Sigma J Eng Nat Sci, Vol. 42, No. 3, pp. 633–641, June, 2024

Sigma Journal of Engineering and Natural Sciences
Web page info: https://sigma.yildiz.edu.tr
DOI: 10.14744/sigma.2024.00060

Research Article

Use of scattering parameters measured via a patch antenna and processed via regression analysis for flash point prediction of flammable liquid mixtures

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ARTICLE INFO

Article history
Received: 18 May 2022
Revised: 20 September 2022
Accepted: 14 February 2023

Keywords:
Flash Point; Hazardous Liquids; Prediction; Regression Analysis; Scattering Parameter; Security Checks

ABSTRACT

Flash point is an important parameter to determine the flammability properties of liquids. For this reason, determining the flash points of liquids is necessary for taking the necessary precautions for the storage and transportation of liquids. We propose that scattering parameters can be used to predict the flash points of flammable liquid mixtures. Therefore, in this study scattering parameters of various aqueous solutions were obtained using an antenna designed for this purpose and then the relationship between the flash points of the aqueous solutions and the scattering parameter was revealed by regression analysis. The results show that it is possible to predict the flash points of pure liquids and aqueous solutions from their scattering parameter measurement.

Cite this article as: Efeoğlu E, Tuna G. Use of scattering parameters measured via a patch antenna and processed via regression analysis for flash point prediction of flammable liquid mixtures. Sigma J Eng Nat Sci 2024;42(3):633–641.

INTRODUCTION

The release of flammable chemicals can lead to fire and/or explosion because flammable compounds develop when the released chemicals mix with air [1]. Vapour cloud explosion and boiling liquid expanding vapour explosion are the most common types of explosions in industries that involve chemical processes [2]. Considering its importance, the role of flammable liquids and vapours produced by flammable liquids was examined by comparing their physical properties [3]. While mixing hazardous chemical liquids can lead to fire pools, mixing incompatible chemicals can lead to exothermic oxidation [4]. Self-incineration is an accelerating factor for fires depending on the proximity of spilled liquid to the surrounding material and its characteristics.

When chemical reactions are not kept under control and stopped, they can lead to catastrophic consequences, such as explosions, fires, and toxic fumes [5]. As explained in [6], oxidisers can both cause fires and contribute to their increasing coverages. As it was explained in [7], accidental spillage and contamination of chemical liquids stored in small containers with open lids may lead to fires. As reported in [8], when candle lighting was replaced by

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This paper was recommended for publication in revised form by Regional Editor Gülhayat Gölbaşı Şimşek

kerosene lighting, many fires and explosions happened because of the application of unsafe kerosene. It was clear that there was a need to develop reliable indicators, flash and ignition points, for safe kerosene [8].

Flash point is a critical parameter for the flammability properties of liquids and knowledge of flammability properties of chemicals are highly important to protect against fires and explosions [9]. Therefore, liquids must be classified based on their flash points and then be appropriately stored [10]. Considering this, fire and explosion hazards of some flammable liquid mixtures were estimated in [11] and how the pressure affects the flash points of various fuels and their binary mixtures were studied in [12]. Experimental studies on the flash point of flammable liquids at high altitudes and small-scale combustion tests using flammable liquids in plastic containers for hazard analysis [13] and large-scale experimental studies have been conducted to investigate the combustion behaviour of these liquids [14].

In fires, flame, heat, smoke, toxic gases create a complex, toxic environment and leads to oxygen depletion [15]. In fires, most of the deaths are the consequence of the inhalation of toxic gases produced during combustion. Therefore, researchers provided methods for assessing life safety hazards in fires and understanding the hazardous effects of toxic gases, smoke, and heat on humans [16]. Fire risk assessments take the flash points of flammable liquids into consideration. Flash point is an important parameter in the use, storage and disposal of liquids. Because liquids with a low flash point ignite more easily, so they should be stored in cool environments.

In this study we provide an alternative approach to predict the flammability properties of aqueous solutions. The approach we proposed relies on using regression analysis with scattering parameters obtained by a patch antenna. Therefore, we designed an antenna to measure scattering parameters of liquids and then connected it to a vector network analyser. The rest of this article is as follows. A focused related work is given in the following section. Information about materials and methods used in this study is presented in the third section. The fourth section presents the results and our comments on the results. Finally, the article is concluded in the fifth section.

Related Work

Flash point is the lowest temperature at which the vapours of a volatile material can ignite. Materials with a low flash point can easily ignite; therefore, the flash point is taken into consideration while safety precautions are being evaluated. Safety precautions in a plant must ensure that the temperature in the environment never approaches the flashpoints of various materials. This phenomenon is known as auto-ignition temperature, and always great care must be taken whenever there is a need to work with hazardous materials or handle them in temperatures anywhere near their flashpoint or auto-ignition temperature. Though there are some equations used to calculate the flash points of various materials, they are seldom used because of significant variations in their accuracy [17].

If materials are classified according to their flammability risks, their safe storage and use can be ensured. Flash point testing is performed to determine whether a liquid is classifiable as flammable, ignitable or combustible. As flash point is the key factor to classify hazardous liquids into different categories according to their flammability risks, the accuracy and reliability of flash point measuring instruments must be evaluated [18]. Considering the vital of the measuring instruments, their accuracy and reliability must be validated and verified [19]. In the verification and validation of methods used in that process, reference materials play a key role [20].

There are many microwave methods for characterisation of different materials [21]. These measurement methods can be broadly divided into two categories as 1) resonant measurement methods and 2) non-resonant measurement methods [21]. Resonant measurement methods offer better accuracy and sensitivity than non-resonant ones at discrete frequencies and are more suitable for low-loss materials [21]. Non-resonant measurement methods use a wide frequency band, have high accuracy, and it necessitate less sample preparation [21]. Microwave non-resonant measurement methods can also be divided into two categories as 1) calibration-dependent non-resonant measurement methods and 2) calibration-independent non-resonant measurement methods [22,23]. Calibration-independent non-resonant measurement methods avoids the usage imperfect calibration standards, this way they may offer better accuracy [22,23]. They also reduce the overall measurement time.

The scattering parameter analysis is used to analyse the stability, input, and output reflection coefficients, power gains, and other parameters in a circuit and can be applied to any network at any frequency [24]. In [24], a new transmission-reflection method was proposed to measure reference-plane invariant constitutive parameters of liquid materials sandwiched in between two identical low-loss plugs. The method was shown to work well in limited frequency-band applications or for dispersive materials and was successfully used to measure the complex permittivity of methanol sample [24]. Its validity was proved by different methods. Calibrating a measuring instrument ensures that the instrument can accurately and reliably measure within the desired range for an application. However, it is sometimes neglected and this may lead to inaccurate measurements. In [25], a microwave method was proposed to determine non-ambiguous complex permittivity of liquid materials. The method relies on two-independent measurements of Eigen values for unique permittivity determination at one frequency.

In particular applications, complex permittivity data of liquids and binary mixtures at microwave frequencies is needed and they can be measured by using the waveguide techniques at the X and Ku band [26]. For this purpose, in

[26], the data measured using the open-ended waveguide technique are, compared with the data measured using the waveguide technique. The waveguide system was calibrated using custom designed through-reflect-line calibration kits. The measurement of complex permittivity for a number of liquids including ethyl alcohol, propyl alcohol, methanol, dioxane, cyclohexane, chlorobenzene, and binary mixtures demonstrated the dielectric properties of the liquids. In [26], it was also demonstrated that the liquid holders designed were effective for the measurement of complex permittivity of lossy, medium loss and low loss liquids.

Different from the literature, in this study an easy-to-implement testbed was designed and regression analysis, an easy to use quantitative tool, was used to predict the flammability properties of aqueous solutions. Although the literature mainly focuses on the use of complex equipment or methods, the testbed designed consists a commercial single-port vector network analyser (VNA) and a patch antenna designed in this study. Not only the design but also the measurement methodology of the proposed approach is simple and straightforward.

MATERIALS AND METHODS

In this section, materials and methods used in this study are explained.

Experimental Setup and Methodology

To realise the proposed approach, an experimental setup shown in Figure 1 was designed and built. The experimental setup consists of three components: a single-port VNA, a 50 Ohm SubMiniature version A (SMA) coaxial cable, and a 10 cm x 10 cm square-shaped antenna designed in this study. In the proposed approach, the antenna transmits electromagnetic waves to the bottle that contains liquid to be tested and then reflected signals are captured after the electromagnetic radiation interacts with the liquid. The signals captured by the VNA are called scattering parameters (S11 parameters). Scattering parameters are basically the indicator of the reflection properties of electromagnetic waves. The antenna was designed and built on a FR4 based dielectric substrate with a height of 1.6 mm and relative permittivity value of 4.4. The patch part of the antenna was 55 mm in diameter. (1) and (2) can be used to calculate the antenna's resonant frequency.

$$F = \frac{8{,}791x10^9}{f_r\sqrt{\varepsilon_r}} \quad (1)$$

$$a = \frac{F}{\left\{1+\frac{2h}{\pi\varepsilon_r F\left[\ln\left(\frac{\pi F}{2h}\right)+1{,}7726\right]}\right\}^{1/2}} \quad (2)$$

where ε_r denotes relative permittivity of the substrate, f_r denotes the resonant frequency, h denotes the height of the substrate, finally a denotes the radius of the patch.

Measurements should be made by keeping the antenna as close as possible to the container to obtain accurate results. The experimental setup allows measurements to be made when liquids are in closed containers. Therefore, it can be viewed as a non-destructive system and it is enough to approach the antenna to the container around 4-5mm. In this study, measurements were made between 1-3 GHz and in this frequency band S_{11} parameter of each liquid was measured. All of the measurements were made at room temperature and using the same container.

In this study, the relationship between the measurements taken and the flash points of flammable liquids was investigated. Because flash point is one of the key parameters to determine the fire hazard potential of liquids. The flash point values of alcohols in the literature are given in Table 1 [27]. These values were prepared from the data provided by International Safety Chemical Cards (ISCS), Society of Fire Protection Engineers (SFPE) [16], Hazardous Chemicals Handbook, namely CHC, Merck index [28].

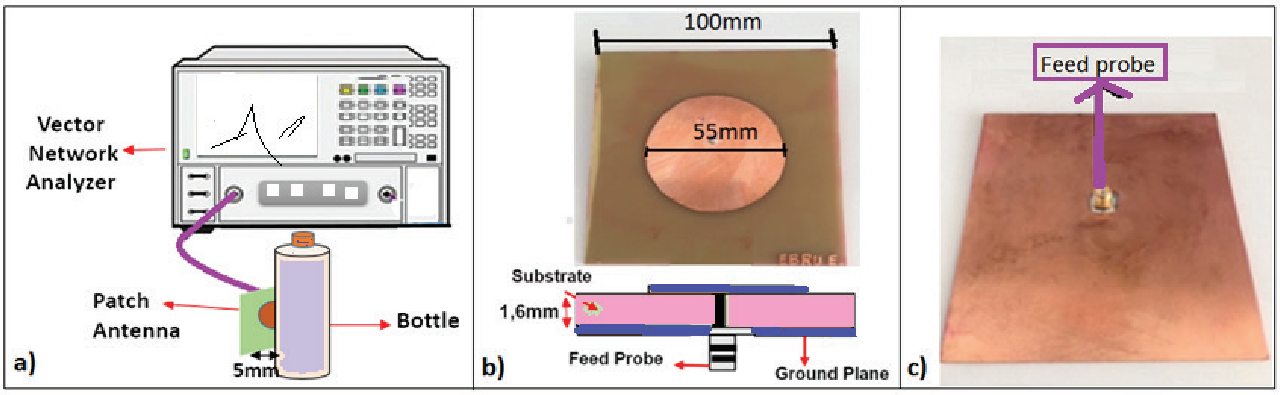

Figure 1. Experimental setup from different views: a) Measurement methodology, b) Design of the patch antenna, c) Feed probe of the patch antenna.

Table 1. Flash and boiling point values adopted from the literature

Component		Closed-Cup Flash Point (°C)			
	Formula	ISCS	SFPE	CHC	Merck
Methanol	CH_3-OH	12	12	12	10
Ethanol	CH_3-CH_2-OH	13	13	12	13
1-Propanol	CH_3-CH_2-CH_2-OH	15	15	15	22
Isopropanol	CH_3 CH-OH CH_3	11.7	12	16	12

Regression Analysis

Regression analysis is a statistical technique that is based on the analysis of the relationship between two or more variables of interest. It basically finds out the influence of independent variables on a dependent variable [29]. As it allows studying and measuring the relation between two or more variables [30], starting from data registered in a sample, it seeks to determine an estimate of a mathematical relation between the variables.

The aim of regression analysis is to estimate the value of one variable as a function of other variables [30]. In regression analysis, the estimated variable is referred as the dependent variable, and it is denoted by Y. Whereas, the variables that disclose the variations in Y are referred as the independent variables, and they are denoted by X [30]. In simple regression analysis, the dependent variable depends on only one independent variable. On the other hand, in multiple regression analysis, the dependent variable depends on more than one independent variable. In linear regression analysis, the relation between the dependent and the independent variables is linear. Regression analysis is typically used to determine the strength of predictors, forecast an effect, and perform trend forecasting.

In this study, the effect of changing alcohol concentration in aqueous alcohol solutions on S parameter (Smax) was investigated and regression analysis was used to predict the flash point from the S parameter measurements of aqueous alcohol solutions with different alcohol concentrations.

RESULTS AND DISCUSSION

Using the experimental setup, S11 parameters of pure and aqueous solutions containing 10-90% flammable liquid by volume were measured. The measurement results are presented in Figure 2. In parallel with the results presented in [31] and [32], as shown in Figure2, an increase in S11 parameter was observed as alcohol concentration in each aqueous solution increased. The flammable liquid concentrations and flash point values contained in these aqueous solutions and Smax values obtained from the S11 parameter measurements given in Figure 2 are listed in Table 2. Here, Smax is the peak value that represents the maximum amplitude value in S11 parameter measurements.

Matlab was used for all calculations in this study. Based on the data listed in Table 2, different curve fitting methods were applied to determine the relationship between Smax-alcohol concentration and Smax-flash points of flammable liquids. The highest coefficient of determination value was obtained when 2nd order polynomial curve fitting was applied. Therefore, 2nd order polynomial curve fitting was preferred. To realise this, a mathematical model was created in curve fitting algorithms and the correlation coefficient in this model was determined. The coefficient of determination (R2) can take a value between 0 and 1. If it is 0, there is no overlap. On the other hand, if it is 1, there is a perfect overlap. Therefore, it is desired to be as close to 1 as possible. The relationship between the Smax values of each aqueous solution and the concentration of the flammable liquid in the aqueous solutions is given in Figure 3. The relationship between these two parameters is shown in Figure 4 by using the Smax values and the flash point values given in Table 1.

In this study, two hypotheses were proposed and their accuracy was investigated. The first of these hypotheses was the relationship between the alcohol concentration of the liquids and their Smax values, and the second one was the relationship between the Smax values of the liquids and their flash points. In the first hypothesis, the dependent variable was the alcohol concentration and the independent variable is the Smax value. The null hypothesis (H_0) was that there was no relationship between the Smax value of liquids and alcohol concentrations in liquids. The alternative hypothesis (H_1) was that there was a relationship between the Smax values of the liquids and the alcohol concentrations in the liquids. In the second hypothesis the dependent variable was the flash point and the independent variable was the Smax value. It was investigated whether there was a relationship between these two variables. The null hypothesis (H_0) was that there is no relationship between the Smax values of liquids and their flash points. The alternative hypothesis (H_1) was that there is a relationship between the Smax value of liquids and their flash points. Root mean square error (RMSE), probability (p) and R^2 values, which show how well the regression model fits the data with 95%

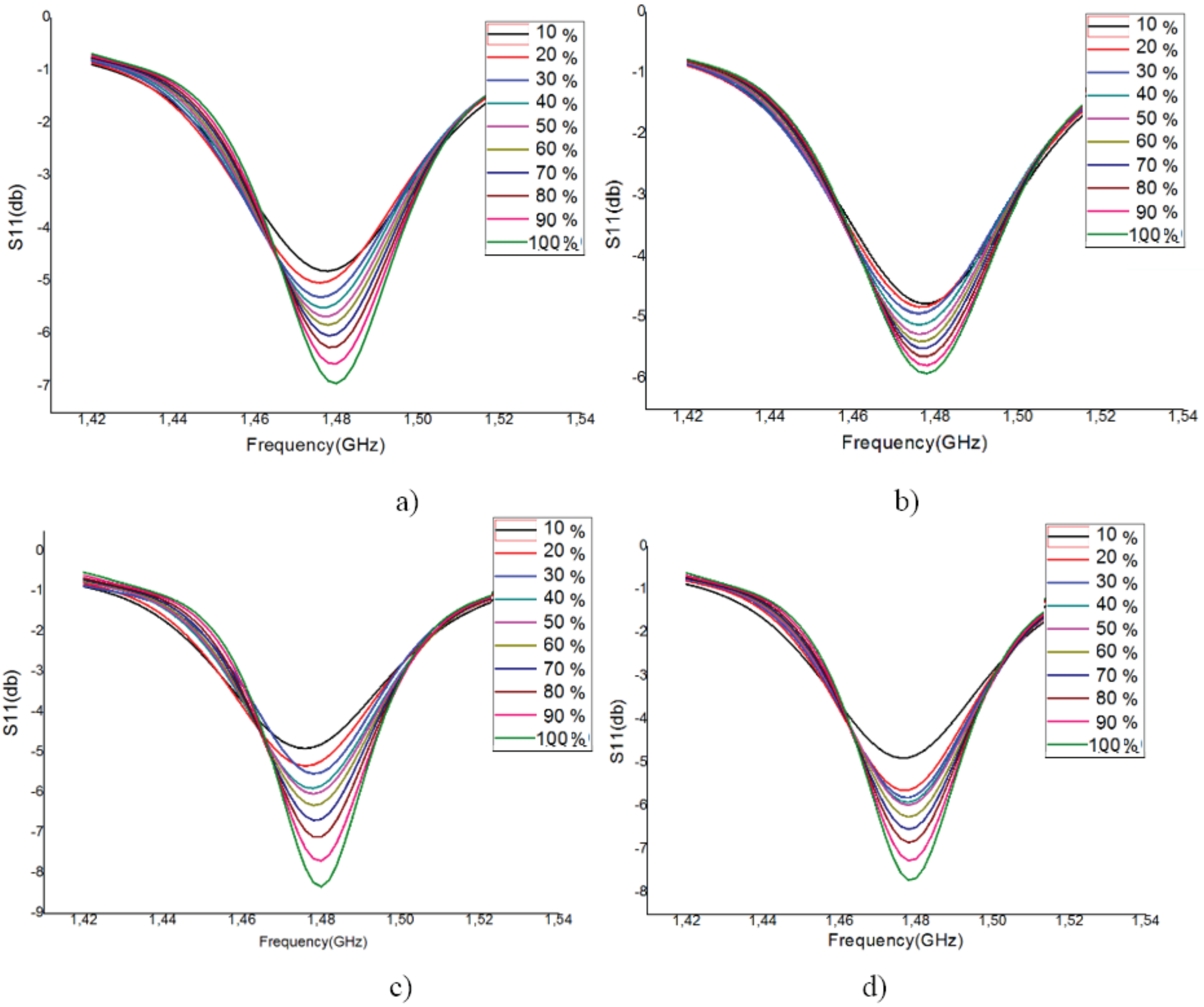

Figure. 2. S11 parameters of aqueous solutions with different flammable liquid concentrations: a) Ethanol, b) Methanol, c) 1-Propanol, d) Isopropanol.

Table 2. Alcohol Concentration and Flash Points in Different Alcohol-Water Mixtures [31,32].

Methanol-Water			**Ethanol-Water**			**1-propanol-Water**			**2-propanol-Water**		
Methanol Concentration	**FP ±2°C**	**S_{max}**	**Ethanol Concentration**	**FP ±2°C**	**S_{max}**	**1-Propanol Concentration**	**FP ±2°C**	**S_{max}**	**2-Propanol Concentration**	**FP ±2°C**	**S_{max}**
10%	53	4.7	10%	50	4.8	10%	43	4.9	10%	41	4.8
20%	46	4.8	20%	38	5	20%	35	5.3	20%	30	5.6
30%	39	4.9	30%	31	5.3	30%	34	5.5	30%	25	5.8
40%	33	5.1	40%	29	5.5	40%	--	---	40%	23	5.9
50%	28	5.2	50%	25	5.7	50%	33	6	50%	22	6
60%	25	5.3	60%	23	5.8	60%	--	---	60%	21	6.2
70%	20	5.4	70%	21	6	70%	31	6.6	70%	21	6.5
80%	18	5.6	80%	19	6.2	80%	30	7.1	80%	19	6.8
90%	15	5.7	90%	18	6.5	90%	26	7.6	90%	18	7.2
100%	12	5.9	100%	13	6.9	100%	22	8.3	100%	14	7.7

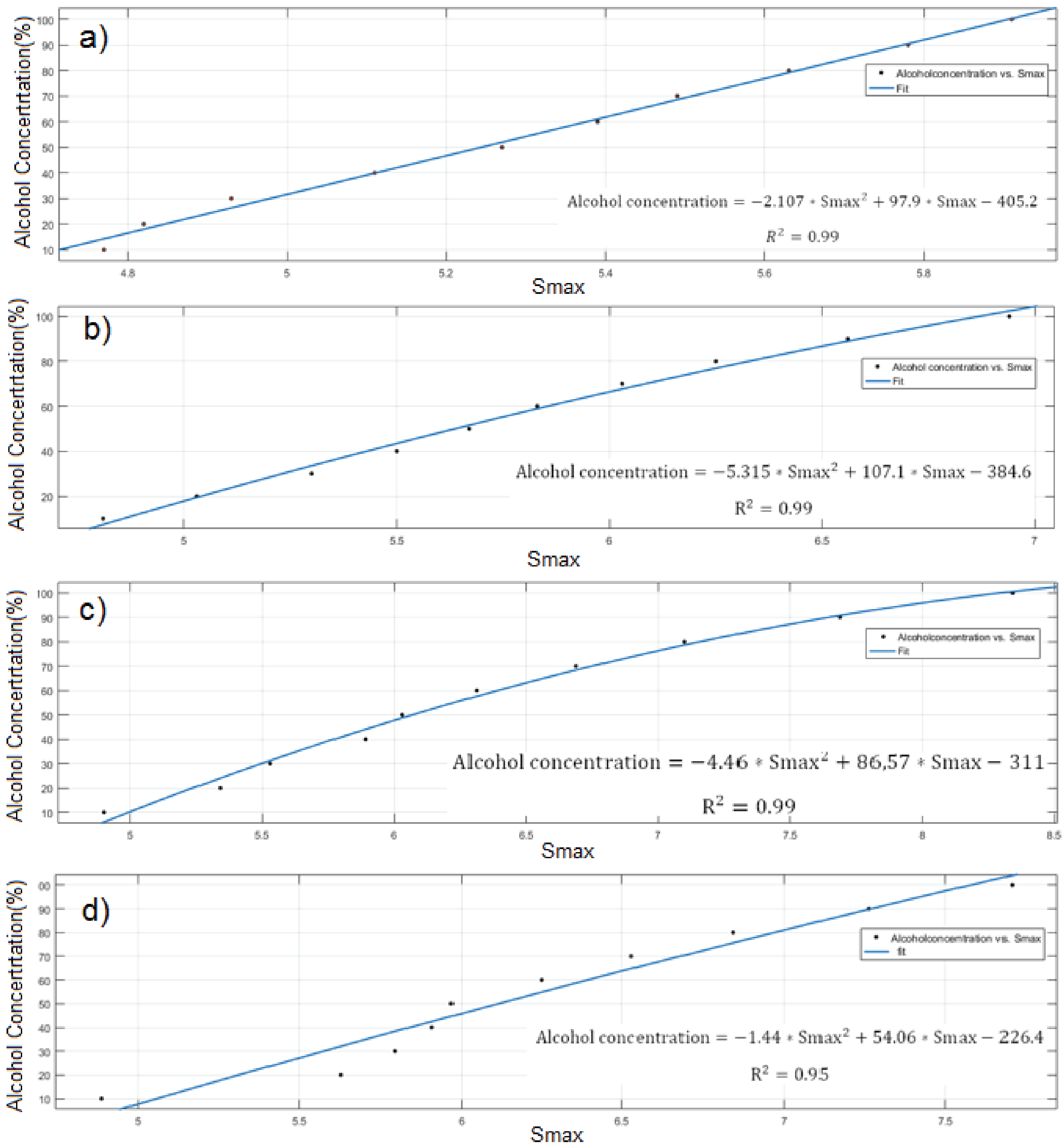

Figure 3. Relationship between alcohol concentration of aqueous solutions and Smax values: a) Methanol, b) Ethanol, c) 1-Propanol, d) Isopropanol.

confidence interval were calculated. The results are given in Table 3.

The error value (α) was taken as 0.05. H0 were rejected because the p values calculated were smaller than the error value. Therefore, it is assumed that there is a relationship between Smax values and flash points and between Smax vaues and alcohol concentrations. As in the literature there is no model that can be used to compare the obtained results, the results could not be compared.

The fire hazard of flammable liquids can be determined using flash point, because the lower the flash point, the greater the risk of fire [33]. Since the presence of water in solutions reduces the flammability of the solution, it should increase the flash point. Therefore, as the water concentration increases, the flash point increases. In other words, as the concentration of flammable liquid decreases, the flash point increases. As the concentration of flammable liquids in solutions increases, the Smax value increases, while the flash point increases, the Smax value decreases.

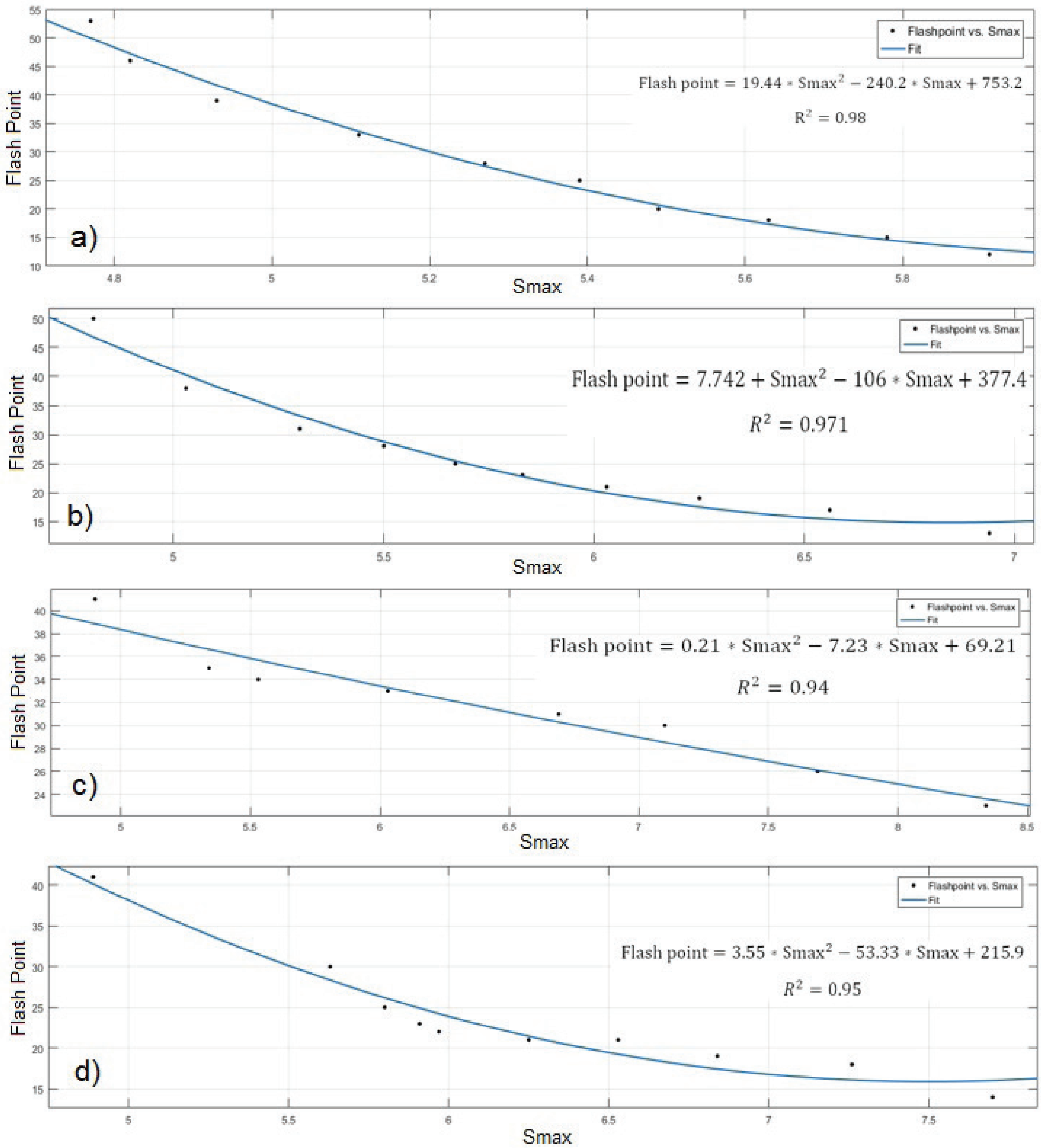

Figure 4. Relationship between flash points of aqueous solutions and Smax values: a) Methanol, b) Ethanol, c) 1-Propanol, d) Isopropanol.

Table 3. RMSE, probability (p) and R^2 values

	Alcohol Concentration and Smax			Smax and Flash Point		
	RMSE	R^2	p-value	RMSE	R^2	p-value
Methanol	0.9	0.99	1.83e-07	1.81	0.98	2.93e-07
Ethanol	0.88	0.99	2.35e-07	1.92	0.97	3.21e-07
1-propanol	0.85	0.99	2.93e-07	1.62	0.94	0.000929
Isopropanol	1.5	0.95	3.93e-07	1.82	0.95	2.51e-05

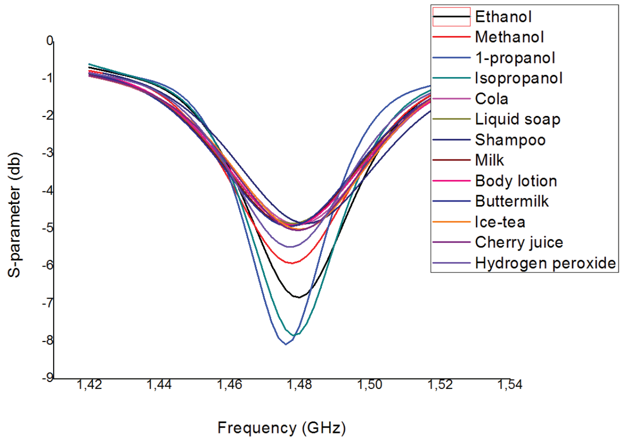

Figure. 5. Relationship between S11 parameter and frequency for different liquids.

This situation shows us that the fire hazard of the liquid increases with the increase of Smax value. Considering the S11 parameter measurement graph given in Figure 5, it is also seen that the flammable liquids, which are used in daily life and do not contain hazardous liquids, give a low Smax value, give a higher Smax value than these liquids.

CONCLUSION

Flash point is an important parameter to determine the flammability properties of liquids; therefore, in this study, the reflection properties of electromagnetic were used to predict the flash points of aqueous flammable liquids is proposed. As the results show, the fire risk of flammable liquid mixtures can be predicted using regression analysis and the scattering parameters of aqueous flammable liquids.

The experimental setup designed and built in this study can be used to take the necessary precautions for the storage and transportation of flammable liquids. However, implementation of the proposed approach on a prototype device is important for straightforward application in real world scenarios. After the prototype device is tested in a group of field tests, it is anticipated that the prototype device can be used to quickly examine the flash point of liquids at safety control points and this way decide whether they can be allowed to carry in or not.

AUTHORSHIP CONTRIBUTIONS

Authors equally contributed to this work.

DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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