
|                           | Q factor                      | 3174.9 |

Table 2. Results of optimized SRR.

| Parameter               | Min   | Max   | Δ     |
|-------------------------|-------|-------|-------|
| Resonant frequency 𝑓₀ (GHz) | 2.394 | 2.571 | 0.177 |
| Q Factor                | 3007.6| 3174.9| 167.3 |

Figure 2. Variations in resonant frequency against 𝜀ᵣ′.

tube were initially measured. Unloaded Q factor was calculated as:

\[
Q = \frac{Q_L}{1 - 10^{IL/20}}
\]  

where IL represents measured insertion loss which was −15.68 dB. Measured and calculated values of resonant frequency and Q factor were 2.6974 GHz and 2312.82 against 2.55 GHz and 2818, respectively.
Figure 3. Variations in $Q$ factor against $\varepsilon'_r$.

Figure 4. (a) Top view of resonator without lid. (b) Experimental setup.

Figure 5. $S_{21}$ versus resonant frequency measurements.

This difference is attributed to factors as mentioned in earlier works [35].

Mixtures of ethanol and methanol were prepared with different volume ratios. Measurements were made for mixture samples placed inside SRR structure. Figure 5 shows results of these measurements. Shifts in resonant frequency and $Q$ factor were observed due to change in effective permittivity of mixtures. Values of resonant frequency and loaded $Q$ factor in each case were measured as shown in
Table 3. Measured results.

| Volume Ratio Ethanol : Methanol | Resonant Frequency $f_0$ (GHz) | Loaded Q Factor $Q_L$ | Insertion Loss IL (dB) | Calculated Q Factor |
|---------------------------------|-------------------------------|------------------------|------------------------|---------------------|
| 0 : 1                           | 2.4944                        | 67.63                  | −34.95                 | 68.86               |
| 1 : 3                           | 2.4999                        | 61.80                  | −35.00                 | 62.92               |
| 1 : 2                           | 2.5022                        | 59.31                  | −35.41                 | 60.34               |
| 1 : 1                           | 2.5071                        | 54.33                  | −35.61                 | 55.25               |
| 2 : 1                           | 2.5125                        | 49.48                  | −35.26                 | 50.35               |
| 3 : 1                           | 2.5151                        | 47.36                  | −35.40                 | 48.18               |
| 1 : 0                           | 2.5209                        | 44.01                  | −35.92                 | 44.73               |

Figure 6. Resonant frequency and $Q$ factor against volume ratio.

Table 4. Fraction of ethanol calculation.

| Volume Ratio Ethanol : Methanol | Resonant Frequency $f_0$ (GHz) | Calculated Q Factor | Fraction of Ethanol Actual | Calculated |
|---------------------------------|-------------------------------|---------------------|-----------------------------|------------|
| 0 : 1                           | 2.4944                        | 68.86               | 0.00                        | 1.023 × 10$^{-11}$ |
| 1 : 3                           | 2.4999                        | 62.92               | 0.25                        | 0.25       |
| 1 : 2                           | 2.5022                        | 60.34               | 0.3333                      | 0.33333    |
| 1 : 1                           | 2.5071                        | 55.25               | 0.50                        | 0.50       |
| 2 : 1                           | 2.5125                        | 50.35               | 0.6666                      | 0.66666    |
| 3 : 1                           | 2.5151                        | 48.18               | 0.75                        | 0.75       |
| 1 : 0                           | 2.5209                        | 44.73               | 1.00                        | 1.00       |

Table 3. Figure 6 shows graphical results for different volume ratios.

Obtained values of resonant frequency and $Q$ factor given in Table 3 were used to derive 3rd order equation with Wolfram’s Mathematica [38]. Volume fraction of ethanol was extracted using following
equation:

\[
\text{Volume fraction}_{\text{Ethanol}} = 860.470179 + 100.477334 \times f_0 - 44.130482 \times f_0^2 - 47.232291 \times f_0^3
- 35.791211 \times Q + 0.483681 \times Q^2 - 0.000333 \times Q^3 - 1.717178 \times f_0 \times Q
- 0.167012 \times f_0 \times Q^2 + 5.710639 \times f_0^2 \times Q
\] (5)

Table 4 shows results obtained for volume ratios against resonant frequency and \(Q\) factor. The obtained equation represents a model to predict fraction of ethanol in sample mixtures with high accuracy.

Resonant frequency and \(Q\) factor has been shown graphically as a function of volume ratio in Figures 7 and 8, respectively. Such a behavior conforms to earlier analysis.

**Figure 7.** Resonant frequency as a function of volume ratio.

**Figure 8.** \(Q\) factor as a function of volume ratio.
5. CONCLUSION

An optimized SRR and mathematical model for volume fraction extraction has been presented. Optimized resonator was designed, fabricated, and tested. Optimization was focused for enhancing sensitivity of resonator to permittivity variation. Mixtures of ethanol and methanol in different volume ratios were prepared. $S$-parameters were measured for each sample of mixture. Resonant frequency and $Q$ factor were observed as a function of volume fraction and mixture permittivity. A mathematical model for extracting volume fraction of ethanol in mixture was worked out. Optimized structure exhibited large variation in resonant frequency for small variation in composition of liquid mixture. Resonant frequency varied by 25.8 MHz for complete range whereas $Q$ factor varied by 21.83. This means that change in volume fraction as small as 1/100 would result in shift about 0.25 MHz in resonant frequency. A fractional change as small as 1/1000 could be detected if measuring equipment can sense variation of 0.025 MHz. Benefits of optimized resonant structure can also be utilized in applications like oscillator, frequency meter, tuned amplifier, filter, electron paramagnetic resonance, food grading, quality control, etc.

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