OPTOELECTRONICS AND ADVANCED MATERIALS – RAPID COMMUNICATIONS Vol. 17, No. 7-8, July-August 2023, p. 265 - 271

HEC/PVDF coated glass substrate for acetone liquid sensor

MARLINA JAHAVA[1], MOHD HAFIZ JALI[1,*], AHMAD NUR AZEIM AHMAD ZAMZURI[1], MD. ASHADI MD JOHARI[2], AMINAH AHMAD[2], HAZIEZOL HELMI MOHD YUSOF[3], HUDA ADNAN ZAIN[4], SULAIMAN WADI HARUN[4], MOH YASIN[5]
[1]Faculty of Electrical Engineering, Universiti Teknikal Malaysia Melaka, 76100 Durian Tunggal, Melaka, Malaysia
[2]Faculty of Electrical and Electronics Engineering Technology, Technical University of Malaysia Melaka, 76100 Durian Tunggal, Melaka, Malaysia
[3]Faculty of Electronic Engineering and Computer Engineering, Technical University of Malaysia Melaka, 76100 Durian Tunggal, Melaka, Malaysia
[4]Department of Electrical Engineering, University of Malaya, 50603 Kuala Lumpur
[5]Department of Physics, Faculty of Science and Technology, Airlangga University, Surabaya, Indonesia

An acetone liquid sensor has been successfully demonstrated by utilizing glass substrate coated with Hydroxyethylcellulose/Polyvinylidene Fluoride (HEC/PVDF). The sensing mechanism is based on the change in refractive index (RI) of the HEC/PVDF coating layer when exposed to acetone concentration variations. When the light progress through the sensing material, the intensity decrease due to absorption and scattering. The proposed sensor's produces exceptional results where output voltage has decreased linearly from 1.8 V to 1.3 V. It showed a good response with sensitivity of 0.0331 V/% and a linearity of 99.1 % as compared to an uncoated glass substrate.

(Received February 2, 2023; accepted August 7, 2023)

Keywords: Acetone, HEC/PVDF, Glass substrate

1. Introduction

A significant number of industries employ acetone in their operation, which makes it an essential chemical substance to be investigated; each with its unique detection method has been devised [1-3]. Different sensing mechanisms for gas/liquid chromatographic have been developed to determine the concentration of acetone [4-6]. Each sensing capabilities has own sensing operation compared to existing acetone liquid sensor such as ethanol [7], formaldehyde [8], and Glucose [9]. Currently, numerous types of acetone sensors based on diverse sensing principles have been developed such as semiconductor sensor [10], electronic sensor [11], and microspheres-based sensor [12]. However, these sensors still have shortcomings, including low sensitivity, short life spans, and instability. Various proposed sensors have been researched and compared in an effort to find a better sensor in terms of sensitivity stability, such as carbon-based 1-D has low selectivity [13], porous silicon has poor stability [14, 15] and wide band gap semiconductor also has low sensitivity [16]. Therefore, the optical-based device for acetone detection is an excellent candidate to producing exceptional sensors with high sensitivity, fast response time, and stability. Optical sensor has superiority as compared to other type of sensor in term of sensitivity because has good evanescent coupling with other waveguides such as metal, semiconductor and substrate as well as offers strong near-field interaction with its surrounding [17, 18]. Therefore, this work aims to develop an acetone sensor that is easier to operate, more effective, and less expensive, and does not pose an increased risk to human health or the environment with encompasses sensing response toward coating material.

Over the past decades, with increased concerns about the energy issue, coating nanomaterial on glass has been the subject of numerous research and is widely employed in contemporary construction. Identifying coating materials that are fit for the task of providing many benefits is necessary [19]. Several research projects on investigating suitable materials to be used for sensing have been carried out on a variety of platforms, such as Hydroxyethyl-cellulose and Polyvinylidene Fluoride (HEC/PVDF) [20], Zinc Oxide Nanorods [21], ZnFe-LDH/GO nanocomposite [22], and Agarose gel coated layer [23]. Hydroxyethyl cellulose (HEC) is a cellulose-derived gelling and thickening substance [24], and Polyvinylidene Fluoride (PVDF) is a thermoplastic fluoropolymer made from the polymerization of vinylidene difluoride that is very resistant to chemical reactions and is employed as an internal coating due to its ease of absorption [25]. According to the literature, HEC/PVDF exhibits good sensing material characteristics, such as the high capacity for water absorption, which modifies the coating refractive index and could indeed increase the sensitivity of the sensing structure. Furthermore, it is more stable, cost-effective, and widely used [26]. This makes it a suitable nanomaterial to produce a low-cost and high-sensitivity sensor. It could be realized by integrating the nanomaterial-coated glass substrate with the sensing circuit. This is due to the uncoated glass substrate being less sensitive because it has no sensitive substance coated on it. It has poor sensing performance because of a low refractive index mismatch between the ambient analyte and the sensor.

Existing optical sensors commonly comprise a light source, an optical spectrum analyzer, and a photodetector, a more expensive and complicated technique for the sensing operation. To achieve our aim to produce a low-cost and highly sensitive sensor, a glass substrate coated with HEC/PVDF nanomaterial is integrated with a sensing circuit that includes a main component, such as a LED to transmit light through the glass substrate and a photodiode that converts light into a voltage signal [27, 28]. In terms of the output signal analysis, it is connected to the conditioning amplifier circuit so that the suitable signal may be sent to the Data Acquisition (DAQ) unit for signal processing of the transmitted light [29]. Arduino microcontroller is used as DAQ to process the data from the signal. To perform the measurement, the detector was carefully attached at the edge of the coated glass substrate, one end of the coating material was connected to a light source, and the other was connected to a photodetector. The proposed HEC/PVDF coated glass substrate for the acetone liquid sensor has been demonstrated for the first time in our knowledge.

2. Sensing mechanism

Nanomaterial have evolved into a crucial element in nanotechnology. The majority of those who utilize the nanomaterial as sensitive materials pay less attention to exploring the light scattering phenomena feature that occurs on the coating material [30]. Light transmission through the glass substrate is significantly influenced by the dispersion coefficient of the coating material and the total fractional power carried in the sensing region. This can be demonstrated by an equation (1) discovered by Beer-Lambert Law:

$$I = I_o e^{-\alpha L} \tag{1}$$

where I signifies the intensity of light leaving the sensing sector, while I_o indicating the intensity of light entering the sensing territory, α is the scattering coefficient, and L is the sensing region length. To quantify these functions, an instrument that measures the angular distribution of light reflected or transmitted by a surface must be used to complete knowledge of optimize the optical properties. To fully characterise the optical properties of each layer, the reduced scattering coefficient (μs*), absorption coefficient (μ_α) and phase function must be known [31]. This is because impacted by the bulk absorption coefficient, concentration, and effective fraction of total directed power of the absorbing material [32]. An overview of the sensing mechanism is shown in Fig. 1 where transmission of light through to the sensing substance. This can cause the intensity of the light to be reduced due to factors such as absorption and scattering. Besides, the tone of the scattering surface affects incident light intensity and wavelength of the reflected light are also two factors that need to determine the strength of the scattered light. The intensity of dispersed light is determined by the particle size and the light's wavelength. To get a good scattered of light is with considering P to be the possibility of scattering and λ to be the wavelength of light, the equation (2) is as follows:

$$\mathrm{P}\ \alpha\ \frac{1}{(\lambda)^4} \tag{2}$$

This can also contribute where there is a refractive index mismatch at the boundary. With a higher refractive index (RI) solution, more light would leak out in point of fact optical transmittance (T) also reduces due to the molecular content increases which increases sensitivity to acetone levels [33, 34]. The percentage of incoming light that makes it through a substance is known as its transmittance, or T, where I signifies transmitted light (output) and I_o indicates incident light (input). Given the theoretical value output, using the following Equation (3) to get the optical transmission (T) at the sensor [33].

$$\mathrm{T} = \frac{I}{I_o} = e^{-\alpha L} \tag{3}$$

A proposed sensor is shown in Fig. 1 is utilized pertaining to the characterization of the sensing structure. In order to observe the sensing response, a light source is supplied at one edge of the glass substrate to expose the concentrations of acetone liquid. The amount of light emitted will gradually decrease subsequent to its initiation of interaction with the sensor substrate [35]. The increase in refractive index contrast between surrounding of coated glass layers as a result of scattering will lead to more light leakage as concentrations grow, resulting in a lower output voltage value [36]. There are many other factors that contribute to this scenario; however, the structure that has been proposed is aimed at achieving notable sensing properties such as high sensitivity, high linearity, and reliable measurement.

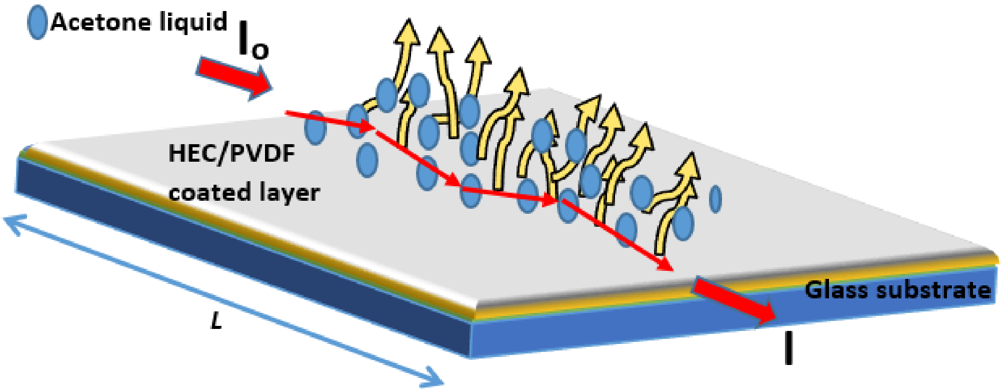


Fig. 1. Sensing mechanism of the proposed sensor

3. Experiment detail

The experimental approach for this project comprises several phases. Before the coating process started, the glass substrates had to go through preparatory steps. The edges of the glass have been cut into several little shards, each measuring 22 millimetres (L) x 15 millimetres (W). Prior of ultrasonic cleaning process, the glass substrates (Heathrow Scientific LLC) were sequentially submerged for 15 minutes in a container containing soap water, clear water, and acetone (CH3COCH3, Bendosen Laboratory Chemical). After that, it was placed in an oven at a temperature of 90 degrees Celsius for one hour in order to eliminate any biological material [20]. The HEC/PVDF solution was prepared by dissolving 1g of PVDF powder (Mw=275,000) in 120 ml of dimethyl form amide (DMF) in a water bath at 90^oC. Subsequently, 4 g of hydroxyethyl cellulose were added to the PVDF solution (HEC). The three-dimensional structure of the mesh gel was developed when the combined solution was agitated continuously for around 10 hours at room temperature (hydrogel). After waiting 48 hours for the HEC/PVDF combination to dry, it was carefully applied to the glass substrate [37]. Subsequently, a little quantity of the mixture was gently placed onto the glass substrate, then exposed to particles at ambient conditions for 24 hours. Fig. 2 depicts a glass substrate with HEC/PVDF coated glass substrate. To develop a low-cost sensor device in this work, a commercial green LED light source with a wavelength between 495 nm and 570 nm was used [38]. The LED and photodetector were placed as close to the edges of the glass substrates, with as little space between them as possible. To ensure total internal reflection is maximized, the LED was positioned at a 50° angle near the glass substrate's border, resulting in a 60° incident angle. A photodetector is linked to a precondition amplifier, which elevates the voltage signal so it can be processed by a data acquisition (DAQ) device. The photodetector, which transformed light intensity into an electrical signal, and the light source were separated by a glass substrate. For this reason, a commercial green LED was implemented as the light source, and a photodiode was used as the light detector [39]. The suggested sensor, based on a HEC/PVDF-coated glass substrate, was tested in a controlled environment utilizing a data acquisition (DAQ) device as shown in Fig. 3. In additionally, the experiment was performed at room temperature, with acetone concentration levels ranging from 0% to 15%, and the started were investigated with 0% as a reference value [36]. This was accomplished by using pure water (0%). The experiment was continuously done with acetone sensing to obtain different results by placing on the surface HEC/PVDF coated glass substrate. Eventually, the behavior of the output voltage is analyzed according to the performance of the sensing parameters such as sensitivity, linearity, repeatability, hysteresis, response time and stability.

Fig. 2. HEC/PVDF coated glass substrate

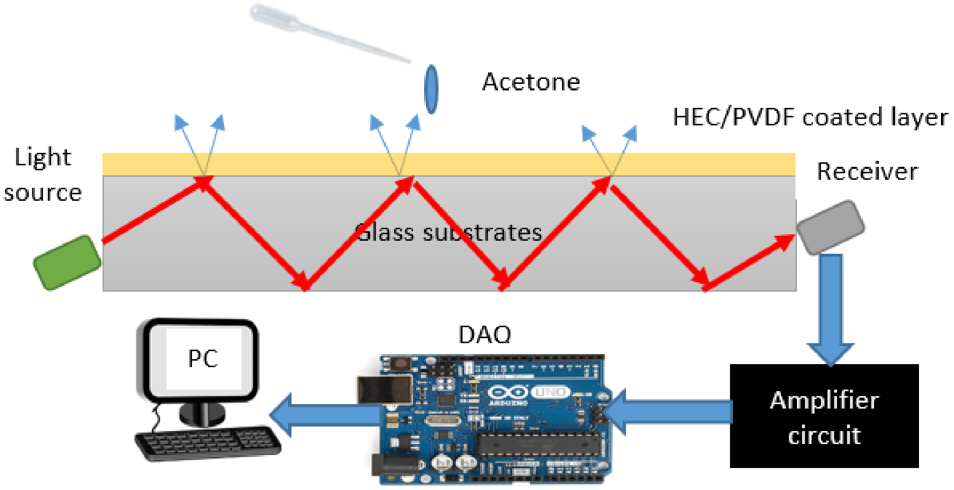
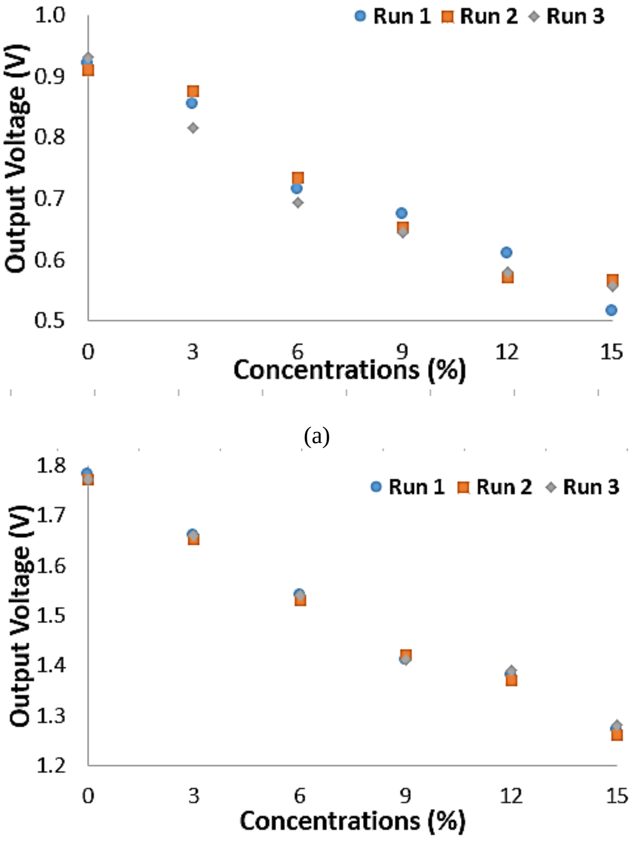


Fig. 3. Experimental setup of the proposed sensor

4. Result and discussion

Figs. 4 (a) and 4 (b) show that the proposed sensor was tested at least three times to determine the standard deviation of the output voltage, which is a response of the sensor acetone towards HEC/PVDF coated glass and uncoated glass by light scattering effect on glass substrates. Referring to Fig. 4 (a), the uncoated glass showed that the differences in deviation voltage ranged around 0.06 compared to the coated glass, as shown in Fig. 4 (b), where the differences in deviation voltage ranged around 0.03, and compared to the uncoated glass, the accuracy of the results for each concentration was higher. More importantly, the whole range shows that the voltage value will be lower at the higher concentration level. Fig. 5 shows hysteresis performances based on the output voltage that was measured in both forward and reverse directions. As depicted in Fig. 5 (a), the variations between forward and reverse values are extremely evident, particularly at concentration levels of 9% and 12% for uncoated glass. For uncoated glass, the difference between forward and reverse measurements is around 0.1V. The minor discrepancy in sensor response is due to the different adsorption and desorption rates of analyte molecules on the coating layer [38]. Furthermore, the output voltages versus concentration in the ranges of 0% to 15% show for both forward and reverse variations has a very small margin of error across the range, as shown in Fig. 5(b). Especially at 9% concentration, where there is a smaller divergence with a lesser output voltage differential of roughly 0.01V.

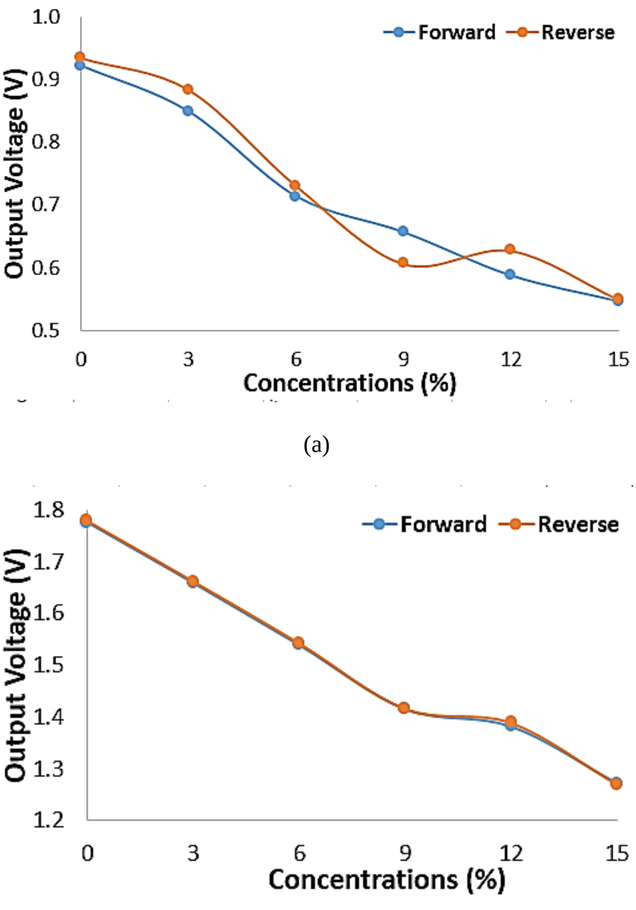


(a)

(b)

Fig. 4. The repeatability of; a) Uncoated glass and b) HEC/PVDF coated glass (color online)

Moreover, to verify the sensor's usefulness, a stability test was conducted by recording sensor readings for 600 seconds, and each second at five concentration levels with a total time is 10 minutes. A stability reading of uncoated glass is depicted in Fig. 6(a), shows a less stable output voltage during the whole period of the test. This is due to the fact that the uncoated glass has a low refractive index contrast as compared to the coated glass. Therefore, more consistent light refraction occurred to the HEC/PVDF coated glass substrate. This lead to more stability to the proposed sensor. This is demonstrated by the data from the stability output voltage readings shown in Fig. 6 (b). Fig. 7 depicts the experimental setup for the response and recovery time for both sensing, which were achieved by continuously adding acetone from the lowest concentration value to the highest concentration value during the test duration of 480 seconds, or 8 minutes. The concentration of acetone has also been adjusted from the minimum to the maximum concentration, and likewise every 120 seconds or 2 minutes until two cycles have been completed [36]. The overall response time for uncoated glass and HEC/PVDF coated glass is displayed in Fig. 7 (a), which is the uncoated glass react from the lowest to the greatest concentration is 1.5 second has a slower time as compared to a HEC/PVDF coated glass substrate, the higher output voltage concentration is 1.47 second. While recovery time shows it was reacting directly from the highest concentration value to the lowest concentration.

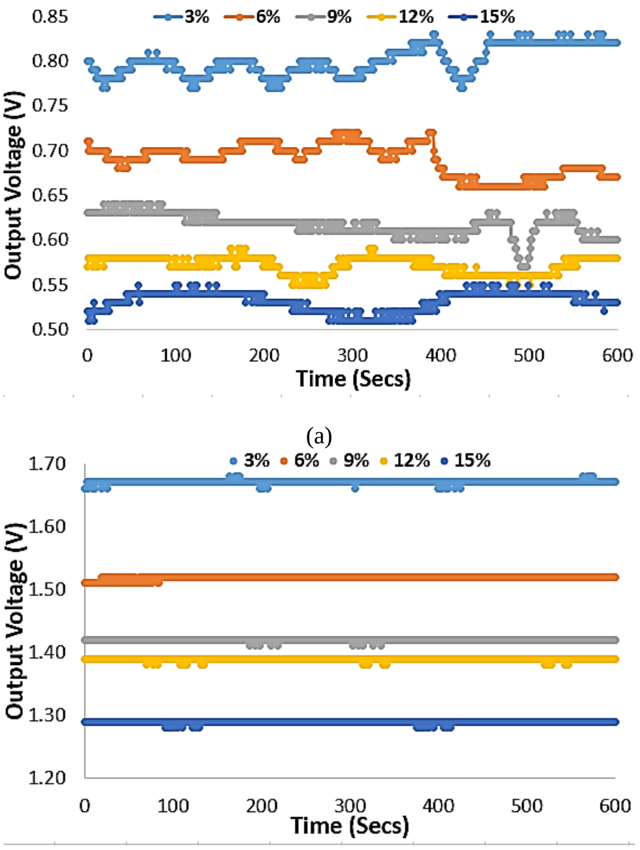


(a)

(b)

Fig. 5. The hysteresis graph of; a) Uncoated glass and b) HEC/PVDF coated glass (color online)

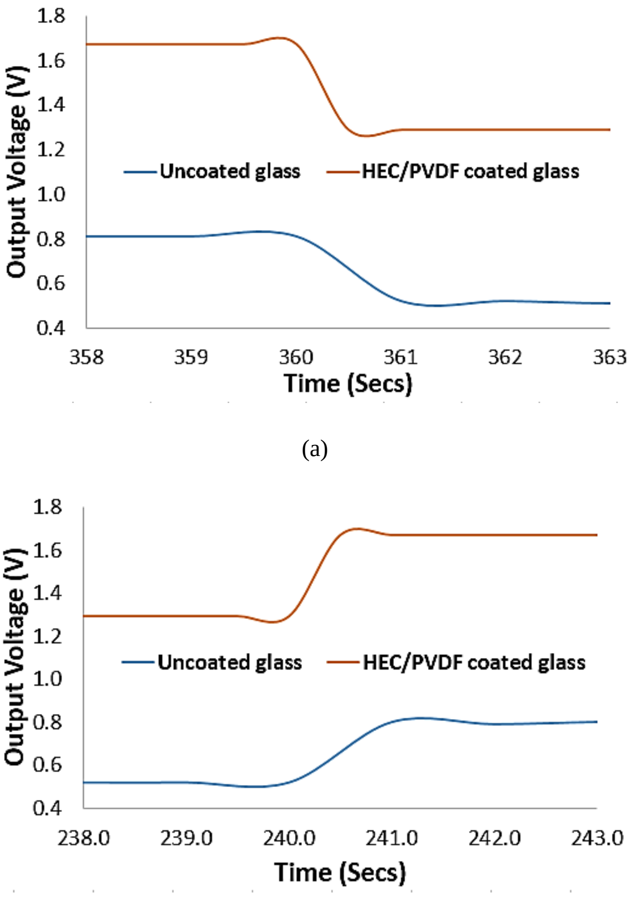
where the output voltage of HEC/PVDF coated glass is greater than that of uncoated glass, with a reaction time of 1.57 seconds against 2.0 seconds, as shown in Fig. 7 (b). These findings demonstrated that the time response of the glass substrate was enhanced after being coated with HEC/PVDF. Fig. 8 depicts the sensing response when exposure to increasing acetone concentrations levels for both sensors along the concentrations level range of 0% to 15%. In addition, the performance parameters of both sensors have been compared and summarised in Table 1. This table shows that coated glass has superior performance, as indicated by the percentages of sensitivity (0.0331%) and linearity (99.1%), in contrast to uncoated glass, which has a lower sensitivity (0.025%) and linearity (only 95.41%). The detection of the acetone sensor on the coating material is based on the fact that the acetone molecule has a higher dipole moment [1, 40], making it easier for it to react with the HEC/PVDF coated glass substrate compared to uncoated glass. Our proposed sensor also shows better performances in term of linearity as compared to other acetone liquid sensors found in the literature [41, 42] which exhibit linearity of 94.38% and 96.91% respectively as compared to our result which is 99.1%. Sensitivity of our sensor (0.0331 V/%) also better as compared to acetone liquid sensor in [42], which is 0.0269 V/%. The change in the refractive index (RI) of the coating materials also can modify the light output intensity [37]. The findings have demonstrated that all of the sensor's properties, including its stability, linearity, and sensitivity, have been successfully improved.



(a)

(b)

Fig. 6. The stability of; a) Uncoated glass and b) HEC/PVDF coated glass (color online)



(a)

(b)

Fig. 7. Time response; a) Response time and b) Recovery time (color online)

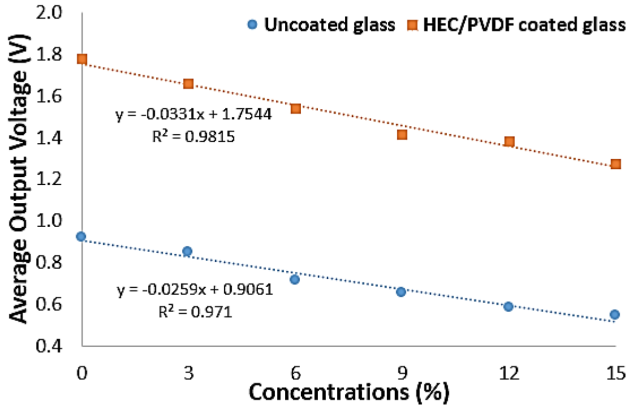


Fig. 8. Trend line of both sensors (color online)

Table 1. Summary of the sensing performances

Parameters	**Uncoated glass**	**HEC/PVDF coated glass**
Sensitivity (V/%)	0.025	0.0331
Linearity (%)	95.41	99.1
Std. deviation (V)	0.0153	0.00718
Resolution (%)	0.612	0.217
Time response (Secs)	1.0	0.6

5. Conclusions

In summary, the proposed acetone liquid sensor based on HEC/PVDF coatings layer that act as changeable reflective index (RI) on glass has been successfully implemented. The proposed sensor exhibits several advantages, such as high sensitivity, fast response, and stability. The sensor shows a strong linear response when exposed to concentration levels between a range of 0% to 15%. In each tested concentration range, the proposed sensor shows superiority in all sensing performance parameters as compared to the uncoated glass. The experimental results show that a sensitivity of 0.0331 V/% as compared to the sensitivity of uncoated glass of 0.025 V/% and a linearity of 99.1%, which is higher than uncoated glass, demonstrating a greater potential as an acetone sensor. Overall, the proposed sensor produces better sensing performances in terms of sensitivity, linearity, repeatability, hysteresis, stability, and time response. In the future, a wide variety of coating materials might be used as sensitive materials to examine sensing performance in relation to acetone concentrations.

Acknowledgements

This research was funded by UTeM and MOHE through Fundamental Research Grant Scheme (No. FRGS/1/2022/FKE/F00499).

References

[1] H. Zhang, H. Qin, C. Gao, J. Hu, Sensors **18**(7), 2318 (2018).
[2] F. Liu, Y. Guan, R. Sun, X. Liang, P. Sun, F. Liu, Sensors and Actuators B: Chemical **221**, 673 (2015).
[3] X. Liu, C. Qu, S. Zhou, W. Cao, M. Xu, Y. Liu, Optics Communications **526**, 128890 (2023).
[4] C. Deng, W. Zhang, J. Zhang, X. Zhang, Journal of Chromatography B **805**(2), 235 (2004).
[5] K. Drozdowska, T. Welearegay, L. Österlund, J. Smulko, Sensors and Actuators B: Chemical **353**, 131125 (2022).
[6] X. Zhang, B. Dong, W. Liu, X. Zhou, M. Liu, X. Sun, J. Lv, L. Zhang, W. Xu, X. Bai, L. Xu, Sensors and Actuators B: Chemical **320**, 128405 (2020).
[7] J. G. Sotelo, J. Bonilla-Ríos, F. García-Escobar, J. L. Gordillo, Coatings **11**(3), 303 (2021).
[8] S. H. Johari, T. Z. Cheak, H. R. A Rahim, M. H. Jali, H. H. M. Yusof, M. A. M. Johari, S. W. Harun, IEEE Access **10**, 91445 (2022).
[9] S. Mishra, P. Yogi, P. R. Sagdeo, R. Kumar, Nanoscale Research Letters **13**, 1 (2018).
[10] Y. Anno, T. Maekawa, J. Tamaki, Y. Asano, K. Hayashi, N. Miura, N. Yamazoe, Sensors and Actuators B: Chemical **25**(1-3), 623 (1995).
[11] H. Zhang, A. Kulkarni, H. Kim, D. Woo, Y. J. Kim, B. H. Hong, J. B. Choi, T. Kim, Journal of Nanoscience and Nanotechnology **11**(7), 5939

(2011).

[12] P. Cheng, L. Lv, Y. Wang, B. Zhang, Y. Zhang, Ya. Zhang, Z. Lei, L. Xu, Sensors and Actuators B: Chemical **332**, 129212 (2021).

[13] G. Korotcenkov, V. Brinzari, M. Ham, Key Engineering Materials **780**, 80 (2018).

[14] J. Salonen, V. P. Lehto, M. Björkqvist, E. Laine, L. Niinistö, Physica Status Solidi (a) **182**(1), 123 (2000).

[15] S. Cherevkov, R. Azizov, A. Sokolova, V. Nautran, M. Miruschenko, I. Arefina, M. Baranov, D. Kurdyukov, E. Stovpiaga, V. Golubev, A. Baranov, E. Ushakova, Nanomaterials **11**(1), 119 (2021).

[16] N. Bakranov, A. Zhabaikhanov, S. Kudaibergenov, N. Ibraev, IOP Conf. Series: Journal of Physics: Conf. Series **987**, 012028 (2018).

[17] T. Subashini, B. Renganathan, A. Stephen, T. Prakash, Materials Science in Semiconductor Processing **88**, 181 (2018).

[18] M. M. Rahman, M. Alam, A. M. Asiri, Nanoscale Advances **1**(2), 696 (2019).

[19] G. Duan, G. Zhao, X. Lin, T. Zhang, G. Han, Conference on Lasers and Electro-Optics/Pacific Rim 2009), paper TUP6_32 (2009).

[20] M. Jali, PrzeglĄd Elektrotechniczny **1**(4), 42 (2022).

[21] H. H. M. Yusof, M. H. Jali, M. A. M. Johari, K. Dimyati, S. W. Harun, M. Khasanah, M. Yasin, IEEE Photonics Journal **11**(1), 1 (2019).

[22] P. Y. Motlagh, A. Khataee, A. Hassani, T. S. Rad, Journal of Molecular Liquids **302**, 112532 (2020).

[23] M. Jali, Przegląd Elektrotechniczny **1**(8), 97 (2022).

[24] M. Zhang, M. X. Li, Z. Chang, Y. F. Wang, J. Gao, Y. S. Zhu, Y. P. Wu, W. Huang, Electrochimica Acta **245**, 752 (2017).

[25] G.-D. Kang, Y.-M. Cao, Journal of Membrane Science **463**, 145 (2014).

[26] H. A. Zain, M. Batumalay, H. R. A Rahim, M. Yasin, S. W. Harun, Optik **232**, 166534 (2021).

[27] P. Yeh, N. Yeh, C. H. Lee, T. J. Ding, Renewable and Sustainable Energy Reviews **75**, 461 (2017).

[28] S. H. Zyoud, N. M. Ahmed, A. S. Z. Lahewil, A. F. Omar, Sensors and Actuators A: Physical **338**, 113485 (2022).

[29] M. H. Jali, N. N. Malek Faisal, H. R. Abdul Rahim, M. Johari, M. Ashadi, A. Ahmad, M. F. Baharom, H. A. Zain, H. W. Harun, Przegląd Elektrotechniczny **98**(8), 97 (2022).

[30] M. P. Diebold, Application of Light Scattering to Coatings: A User’s Guide, Springer 2014.

[31] V. M. Turzhitsky, A. J. Radosevich, J. D. Rogers, N. N. Mutyal, V. Backman, Journal of Biomedical Optics **16**(6), 067007 (2011).

[32] S. Azad, E. Sadeghi, R. Parvizi, A. Mazaheri, M. Yousefi, Optics & Laser Technology **90**, 96 (2017).

[33] H. H. M. Yusof, S. W. Harun, K. Dimyati, T. Bora, W. S. Mohammed, J. Dutta, Photonics and Nanostructures-Fundamentals and Applications **30**, 57 (2018).

[34] M. Mazilu, N. Tigau, V. Musat, Optical Materials **34**(11), 1833 (2012).

[35] X. Fang, Y. Bando, U. K. Gautam, T. Zhai, H. Zeng, X. Xu, M. Liao, D. Golberg, Critical Reviews in Solid State and Materials Sciences **34**(3-4), 190 (2009).

[36] M. Jali, Przegląd Elektrotechniczny **1**(5), 73 (2022).

[37] M. Batumalay, S. W. Harun, N. Irawati, H. Ahmad, H. Arof, IEEE Sensors Journal **15**(3), 1945 (2014).

[38] M. H. Jali, H. R. A. Rahim, M. A. M. Johari, A. Ahmad, H. H. M. Yusof, S. H. Johari, M. F. Baharom, S. Thokchom, K. Dimyati, S. W. Harun, Optics & Laser Technology **143**, 107356 (2021).

[39] H. H. M.Yusof, S. W. Harun, K. Dimyati, T. Bora, K. Sterckx, W. S. Mohammed, J. Dutta, IEEE Sensors Journal **19**(7), 2442 (2018).

[40] L. Wang, A. Teleki, S. E. Pratsinis, P. I. Gouma, Chemistry of Materials **20**(15), 4794 (2008).

[41] N. F. H. Zailani, N. Saidin, M. F. M. Rusdi, S. W. Harun, P. M. Thirunavakkarasu, 2021 IEEE Regional Symposium on Micro and Nanoelectronics (RSM) **100**, 103 (2021).

[42] M. H. Jali, A. N. Zamzuri, H. R. Rahim, M. A. Johari, A. Ahmad, M. F. Baharom, H. A. Zain, S. W. Harun, Przegląd Elektrotechniczny **98**(5), 7175 (2022).

---

[*] Corresponding author: mohd.hafiz@utem.edu.my