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Original Article

Characterization of Alcohol Water Mixture Using Ensemble Machine Learning Method

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Abstract *- Material characterization is important to ensure the quality of composites. Traditional methods for assessing alcohol content involve intricate chemical processes. Complex permittivity measurement is a good method to characterize the composites. This paper focuses on binary polar liquid mixtures, particularly alcohol-water mixtures, essential in many industries. Characterizing dielectrics, such as polar liquids, is challenging due to their frequency dispersion. To address this challenge, the paper proposes an ensemble machine learning-based classification model that uses complex permittivity measurements and frequency to accurately identify the type and volume fraction of alcohol in aqueous solutions. This model offers an accuracy rate of 98.4% and can accommodate a measurement error of ±5.5%. This approach simplifies assessing aqueous alcohol solutions and can serve as a supporting tool for various measurement systems.*

Keywords *- Composites, Complex permittivity, Classification, Identification, Polar liquids.*

1. Introduction

A composite is a unique material combining two or more substances with significantly different chemical and physical properties. Polar liquid composites, composed of liquids with polar molecules, offer a broad spectrum of applications. This work specifically examines binary polar liquid composites consisting of two polar liquids. In particular, the focus is on Alcohol-Water Mixtures (AWM). Alcohols are widely used in many industries, including chemical, pharmaceutical, food, and beverage sectors. Regular monitoring of alcohol content in these mixtures is essential to ensure product quality [1-4]. The work examines the most commonly used alcohols like methanol, ethanol, propanol, and isopropyl alcohol.

These are highly soluble in water and have a wide range of uses. Traditional methods for identifying the type and concentration of alcohol include chemical and optical techniques. Chemical methods, such as titration, High-Performance Liquid Chromatography (HPLC) and Gas Chromatography (GC) involve the use of hazardous chemicals like potassium dichromate and concentrated sulfuric acid. These methods typically require around 30 minutes. They are complex, time-consuming and require skilled operators. They also pose serious health risks, such as respiratory issues, due to prolonged exposure to harmful chemicals [5, 6]. Optical methods like Raman spectroscopy and Infrared (IR) spectroscopy use lasers for analysis. Although these methods are generally safer than chemical techniques, proper safety measures are essential due to the risks associated with laser usage [7]. Complex permittivity measurements employing microwave-based methods such as open-end coaxial probes and sensors are being considered as potential choices for identifying composite solutions. Coaxial probes are utilized for various frequencies, whereas sensors are only confined to a specific frequency range. However, the permittivity of polar liquids changes with frequency [8-14]. Identifying an unknown polar liquid requires measuring the complex permittivity and carefully examining the current standard report for near matches. In mixtures, this becomes even more complicated, as the composition of the mixture also influences the complex permittivity.

The study on techniques for material characterization of liquid mixtures identifies a significant research gap: the need for faster and safer methods for alcohol characterization. Machine Learning (ML) is a powerful tool for material characterization. It can analyze complex data and uncover patterns that traditional methods often miss. Machine Learning (ML) techniques are proposed to address these challenges and make the process more efficient. The work introduces a classification model based on an ensemble machine learning approach. It is designed to identify the type and concentration of alcohol in AWM. The model achieves this using a minimal set of relevant and readily available parameters. This Machine Learning (ML) model uses the real and imaginary parts of complex permittivity (ε' and ε'') along with frequency (f) as inputs to analyze mixtures. It determines both the type and alcohol content in aqueous solutions over a

frequency range of 0.2 - 20 GHz at 25°C, with an accuracy of 98.4%. Furthermore, Graphical User Interface (GUI) is developed to facilitate the application of the model. The proposed model has the novelty of achieving fast identification of AWM without any health risks.

2. Methodology

Machine learning plays a crucial role in categorizing data into various classes, and this work utilizes this capability through a classification-based Machine Learning (ML) model. The complete workflow is depicted in Figure 1.

The dataset is generated by measuring the complex permittivity (ε*) of AWM solutions using a dielectric probe kit - N1501A of Keysight Technologies. These measurements include both the real part (ε′) and the imaginary part (ε″) of the complex permittivity, as well as the frequency (f) at which the measurements are taken. The data collection is conducted at a constant temperature of 25°C across a frequency range of 0.2 to 20 GHz. This work focuses on four commonly used aliphatic alcohols: methanol, ethanol, propanol, and isopropyl alcohol. These alcohols are tested in various volume fractions within water: 25%, 50%, 75%, and 100%. For each volume fraction, 100 data points are measured across the specified frequency range, resulting in a total of 400 data points per alcohol type. Therefore, the entire dataset comprises 1600 data points representing four different mixtures. This dataset creates a multi-class classification problem with 16 distinct classes, named from 1 to 16, as detailed in Table 1. Each class corresponds to a specific combination of alcohol type and volume fraction. To train and evaluate the ML model, the dataset is split into two parts: 80% for training and 20% for testing. The training set is used to develop the model, while the testing set is employed to assess its performance.

A variety of classification-based ML algorithms are applied to train the model. Hyperparameter optimization is performed by fine-tuning the parameters to ensure optimal performance. The performance of ML models is evaluated using performance metrics such as accuracy, precision, sensitivity, and specificity. 5 fold cross-validation and external datasets are used to validate the model.

The stacking-based ensemble machine learning technique proved the most effective among the different approaches tested. This technique combines multiple learning algorithms to improve the overall accuracy and robustness of the model. The measurement setup used for dataset generation and the ensemble ML method used in the work are described in Sections 2.1 and 2.2

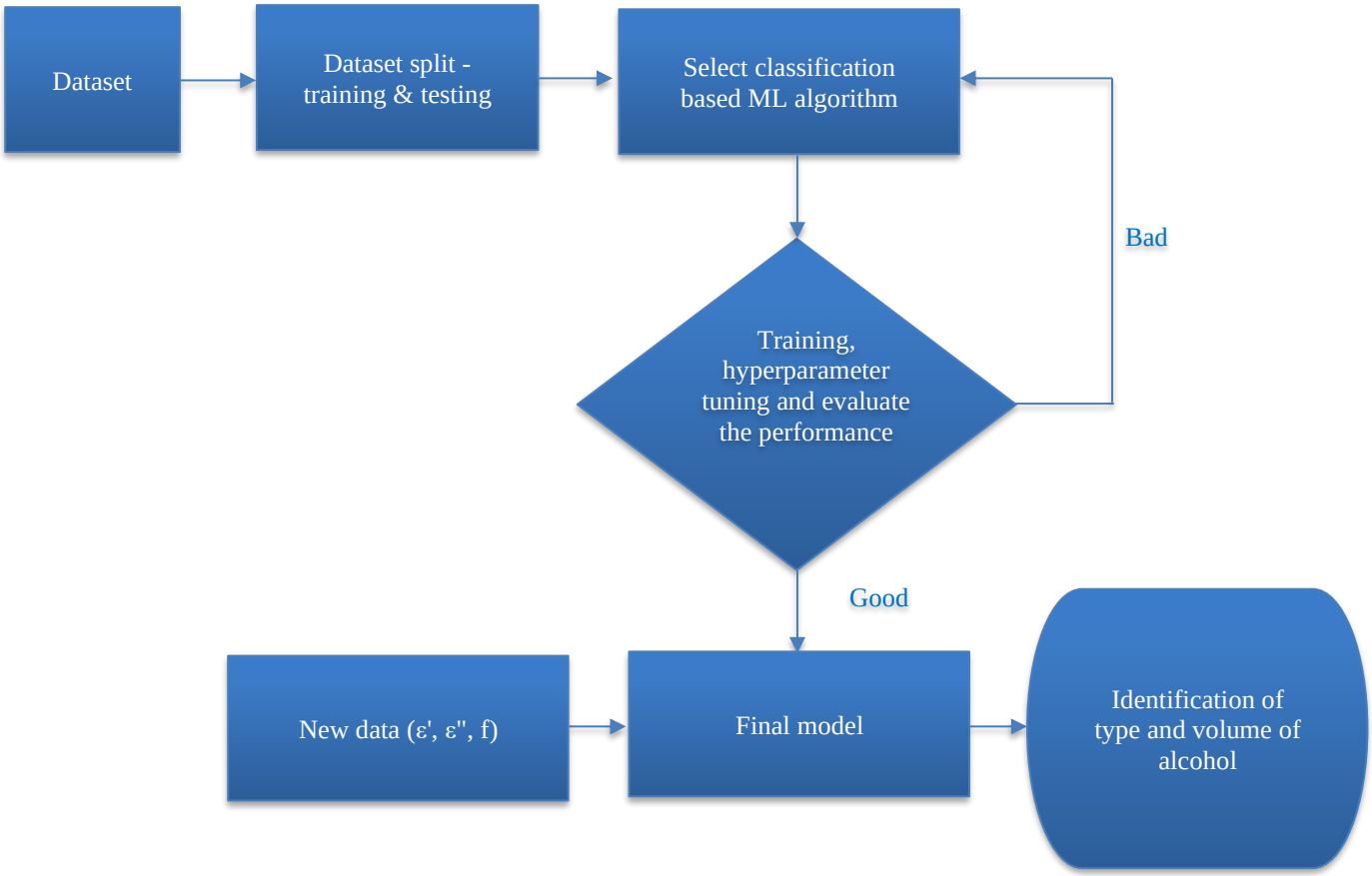


Fig. 1 Methodology

Table 1. Dataset formation

No	Inputs						
1	Frequency (f) in GHz						
2	Permittivity (Real part)						
3	Permittivity (Imaginary part)						
Output features							
No	**Liquid**	**Number of Samples**		**Volume Fraction of Alcohol (%) and Associated Class Assignment**			
				25%	**50%**	**75%**	**100%**
1	Methanol – water	400	**Class Assigned**	1	2	3	4
2	Ethanol – water	400		5	6	7	8
3	Propanol – water	400		9	10	11	12
4	Isopropyl alcohol – water	400		13	14	15	16

2.1. Measurement Setup

Measurement is carried out using a dielectric probe kit – N1501A of Keysight Technologies at a temperature of 25°C. Dielectric probe supports a broad frequency range. It provides high precision and enables rapid, non-destructive testing. The probe measures the reflected signal at the probe-material interface, calibrated to measure the complex permittivity. The device is user-friendly, works with small sample volumes and is versatile for various materials.

Additionally, it is safer than chemical methods and ensures efficient and accurate measurements. The measurement setup is shown in Figure 2. Calibration is a crucial step in ensuring the accuracy and reliability of measurement instruments.

Calibration of the probe is done using air, short and distilled water. Alcohol samples with a 99% purity level are procured from Alpha Chemicals, located in Cochin, Kerala, India. Aqueous solutions of alcohols are prepared by mixing them with distilled water.

AWM of 10 ml is prepared, and the volume fraction of alcohol used in this work is 25%, 50%, 75% and 100%. The probe is immersed in the 10 ml mixture solution so there is no air gap between the tip and the solution. The probe position is shown in Figure 3. The systematic measurement system ensures consistent and reliable data, reducing the necessity for preprocessing or addressing outliers. The noise that may occur is handled by error analysis, as explained in Secion.3.

2.2. Ensemble Machine Learning

Stacking is a sophisticated ensemble technique in machine learning designed to enhance predictive accuracy by utilizing the strengths of multiple base models. These base models can include various algorithms such as decision trees, Support Vector Machines (SVMs) or neural networks [15-19]. The process begins by training these base models on the same dataset. The predictions from these models are then utilized to generate new features, referred to as meta-features.

Fig. 2 Measurement setup

Fig. 3 Position of the probe in the mixture

These meta features serve as inputs for a meta-model, which is trained to effectively combine and weigh the base models' predictions.The primary function of the meta-model is to synthesize the information provided by the base models, often resulting in superior predictive performance compared to any single base model. Once the meta-model is trained, it can be applied to new unseen data, thereby improving the overall accuracy and robustness of the predictions [20-22].

In this work, the inherent nonlinearity in the data necessitates using nonlinear classification techniques. As base models, Support Vector Machines (SVMs) with polynomial and Radial Basis Function (RBF) kernels are employed.

SVMs are well suited for this task due to their ability to handle complex decision boundaries through kernel functions. The polynomial kernel can capture interactions up to a certain degree, while the RBF kernel can model even more intricate patterns by mapping the data into a higher-dimensional space.

Hyperparameters of each classifier are tuned to achieve optimal performance. For SVMs, this involves selecting the appropriate kernel parameters and regularization parameters. For the random forest meta classifier, the tuning process includes adjusting the number of trees (number of estimators), the depth of each tree and other relevant parameters [23-28].

The stacking approach effectively captures the complex relationships in the data by combining SVMs with different kernels as base models and utilizing a random forest as the meta-classifier. This methodology is versatile and can be extended to other mixtures by systematically extending the dataset.

3. Result and Analysis

The complex permittivity of an Alcohol-Water Mixture (AWM) varies with changes in both the frequency of the applied electromagnetic field and the volume fraction of the liquids within the mixture. This work utilizes the complex permittivity measurements obtained using a dielectric probe to characterize the AWM solutions accurately. To validate the measurement procedure, the complex permittivity values of polar liquids (ε^*) measured in this work are compared with the reference values documented for the frequency range 0.1 - 5 GHz in the National Physical Laboratory (NPL) report MAT 23 [13]. This comparison confirms the accuracy of the measurements, as the observed variations are within a range of ±1%, indicating a high degree of consistency with the established standards.

A sample plot of the complex permittivity of ethanol is presented in Figure 4, illustrating the agreement between the measured values and the reference data. To identify the type and volume fraction of alcohol in mixtures accurately, several classification-based machine learning techniques are applied to the dataset. These techniques are evaluated based on various performance metrics, including accuracy, error rate, specificity and sensitivity, as detailed in Table 2. The results indicate that among the single classifiers tested, the Support Vector Machine (SVM) with a polynomial kernel achieves an accuracy of 94.0%. In addition to single classifiers, ensemble techniques are employed to further enhance the model's performance. Among the ensemble techniques tested, stacking proved to be particularly effective. Table 3 presents the accuracy of the stacking classifiers, showing a significant improvement compared to the performance of individual classifiers.

Table 2. Performance comparison

Technique	Classifiers	Accuracy (%)	Precision (%)	Sensitivity (%)	Specificity (%)
Single classifier	Random Forest	82.1	83.2	81.3	82.0
	Decision Tree	83.4	84.1	83.1	83.9
	KNN	87.5	87.4	87.3	87.5
	SVM	94.0	94.1	93.8	94.1
Ensemble	Boosting	81.6	82.0	79.6	81.5
	Bagging	86.2	86.4	85.1	86.4
	Stacking	**98.4**	**98.2**	**98.5**	**99.7**

Table 3. Performance comparison of stacking classifier

Base Classifier	Meta Classifier	Accuracy (%)
SVM (kernel = RBF)	Decision tree	93.8
SVM (kernel = RBF)	Random forest	96.9
SVM (kernel = polynomial)	Decision tree	93.2
SVM (kernel = polynomial)	Random forest	97.8
SVM (kernel = polynomial) + SVM (kernel = RBF)	Decision tree	93.4
SVM (kernel = polynomial) + SVM (kernel = RBF)	Random forest	98.4

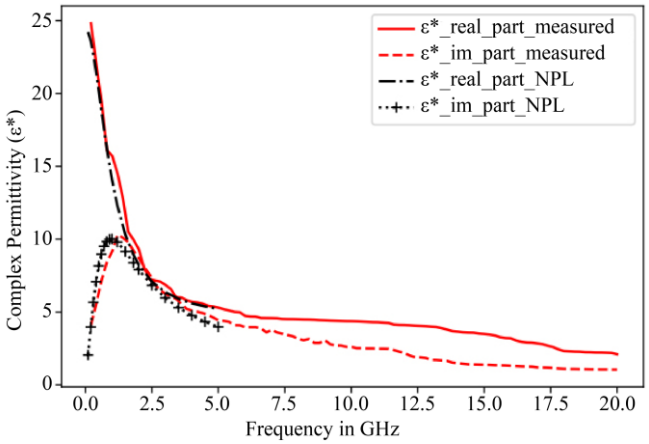


Fig. 4 Comparison of the measured value of complex permittivity (ε*) of ethanol with NPL data

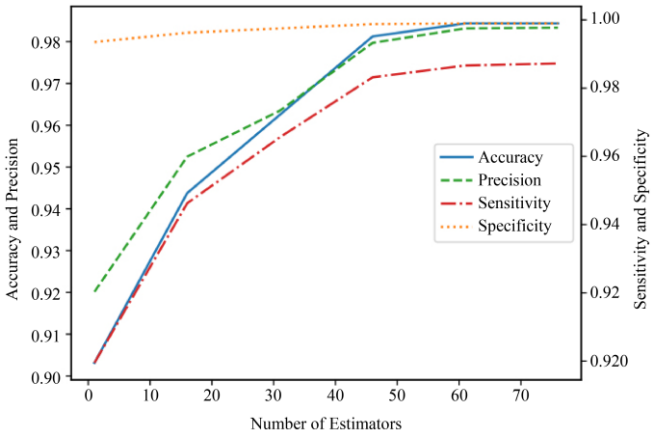


Fig. 5 Performance of stacking classifier

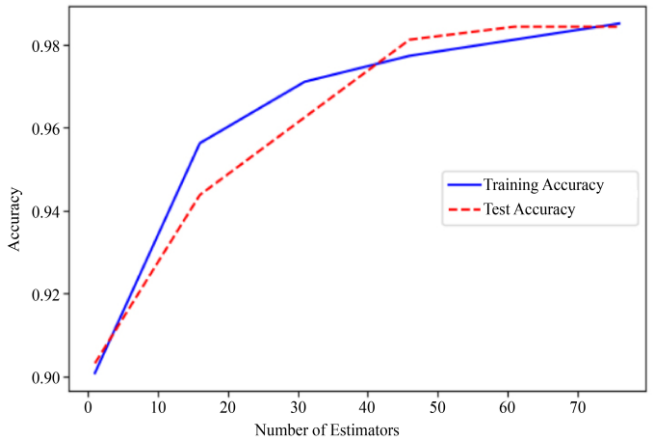


Fig. 6 Tuning of stacking classifier

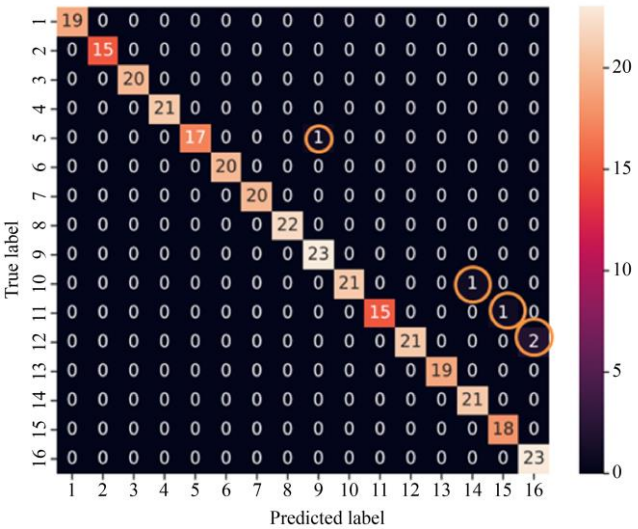


Fig. 7 Confusion matrix of test set

The characteristics of stacking employed in this work are as follows.

- The best performance is achieved by stacking two SVMs with kernel polynomial and Radial Basis Function (RBF) as a base classifier and random forest classifier as a meta classifier.
- K- fold cross validation with K = 5 is applied on the dataset to ensure the performance of the model.
- The hyperparameters of SVM and random forest are tuned to improve the performance.
- In the SVM with a polynomial as the kernel, the degree of the polynomial is tuned to 3, and in the SVM with RBF as the kernel, the penalty parameter C is tuned to 6.
- In the random forest classifier, the number of estimators is tuned to 70 to get the best result.
- The overall accuracy achieved in this case is 98.4%.
- All three input parameters (f, ε', and ε") are essential to achieving an accuracy of 98.4%. Removing any one of these parameters will negatively impact the performance of the model.

Figure 5 illustrates the performance metrics of the model. The model achieves maximum performance with 70 estimators attaining an accuracy of 0.984, precision of 0.982, sensitivity of 0.985 and specificity of 0.997. Figure 6 shows the performance of the proposed model on training and test sets. Since both training and test accuracy are comparable, overfitting is not observed. The confusion matrix for the test set is shown in Figure 7. The diagonal elements in the confusion matrix show the correct prediction, whereas the other elements show the misclassification. It shows that out of 320 elements, only 5 elements (encircled) are misclassified, resulting in an accuracy of 98.4%. To evaluate the robustness of the model, errors that may occur from measurement variations, temperature fluctuations and sample purity are systematically introduced into the dataset. These errors are incrementally added as steps of ±0.5% from the original measured values.

When the error exceeds ±5.5%, the accuracy of the proposed model drops to 97.1%. This is illustrated using a confusion matrix. Figures 7 and 8 present the confusion matrices for the model without any errors introduced and with errors, respectively.

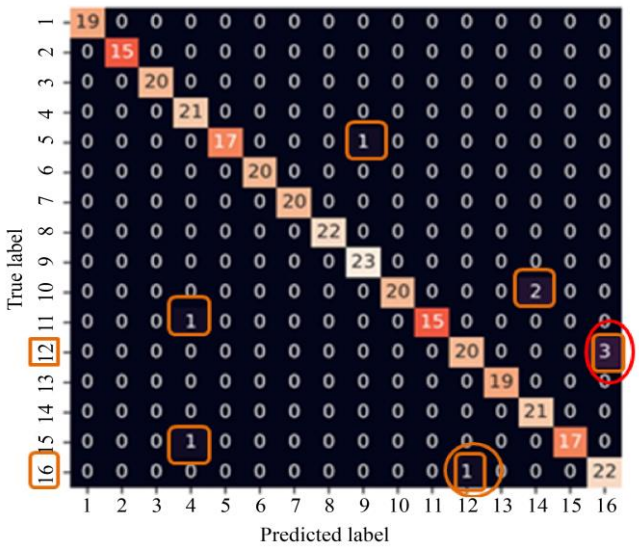


Fig. 8 Confusion matrix of the test set with the error of ±5.5%

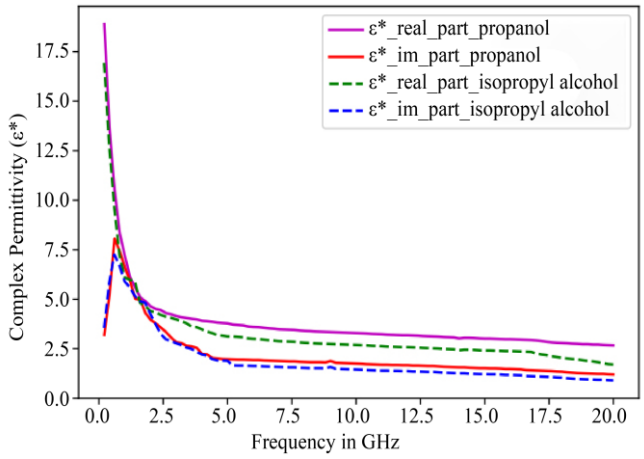


Fig. 9 Complex permittivity plot of propanol and isopropyl alcohol

When an error greater than ±5.5% is introduced, the number of misclassified elements increases to 9 (enclosed in the box inside the confusion matrix), reducing the accuracy to 97.1%. As depicted in Figure 8, these misclassifications predominantly occur between propanol and Isopropyl Alcohol (IPA) (classes 12 and 16). These classes are enclosed in the box outside the confusion matrix. The 4 misclassified elements in class 12 and class 16 are shown as encircled and boxed. To investigate this further, the complex permittivity of propanol and IPA is plotted in Figure 9, revealing that their values are particularly similar in the 0.2-3 GHz frequency range. These findings suggest that the model's accuracy is highly dependent on the precision of the permittivity measurements. This poses the limitation of the model. When detecting an unknown aqueous alcohol mixture using the proposed model, it is crucial to use well-calibrated instruments to measure complex permittivity with a measurement error margin of less than ±5.5%. This level of precision is necessary to minimize the likelihood of misclassification, particularly between substances with closely related permittivity values. Several techniques are available for measuring complex permittivity, including the coaxial probe, sensors and coaxial cell. The coaxial probe and sensors are the most frequently used among these methods due to their reliability and accuracy. Its performance is validated using a new test set to ensure that the proposed model can accurately follow these measurements. Complex permittivity values published in the scientific literature are gathered to create this new test set, resulting in a new test set of 30 samples in 8 classes. These values are deliberately chosen to highlight significant deviations from the measured data used in this work [2, 29]. This inclusion of values allows for a thorough assessment of the robustness of the model and accuracy.

The newly compiled dataset is used to validate the performance of the model. The results indicate that the model successfully identifies the complex permittivity values for 27 out of the 30 samples, achieving an accuracy rate of 90%. This high level of accuracy demonstrates the model's effectiveness in generalizing new data. A confusion matrix for the new test set is presented in Figure 10, providing a detailed view of the performance of the model across different classes. Additionally, the maximum permittivity error (E) between the measured values used in this work and the values reported in the literature is summarized in Table 4. The analysis reveals that most misclassifications occur when the error (E) exceeds ±5.5%.

The proposed approach serves as a valuable supplementary tool for complex permittivity measurement-based characterization. To facilitate the testing of unknown AWM, a Graphical User Interface (GUI) has been developed using the Flask framework [30]. This user-friendly interface simplifies the process, requiring only the measured values of complex permittivity and the measurement frequency.

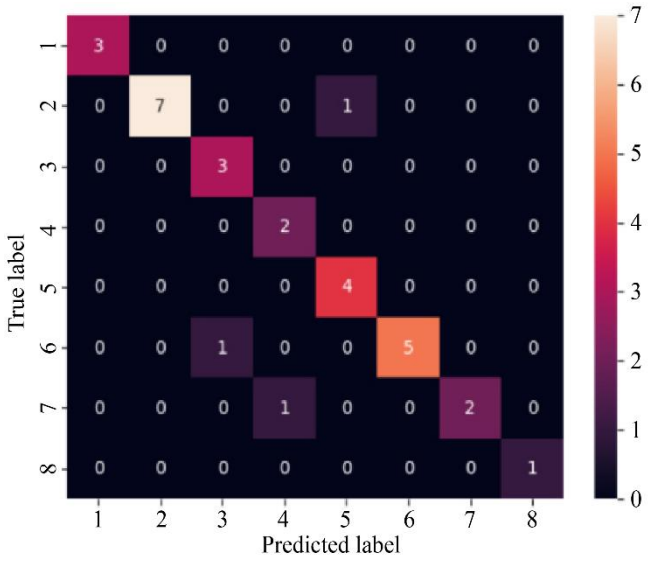
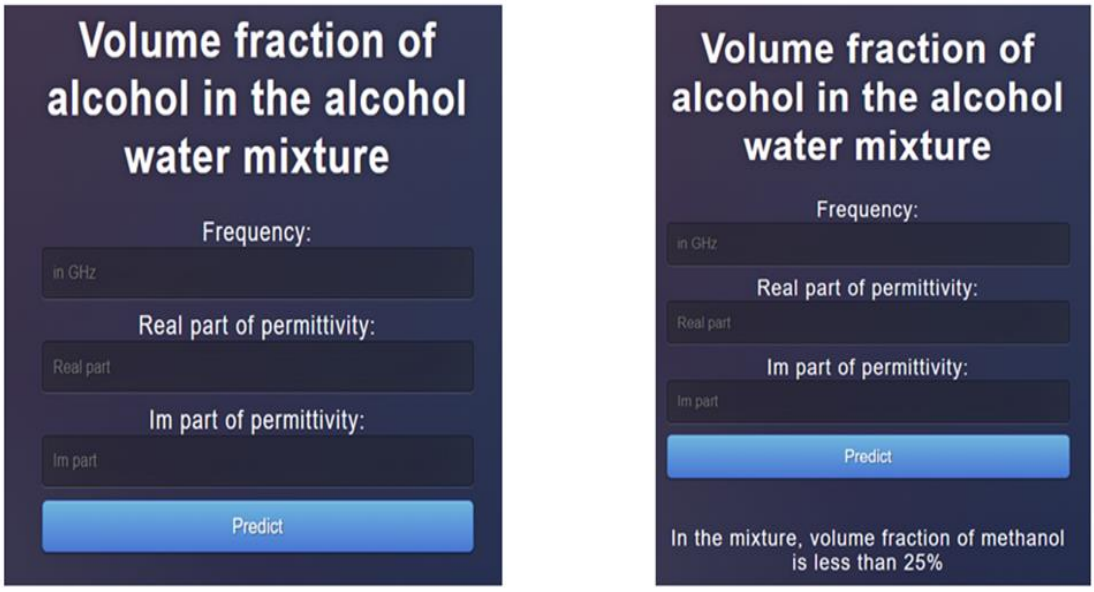


Fig. 10 Confusion matrix of the new test set

Table 4. Robustness of the proposed model

Liquid [Ref. No.]	Measurement Technique	F (GHz)	Max. Error in Permittivity E (%)	Number of Samples Supplied to the Proposed Model	Number of Samples Identified by the Proposed Model
Methanol-water [2]	Probe	0.1 – 20	1.62	16	15
Ethanol-water [2]	Probe	0.1 – 20	0.94	10	10
Ethanol-water [26]	Sensor	4.5	5.97	4	2
Total				**30**	**27**



(a) **(b)**

Fig. 11 Graphical user interface (a) Appearance, and (b) Example

The GUI is designed to identify both the type and fraction of alcohol in the mixture for a frequency range of 0.2 to 20 GHz at a constant temperature of 25°C. When the user inputs the necessary data, the system processes this information and displays the alcohol type and volume fraction. If the alcohol content in the sample falls below specific thresholds of 25%, 50%, 75%, or 100%, the corresponding message is shown on the front end of the GUI. In addition, if the measurement frequency exceeds the specified range, a warning message is displayed to alert the user. Figures 11(a) and 11(b) visually represent the GUI. Figure 11(a) shows the overall appearance of the interface, whereas Figure 11(b) demonstrates an example.

4. Conclusion

This study focuses on a major challenge in identifying and analyzing composite solutions. It specifically examines alcohol-water mixtures, which are widely used in different industries. Traditional methods for determining alcohol content are time-consuming and pose potential health risks due to chemicals and laser exposure. This work introduces an innovative approach that overcomes these challenges by utilizing ensemble machine learning and complex permittivity measurements. The measurements are taken in the frequency range of 0.2 to 20 GHz at 25°C. This method helps to identify the type and volume fraction of alcohol in aqueous alcohol solutions. The proposed model uses only a minimum number of parameters – complex permittivity and frequency data to accurately determine the type and concentration of alcohol in aqueous solutions, achieving an accuracy of 98.4%. It can also tolerate an error of ±5.5% due to temperature, measurement or sample purity variations. This method simplifies the evaluation process while ensuring reliable results. A GUI is designed to improve accessibility and ease of use. The model ensures fast characterization of polar liquids with a minimum number of input parameters and without any health risk.

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conceived the research idea. THP conducted the experiments, analyzed the data, and wrote the paper. MS reviewed and edited the manuscript.

Data Availability

The dataset generated during this work is available from the corresponding author upon reasonable request.

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