Direct Broadband Dielectric Spectroscopy of Liquid Chemicals Using Microwave-Fluidic Two-Wire Transmission Line Sensor

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Abstract—A novel robust approach for permittivity measurements of liquid materials in a wide frequency range is proposed. A high-sensitivity microwave-fluidic sensor is developed enabling direct determination of liquid samples’ permittivity from the measured sensor’s propagation constant. Importantly, neither sensor calibration using liquid standards nor sensor’s geometry (except length) information is required, which is very advantageous for industrial applications. This is achieved through the sensor’s geometry, which is a two-wire transmission line (TWL) encapsulated in a 3-D printed container, and thus, the liquid-under-test fully encloses the propagating electromagnetic waves. The measurement setup comprises the sensor cascaded between two microstrip-to-TWL transitions for which a simplified de-embedding procedure is proposed using only a THRU standard. For demonstration, a setup was developed with transitions operating within 1–10 GHz and tested up to 18 GHz by measurement of a set of alcohols to produce useful data within the transition bandwidth, which can be extended at the cost of increased uncertainty by postprocessing. Finally, the measurement error study has shown that a relatively low uncertainty and error with respect to the reference is obtained, verifying the usability of the developed measurement technique.

Index Terms—Broadband liquid sensing, microwave-fluidic, permittivity measurement, two-wire sensor.

I. INTRODUCTION

CHARACTERIZATION of and monitoring the properties of resources for quality assessment are important industrial processes. The growing demand that increases the pace of production currently enforces a need for the development of suitable measurement techniques that are fast and accurate. Among others, methods utilizing electromagnetic (EM) waves are of great interest since the EM waves penetrate the materials and allow for nondestructive characterization with high measurement speed. In such cases, the electrical permittivity is a quantity of interest. A lot of research effort has already been put into the development of techniques for biological material characterization [1], [2], chemical analysis [3], food products monitoring [4]–[9], etc. In general, the permittivity measurement methods for both solid and liquid materials can be divided into two types: resonant [10], [11] and transmission/reflection types [12]–[19]. The resonant methods can yield high accuracy; however, the material-under-test is characterized only at one or more discrete frequencies. Such methods are suitable, among others for the determination of permittivity deviation or constituents’ mixing ratio in a solution once an appropriate model is known. On the other hand, the transmission/reflection methods are favored for broadband dielectric spectroscopy (BDS). In the MHz to THz frequency range, molecular and collective dipolar fluctuations, charge transport and polarization effects at inner and outer boundaries occur and determine the dielectric properties of the material under study [20]. Therefore, BDS enables to gain a wealth of information on the dynamics of bound (dipoles) and mobile charge carriers depending on the details of the molecular system.

The sensitivity of sensors is determined by their construction, which depends on the amount of the associated EM field that is enclosed within the material sample, leading to either detuning/attenuation of a resonant peak or affecting the sensors propagation constant/characteristic impedance. In the case of [10]–[15], [18], and so on, this is only a fraction of the total field, hence lowering the measurement sensitivity. Out of the sensors devoted for liquid samples’ characterization, [10], [11], [14]–[19], and so on, the sensor proposed in [16] realized as an open-ended two-wire coplanar strip (CPS) transmission line can yield high sensitivity as the liquid-under-test (LUT) constitutes fully the dielectric medium given that the liquid volume is large enough. Following, the sensor in [17] realized as a broadside-coupled-line section with a liquid channel in-between can enforce full-field enclosure within a small volume of a liquid sample. However, a four-port measurement is necessary providing even-/odd-mode excitation. The above mentioned sensors are of broadband type, similarly as in [11] or [18] where an interferometric configuration with an additional reference sensor is used to extract the information about the measured samples. Finally, in many cases, it is required to calibrate the sensing setup that includes the sensor beforehand, usually with the use of well-known liquid standards as in [10]–[14], [16], [18], and [19], to establish model parameters of the relationship between measured data and quantity of interest.

In this article, we propose a novel robust approach for liquid chemicals’ complex permittivity measurements in a wide
frequency range. A transmission-type sensor is proposed, comprised of a two-wire transmission line (TWL) enclosed within a 3-D-printed liquid chamber that is to be filled with LUT. Such a configuration ensures high sensitivity as to when LUT is present, it fully constitutes the dielectric medium for the TWL. Therefore, the LUT’s complex permittivity can be derived directly from the TWL’s propagation constant assuming it being nonmagnetic (relative magnetic permeability μᵣ = 1). As such, the sensor can be of arbitrary impedance, and thus there is no need to know its geometry except for physical length, which ensures its robustness. Importantly, no system calibration is needed as the relation between complex permittivity and the measured sensors’ S-parameters is described through a simple analytical formula. To enable convenient physical realization and sensor excitation, a broadband microstrip (MS) to CPS to two-wire transition is used where the CPS geometry matches roughly the TWL geometry. Along, the Thru-Line de-embedding technique is adopted in a simplified form to deliver the sensor’s propagation constant treating the setup as the LINE standard, and thus only one additional THRU standard is needed. An exemplary measurement setup was developed and verified up to 18 GHz. The measurement results for a set of alcohols yielding useful data within the transition bandwidth with provided measurement error prove the usability of the proposed approach. It is shown that after postprocessing of the measured data, the frequency range of the measured permittivity is not limited to one of the transitions and thus certifying the broadband operation.

II. DERIVATION OF LIQUID PERMITTIVITY BASED ON SENSOR’S MODEL

A schematic view of the proposed microwave-fluidic measurement setup is shown in Fig. 1. The key component is a highly sensitive sensing element realized as a TWL section encapsulated in a liquid chamber as shown in Fig. 2. A detailed description of the setup is provided in the following sections.

A. LUT Complex Permittivity Determination

A TWL sensor model depicted in Fig. 2(a) (cross section) is consisted of two conductors having a circular cross section of diameter dᵣTW that are spaced by DᵣTW (between centers) and

\[
\gamma_{TW} = \sqrt{\frac{(R + j\omega L)(G + j\omega C)}{\sqrt{(R + j\omega L)^2 + (G + j\omega C)^2}}} \quad (1)
\]

\[
Z_{TW} = \frac{R}{\sqrt{(R + j\omega L)^2 + (G + j\omega C)^2}} \quad (2)
\]

where the resistance, inductance, conductance, capacitance (RLGC) are per-unit-length parameters of the lossy transmission line model that reflect the transmission line’s geometry and the properties of the propagation medium. Therefore, having measured (1) and/or (2), one can determine the effective permittivity of the medium. Moreover, if the medium is assumed a pure dielectric, knowledge of only one of the two parameters is sufficient. Considering the application, however, the propagation constant is more favorable since it can be easily delivered when the sensor is embedded into a sensing setup. This technique is covered in detail in Section II-B.

The relationship of interest can be derived when plugging RLGC parameters for a TWL [21] into (1) yielding

\[
\gamma_{TW} = j \sqrt{\left(\varepsilon_{TW}^\prime - j\varepsilon_{TW}''\right)\left(\frac{\omega^2}{c_0^2} - j\omega\varepsilon_0\right)} \quad (3a)
\]

\[
W = R_S \frac{D_{TW}}{0.5d_{TW}\cos^{-1}\left(\frac{D_{TW}}{\pi d_{TW}}\right)} \sqrt{D_{TW}^2 - d_{TW}^2} \quad (3b)
\]

where W is a variable that bounds the conductor resistance Rₛ and wire geometry; ω is the angular frequency, ε₀ is free space permittivity; c₀ is free space velocity. After some algebraic manipulations, the relative complex permittivity εₜₜ′ can be written in terms of γₜₜ as

\[
\varepsilon_{TW}^\prime = \left(\frac{-j\gamma_{TW}}{\frac{\omega}{c_0} - j\omega\varepsilon_0}\right)^2 \overset{\gamma_{TW} \approx 0}{\rightarrow} \left(\frac{-j\gamma_{TW}}{\frac{\omega}{c_0}}\right)^2 \quad (4)
\]

The reduced formula is valid with little error for very good conductors such as copper. Importantly though, the...
information on the TWL sensor cross section geometry is no longer needed.

Following, the proposed construction of the sensor [see Fig. 1(a)] including the liquid chamber provides two main advantages. The first being a theoretically unrestricted band of operation due to transmission line nature with TEM mode of propagation and the second being provision of high sensitivity defined as a fraction by which the LUT affects the sensed effective permittivity. When the liquid chamber is properly sized, the sensitivity reaches unity as the TWL’s conductors are within a homogenous medium constituted solely by the LUT and the electrical field associated with the propagating wave along the line, encloses entirely within this medium. Therefore, the permittivity of LUT is measured directly in the setup through (4) as

\[ \varepsilon_{\text{LUT}} = \varepsilon_{\text{TW}}, \]

(5)

### B. Sensor Parameter De-Embedding

The proposed sensor is intended for use in a two-port transmission configuration in which the propagation constant is directly associated with the transmission coefficient. Therefore, no additional calibration steps are needed as in open/short-ended reflection configuration (see [15]). Nevertheless, the sensor needs to be embedded in a fixture that serves as a mechanical frame and electrical transition to a more convenient guiding structure from the measurement perspective. The full setup model is shown in Fig. 1(a). The raw S-parameters that are measured are referenced at ports #1 and #2, and therefore de-embedding is required. A Thru-Line de-embedding technique from [22] in a simplified form can be adopted for this purpose. In [22], two standards are needed on top of the embedded device-under-test, namely thru and line to deembed the device-under-test with a byproduct of the algorithm being the line’s propagation constant. Since that is the only sensor’s parameter of interest, the embedded sensor is now proposed to be considered as LINE and only an extra THRU standard is required [see Fig. 1(b)]. This is beneficial as neither multi-standard de-embedding nor full two-port S parameters of the sensor are needed, and hence, the measurement error due to manufacturing repeatability is reduced.

To derive the required \( \gamma_{\text{TW}} \) information, first, the matrix \( Q \) is calculated

\[ Q = M_L M_T^{-1} \]

(6a)

\[ M_T = T_X T_Y \]

(6b)

\[ M_L = T_X T_{\text{sens}} T_Y \]

(6c)

\[ T_{\text{sens}} = \begin{bmatrix} e^{i \gamma_{\text{TW}l_{\text{TW}}}} & 0 \\ 0 & e^{i \gamma_{\text{TW}l_{\text{TW}}}} \end{bmatrix} \]

(6d)

where \( T \) presents the wave cascaded transfer matrix, \( M_T \) and \( M_L \) are the measured THRU and LINE T-parameters. The \( T_X \) and \( T_Y \) are assumed to describe identical transitions with opposite orientation and include the impedance transformation matrix, while \( T_{\text{sens}} \) describes the sensor. Then, knowing that similar matrices have identical eigenvalues, the complex propagation constant of the sensor can be calculated as

\[ \gamma_{\text{TW}} = \frac{\ln(\text{eig}(Q))}{l_{\text{TW}}} \]

(7)

where \( l_{\text{TW}} \) is the physical length of the TWL. Finally, the phase unwrapping algorithm needs to be applied to the imaginary part of \( \gamma_{\text{TW}} \), as (7) provides it in the wrapped form, to deliver correctly the \( \varepsilon_{\text{LUT}} \). It is worth noting that \( T_{\text{sens}} \) is impedance invariant and thus the determination of propagation constant does not depend on the sensor’s characteristic impedance making the approach very robust.

### III. Measurement Setup Design

For the sake of experimental verification of the proposed technique, an exemplary sensing setup was developed. The setup is assumed to be a general-purpose one, operating at least within the 1–10-GHz frequency band (assuming 50-\( \Omega \) impedance bandwidth of transitions with sensor exposed to air). Importantly, there is no bandwidth limitation for the applicability of the proposed technique in general. The limitation comes from the design of the transition, the measurement system dynamic range, uncertainty, and noise.

#### A. TWL Sensor

The two-wire sensor is a pair of wires in close proximity. For practical reasons, it is better to restrain the minimal distance between wires to \( D_{\text{TL}_{\text{min}}} = 1 \text{ mm} \), which ensures reliability and minimal wire diameter to \( d_{\text{TL}_{\text{min}}} = 0.5 \text{ mm} \) as it provides enough stiffness (especially if the flow system would be tested). \( D_{\text{TL}}/d_{\text{TW}} = 2 \) yields roughly \( Z_{\text{TW}} = 158 \Omega \) in air and drops when permittivity increases. On the other hand, the closer the wires together, the more confined the field associated with the propagating wave can be. This is important from the perspective of designing the liquid container, which will be explained later. However, if the LUT is a heterogeneous solution with particle/fractions of uneven distribution and relatively large size, the more confined field might lead to results deviating from the large volume average as the LUT permittivity is sensed by the sensor in a region of the field cross-sectional distribution (which depends on the wire geometry and medium permittivity). This can be perceived as the result is an average of the material properties within the volume limited by the EM field cross-sectional distribution times the sensor length. The sensor’s electrical length should be set to at least \( 20\degree \) at the lowest frequency of operation to minimize the measurement errors. This is derived to provide a given minimum signal-to-noise ratio thus providing a clean signal as well as to be high enough above the intended measurement instrument resolution and uncertainty. In general, however, the longer the sensor, the more robust the measurement and the less prone to some of the error sources at the expense of reducing the upper-frequency limit resulting from a finite dynamic range of the instrument used.

The sensor’s geometry was determined as a compromise to all the above-discussed aspects. Wires having a diameter of \( d_{\text{TW}} = 0.8 \text{ mm} \) and length of \( l_{\text{TW}} = 15 \text{ mm} \) were spaced by \( D_{\text{TL}_{\text{min}}} = 1.5 \text{ mm} \). The chosen geometry yields \( Z_{\text{TW}} = 150 \Omega \) and \( 18\degree \) @ 1 GHz, both in air. A silver-clad copper core wire was used as it features a very good conductance (\( \sigma_C = 6.3\times10^7 \text{ S/m} \) for silver at 20 °C), and therefore, the term \( W \) in (4) can be neglected with small added error (e.g., for ethanol \( \text{adderr}_W(\varepsilon_{\text{LUT_ethanol}}) \approx 0.4\% \) for Re...
and \( adderr \approx 0.9\% @ 5 \text{ GHz} \). It must be noted, the printed circuit board (PCB) fabrication tolerances error usually has a greater effect than neglecting \( W \).

### B. MS to Coplanar Strip to Two-Wire Transition

Once the geometry and impedance of the TWL sensor are established, the requirements for the transition design can be defined. The TWL sensor, due to its construction needs to be hosted in an accompanying setup, and to do so a PCB technology can be used. To accept the TWL onto PCB, a CPS transmission line can be used which features similar geometry of two co-planarly located conductors. On the other hand, coaxial type cables are usually used in the measurement setup, which can be easily transitioned into a MS transmission line on PCB. Therefore, to connect the above two, an MS to CPS transition is needed. Such a transition provides not only a smooth transition of the TEM mode of propagation from one type of the TL to another one but also can provide impedance transformation from the measurement system characteristic impedance of 50 \( \Omega \) to the one of the TWL sensor (when those two differ as in this case due to the reasons discussed in Section II-B). The CPW geometry on PCB should closely match one of the TWL to ensure a smooth transition as well.

Since the propagation constant of the sensor is extracted with the use of one extra THRU standard, the selection of laminate for circuit manufacturing is an open choice as the transitions are de-embedded from measurements. Here, a 0.508-mm-thick Arlon 25N laminate having a permittivity of 3.38 and loss tangent of 0.003 was selected for the design. A 0.508-mm-thick Arlon 25N laminate having a permittivity of 3.38 and loss tangent of 0.003 was selected for the design. The container wall thickness was set at 0.8 mm in the TWL to taper toward forming a cylindrical liquid port having an outer radius of 8.6 mm to accommodate a push-on quick coupler. Such dimensions ensure, with a safety margin, that the measured permittivity is an effective permittivity of the LUT and the container wall material and thus increasing the measurement error. Such behavior would be most pronounced for low permittivity LUTs as the EM field tends to “spread” more with decreasing permittivity.

The final shape of the attachment is shown in Fig. 5(a), which is suitable for both liquid-static and liquid-flow applications. For the sake of this study, an oversized chamber was designed. The inner length of the chamber along the TW sensor is equal to the sensor’s length while the inner width perpendicular to the sensor was set as 22 mm. The height of each half was set at 10 mm before the container starts to taper toward forming a cylindrical liquid port having an outer radius of 8.6 mm to accommodate a push-on quick coupler. Such dimensions ensure, with a safety margin, that the entire field encloses within the LUT medium (see Fig. 4). The container wall thickness was set at 0.8 mm in the TWL region to minimize the edge wall effect, i.e., print material affection on the lines, while everywhere else it was equal to 2.4 mm. To fill the vertical part of the container, approximately 5 mL of LUT is required.
D. Fabrication and Assembly

The designed sensor and transitions were integrated into the final LINE and THRU PCBs, which were manufactured in-house out of the same sheet of laminate through the photolithography process and chemical etching. The TW sensor was mounted by soldering wires ends to the respective copper trace on PCB in the CPS-to-TW transition region. The liquid chamber was printed using Prusa i3 MK3s FDM (Fused Deposition Modeling) type 3-D printer out of PET-G (Poly (ethylene terephthalateco-1, 4-cyclohexylenedimethylene terephthalate)) material ($\varepsilon_r = 2.4$, $\tan\delta = 0.02$ @ 1 MHz) which has a stable chemical resistance against the intended test liquids [26]. Along with the container, an accompanying part for the THRU standard [see Fig. 5(b)] was fabricated. The top and bottom printed parts were attached to the PCB using adhesive and screws for leak-proofing and mechanical sturdiness. Printed bars were added on PCB sides for extra stiffness. The THRU PCB was assembled in a way to enable de-embedding solely the TW sensor-related propagation constant, i.e., short wires were soldered to compensate for the CPS-to-TW transitions wires influence while the printed parts were attached using the same epoxy glue to compensate for the liquid chamber end walls material and glue influence. Photographs of the fully assembled sensing setup are shown in Fig. 6.

IV. EXPERIMENTAL RESULTS

The developed sensing setup was experimentally verified by characterizing a set of test liquids. A series of measurements were taken for the following LUTs: methanol, ethanol, isopropanol, and butanol as those are commonly available and well-characterized liquids. All $S$-parameters were done using the Agilent N5224A PNA Microwave Network Analyzer covering a bandwidth of 10 MHz–18 GHz at 1800 points, IF filter bandwidth of 1 kHz, no averaging. The PNA was calibrated using the Agilent 85052D 3.5-mm standard mechanical SOLT calibration kit setting the reference plane at the edge of SMA connectors on the PCB. Prior to measurements, all liquid samples had settled to room temperature of 21°C ± 1°C, which was maintained in the lab. For each measurement, the LUT sample was poured into the liquid chamber of the TWL sensor and the liquid-static measurement was carried out. After each sample, the chamber was thoroughly drained and dried by blowing compressed air.

First, the THRU standard was measured. Then, a set of the LINE measurements were carried out with the TWL sensor’s liquid chamber filled with the test liquids. The collected $S$-parameters for both are provided in Fig. 7. It is seen from THRU measurement that the transitions operate within the designed bandwidth. Outside, strong reflection/attenuation is observed in the vicinity of 0.5, 11, and 14 GHz, hence increased measurement uncertainty is expected. Lower than predicted impedance match above 2 GHz and extra reflections and ripples on the transmission can be attributed to finite fabrication tolerances and assembly accuracy along with epoxy adhered (not EM modeled) 3-D printed attachment on top. Ports mismatch is becoming visible on $S$-parameters above 10 GHz due to the same reasons. It is seen from LINE measurements, that for a general-purpose sensor as the one used here, the higher complex permittivity, the higher noise is observed due to the high dynamic range of the measured
Fig. 8. Measurement derived permittivity (solid lines) with overlaid reference data (dashed lines) of (a) methanol, (b) ethanol, (c) isopropanol, and (d) butanol at 21 °C. Raw data postprocessing applied.

S-parameters. This also relates to the length of the TWL itself as longer would improve sub-GHz performance while shorter could provide signal not exceeding the instrument dynamic range at higher frequencies. As a reference, the TWL sensor de-embedded from LINE reaches phase shift $\hat{\gamma}_{\text{LUT TW}}$ of 20° at 0.19, 0.22, 0.34, and 0.28 GHz for methanol, ethanol, butanol, and isopropanol, respectively.

The derived nominal complex permittivity of each tested LUT, using (4), is provided in Fig. 8. Calculated data was postprocessed for noise reduction and smoothing (outlier removal and Savitzky-Golay filter) to extend their usable frequency range. Moreover, the calculated permittivity was referenced against the one provided in [27] (relaxation model-based data was used (1)–(4) [27]).

It is worth noting the proposed technique provides data over a very broad frequency range, thus it is well suited for fitting a permittivity model being useful for liquids and liquid mixtures properties’ analysis. Moreover, such a model gives more robust information than the raw measurement which can be loaded with noise or ripples (e.g., as here outside the MS-to-CPS-to-TW transition bandwidth). Therefore, the measured permittivity of each LUT was fitted (using nonlinear least-squares solver) with the same model as used in [27] allowing for direct comparison of coefficients as in Table I and curves as overlayed in Fig. 8. It is seen that the obtained results align well with the reference data (except for regions of strong signal reflection/attenuation) validating the proposed technique. It is seen, however, that for liquids having first relaxation frequency far below the operation band of the used transition, the fitted coefficients deviate more from the reference and thus transitions with lower cutoff would be recommended to improve that. A detailed error analysis is provided in Section V.

| Liquid  | Temp. °C | Model | Coeffs’ | Uncert. | Coeffs’ Fitt freq. Resnorm |
|---------|----------|-------|---------|---------|--------------------------|
| Methanol| 21       | Single-Debye | $\varepsilon$ | 33.44      | 0.06                  | 53.58 | 966 |
|        |          |       | $\varepsilon_0$ | 5.636      | 0.118              | 5.574 |       |
|        |          |       | $\varepsilon_0$ | 2.886      | 0.022              | 2.793 |       |
|        |          |       | $\varepsilon_0$ | 25.01      | 0.04               | 24.81 |       |
| Ethanol| 21       | Debye-Gamma | $\varepsilon$ | 4.526      | 0.020              | 4.825 | 48.6 |
|        |          |       | $\varepsilon_0$ | 0.856      | 0.003              | 0.907 |       |
|        |          |       | $\varepsilon_0$ | 0.058      | 0.006              | 0.062 |       |
|        |          |       | $\varepsilon_0$ | 19.95      | 0.08               | 18.34 |       |
|        |          |       | $\varepsilon_0$ | 3.556      | 0.015              | 3.635 |       |
| Isopropanol| 21     | Double-Debye | $\varepsilon$ | 3.059      | 0.044              | 3.098 | 71.5 |
|        |          |       | $\varepsilon_0$ | 0.369      | 0.002              | 0.318 |       |
|        |          |       | $\varepsilon_0$ | 5.777      | 0.062              | 5.500 |       |
|        |          |       | $\varepsilon_0$ | 18.09      | 0.04               | 17.50 |       |
|        |          |       | $\varepsilon_0$ | 3.505      | 0.011              | 3.601 |       |
|        |          |       | $\varepsilon_0$ | 2.904      | 0.031              | 2.909 |       |
|        |          |       | $\varepsilon_0$ | 0.269      | 0.001              | 0.206 |       |
|        |          |       | $\varepsilon_0$ | 5.888      | 0.505              | 5.500 |       |

Coefficient fitted to data measured over 0.03 GHz to 5 GHz. Interpolated for unspecific temperature.

V. ERROR ANALYSIS AND PERFORMANCE ASSESSMENT

To assess the performance of the proposed measurement technique, the associated measurement error was studied. The main sources that contribute to the total error were identified, based on the setup’s model as follows:

1) uncertainty of S-parameters measurement using vector network analyzer (VNA);
2) systematic error due to sensor’s length uncertainty due to finite sensing setup fabrication and assembly tolerances;
3) systematic error due to nonidentical transitions being de-embedded resulting from finite PCB fabrication and assembly tolerances;
4) an error due to neglecting the conductor-related losses being insignificant for high loss dielectrics;
5) an error due to leak-proofing limitations of the fluidic channel and possible seepage onto the CPS-to-TW transition;
6) uncertainty of liquid sample temperature determination.

The above-listed sources of error provide insight into what are the key contributors and indicates the direction, how to minimize the overall measurement error. A detailed study is provided in the following section.

A. Measurement Uncertainty

First, the measurement error was assessed based on the sensing setup model proposed in Section II. On top of the VNA manufacturer-specified S-parameters’ measurement uncertainty $\delta_{\text{params}}$, the MS-to-CPS-to-TW transitions propagation constant uncertainty of $3\delta_{\text{manuf}} = 0.1 \, \text{dB} + 0.5^\circ (\alpha l + \beta l)$ (due to PCB fabrication tolerance, a spill of leak-proofing adhesive), and sensor’s length uncertainty of $3\delta_{\text{eng}} = 0.2 \, \text{mm}$ was incorporated to derive the worst-case scenario. A combined standard uncertainty was calculated through the type A evaluation of uncertainty based on the measured nominal value of permittivity for each measured LUT and is provided in Fig. 9 as normalized-to-mean $\delta/\mu$ value. Regions of increased uncertainty align with the frequency regions of increased noise; thus, the focus is put on the 1.5–10-GHz region along with 15–18 GHz.

The strongest impact is observed from the S-parameter measurement uncertainty contributing to the measured permittivity error (understood as $3\delta/\mu \cdot 100\%$) being less than 0.5\% for the real part and less than 1.8\% for the imaginary part from 2 GHz upward. Importantly, however, this contribution can be easily reduced at the expense of the increased measurement time by narrower IF filter and/or data averaging. The sensor’s length uncertainty increases the error up to 2.4\% for the real part and to be less than 4.2\% for the imaginary part. This error is of systematic nature and can be reduced by accurate length measurement after assembly or through tighter manufacturing and assembly tolerances. The same applies to the transitions mismatch for de-embedding.

B. Repeatability and Reproducibility

The repeatability and reproducibility of measurements were studied on an example of ethanol following the general measurement procedure. This was done to assess the data variation due to procedure repeatability as multiple pouring – single sensor unit and due to sensors fabrication reproducibility as single pouring – multiple sensor units.

The repeatability of data using the same sensor unit was investigated with a total of six measurement sequences. For each sequence, the LUT sample was poured into the liquid chamber of the TW sensor and the steady-liquid type measurement was carried. After each sequence, the chamber was thoroughly drained and dried by blowing compressed air. Having collected the data (eight traces per sequence, averaged value of those used), the intersequence mean value $\mu_{\text{inter seq}}$ and standard deviation $\sigma_{\text{inter seq}}$ were calculated to assess the repeatability. The above was followed by the study of the reproducibility of data for which four sensor units were fabricated and single-sequence measurements using each of them were carried out. Then the interunit mean value $\mu_{\text{inter unit}}$ and standard deviation $\sigma_{\text{inter unit}}$ were calculated out of the collected data to assess general reproducibility. The study is summarized in Fig. 10 in a form of mean-value-normalized deviation $\bar{\sigma} = \sigma/\mu$.

It is seen from the intersequence deviation that the measured permittivity for ethanol is repeatable with the deviation being less than 0.002 for the real part and less than 0.012 for the imaginary part. The behavior aligns with the results of uncertainty analysis with the addition of potential sequence-to-sequence liquid temperature variation that affects its permittivity. Finally, it is seen from the interunit deviation that the measured permittivity for ethanol is reproducible with the deviation being less than 0.030 for the real part and less than 0.041 for the imaginary part. This is higher than repeatability for a single unit which is expected, even though certifies the robustness of the proposed approach. It is worth noting that the interunit reproducibility reflects the in-house fabrication tolerances, especially when it comes to milling and straightness of the soldered wires, and thus is expected to improve when fabricated by a professional PCB/assembly service.

C. Accuracy With Respect to Reference

Subsequently, the measurement accuracy with respect to the reference data was studied. An absolute percentage error between the nominal measured and nominal reference permittivity [27] was calculated for each LUT and is provided in Fig. 11. It is seen that accuracy over frequency aligns with the measurement uncertainty as discussed earlier.
TABLE II
STATE-OF-THE-ART TECHNIQUES FOR CHARACTERIZATION OF LIQUID DIELECTRIC MATERIALS

| Shown range of operation [GHz] | Sensor type | Technique type | Sensitivity | Effective permittivity equal to LUT | Calibration liquid required | LUT volume required | Measured liquids | Permittivity extraction |
|-------------------------------|-------------|----------------|-------------|-------------------------------------|-----------------------------|---------------------|---------------------|------------------------|
| [10] 17.9 – 19.34             | Coplanar resonator | Resonant | Medium | No | No | Small* | Water-ethanol mixture | Model-based |
| [11] 1.25 – 1.6               | Complimentary Split Ring Resonator | Resonant | High | No | Yes | Small* | Water-methanol mixture | Direct |
| [13] 0.01 – 3                 | Open-ended coupled-line section | Transmission / Reflection | Medium | No | Yes | Large | Isopropanol, ethanol, butanol | EM sim.-based |
| [14] 4 – 8                    | Composite right/left-handed transmission lines | Transmission / Reflection | Medium | No | Yes | Small* | Ethanol-methanol mixture, butanol-propanol mixture | Model-based |
| [15] 0.1 – 110                | Coplanar waveguide | Transmission / Reflection | Medium | No | Yes / air | Very small* | Water | Model-based |
| [16] 0.01 – 10                | Substrate-less coplanar stripline | Transmission / Reflection | High | Yes | Yes | Large | Butanol, propanol, isopropanol | Direct from $\gamma$ and $Z$ |
| [17] 0.01 – 4                 | Stripline coupled-line section | Transmission / Reflection | Medium | No | No | Small* | Methanol, ethanol, propanol, isopropanol, butanol | Model-based |
| [18] 1.97                     | Folded microstrip line | Transmission / Reflection | High | No | Yes | Small* | Isopropanol-water mixture | Model-based |
| [19] 0.5 – 50                 | Open-ended coaxial probe | Transmission / Reflection | Medium | Yes | Yes | Medium | - | Model-based |
| This paper                   | Two-wire line | Transmission / Reflection | High | Yes | No | Small* | Methanol, ethanol, isopropanol, butanol | Direct from $\gamma$ |

* microfluidic channel
* sub-mm channel poses an issue for lower permittivity liquids (only high permittivity water shown)
* single/sub-milifluidic channel

Fig. 11. Calculated absolute percentage error between nominal measured complex permittivity and the nominal reference value from [19] for methanol, ethanol, isopropanol, and butanol as LUT. Error for (a) real and (b) imaginary parts.

The resonant techniques despite their high sensitivity and small LUT volume such as [10] and [11] are not suited for BDS applications due to narrowband operation at discrete points. On top of that, the above requires either calibration or a detailed sensor model to extract LUT’s permittivity. On the other hand, a large variety of sensors has been developed relying on the transmission/reflection measurement principle providing broadband information. Such sensors can feature high sensitivity as well when LUT constitutes fully the

D. Performance With Respect to State-of-the-Art

Finally, the proposed measurement technique was compared with state-of-the-art to analyze its advantages and limitations. The summary is presented in Table II.

It is important to note that the uncertainty of the reference data being in the range of 0.15%–1.5% of the nominal value in the range of 0.1–5 GHz is lower for the real part and higher for the imaginary part of complex permittivity. Moreover, it is increasing with frequency and decreasing with increased permittivity value. Similar behavior is observed for

the determined measurement accuracy error. Interestingly, for the real part, the error tends to increase with decreasing permittivity. Higher error for lower permittivity LUTs can be attributed to the potential 3-D-printed container edge-wall effects which are not considered in the sensing setup model. A small fraction of the field might be enclosed either within the chamber edge-wall material (here PET-G) and the substrate as the EM field transitions from one type of wave guiding structure to another or more likely within an excess of epoxy adhesive that could have spilled onto the TW sensor during assembly. Such effects can be more pronounced for low permittivity LUTs or relatively short length sensors or high-permittivity chamber material or epoxy. Finally, LUT heating due to exposure to an alternating electromagnetic field of the TW sensor as an effect of molecular dipole rotation within the dielectric material is not considered. This effect is more pronounced for the imaginary part of permittivity than for the real part. However, the increase in temperature is expected to be lower than the uncertainty of the used temperature measurement instrument.
dielectric medium as in [16] or in this work. Moreover, in that case, LUT permittivity can be extracted directly and only from the measured propagation constant for the purpose of which extra TRHU is used while in [16] the propagation constant, characteristic impedance, and open-end capacitance is required to be established. The above is not a case in [14], [15], [17], and [18], etc., where LUT’s permittivity is only a fraction of the sensed effective permittivity or enclosed by the fringe field as in [19], thus lower sensitivity, and extraction of which is done through the established medium model and/or calibration data. Even though microfluidic sensors are shown in [14], [15], [17], or [18], the one proposed here might operate with single-milliliters volume. The advantage here might be the 3-D printed chamber, which apart from minimal dimensions can be of almost arbitrary shape for better integration with the liquid delivery part of the system. Finally, the frequency range of operation and/or permittivity range poses a requirement on sensor properties to fit the dynamic range. An example is a miniaturized sensor as in [15] enabling operation up to mm-wave range at the expense of being suitable only for very high permittivity liquids. In [11] and [15], the presented sensors were validated only with high-permittivity solutions, whereas in [13], [14], [16], and [17] sensors were shown with measurements of LUTs featuring lower permittivity so their performance for significantly lower/higher permittivity is not assessed. In this study, liquids with a large span of permittivity have been tested. The measurement error can be compared to one of [17] as the same liquids were tested, and no calibration liquids were used. The proposed technique yields lower error with a much broader range of operation. Error comparison with [13]–[16], [18], and [19] might not be adequate as in those a calibration procedure is needed, as on the contrary to this work where only de-embedding is used. On the other hand, lower or comparable measurement uncertainty as in [15] is obtained. Considering the uncertainty of the measured and reference data along with LUT temperature uncertainty and skipping the conductor losses on one hand and simplicity of the approach on the other, it can be stated that a satisfactory measurement accuracy can be obtained for many applications such as food processes monitoring, broadband materials characterization for mixing processes, microwave heating, etc.

VI. Conclusion

A novel measurement technique centered around a two-wire sensor was introduced to enable direct liquid permittivity measurement over a broad frequency range. For the proposed approach, neither knowledge of the TWL sensor geometry, except for length, nor the characteristic impedance is required as the calculation is based on the measured sensor’s propagation constant. The LUT permittivity is calculated directly under the assumption that the EM field associated with the sensor encloses fully within the LUT. The technique was experimentally verified by measurement. Importantly, the proposed technique has no theoretical frequency limitation. However, the bandwidth is limited by a particular design of the required transitions along with uncertainty and noise of the S-parameters measurement equipment. The obtained results aligned with the reference data. Moreover, error sources were identified and strategies to minimize them were described. It can be concluded that LUT complex permittivity measurements performed with the proposed setup have a relatively low measurement error in a broad frequency range thus, proving it as a valuable measurement technique for BDS applications.

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