

SUSTAINABLE ANTIMICROBIAL COPOLYMER NANOEMULSION OF *Aloe Vera* AND ALLYL ALCOHOL: AN *In-situ* APPROACH

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ABSTRACT

The current study describes the formation of a new sustainable *Aloe Vera* and allyl alcohol nanoemulsion with outstanding antimicrobial efficacy against Gram (+ive) and Gram (-ive) bacteria. The sustainable nanoemulsion of allyl alcohol/butyl acrylate/methyl methacrylate/ *Aloe Vera* with surfactant initiated by potassium persulphate was synthesized using a semi-continuous seeded emulsion polymerization technique. Several techniques like ¹H, ¹³C NMR, XRD, SEM with EDX, DSC, TGA, DTA, FTIR spectroscopy, and physicochemical properties were utilized for the characterization of the synthesized sustainable nanoemulsion. A comparative study was done with different sets of organic-aqueous ratios for the determination of the ratio effect on the various properties of nanoemulsion. The results revealed tremendous improvement in the physicochemical characteristics and antimicrobial activity of nanoemulsions with a 50:50 organic-aqueous phase ratio.

Keywords: *Aloe Vera*, Allyl Alcohol, Semicontinuous, Sustainable, Tremendous, Antimicrobial Activity.

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INTRODUCTION

Nanoemulsions are kinetically stable and can be prepared from water, surfactants, and monomers. They are utilized in various areas such as pharmaceuticals, food industries, cosmetics, paints, coatings, adhesives, and biotechnology.¹⁻⁴ Emulsion polymerization is the most used radical polymerization technology; it offers a number of advantages, including good heat transmission, low viscosity, high monomer conversion, direct application, and a large surface area.⁵ In free radical emulsion polymerization, multifunctional monomers have been investigated for the synthesis of industrial-friendly polymers. The polymerization of allyl alcohol (AA) by radical polymerization is different from the polymerization of vinyl monomers.⁶ Allyl alcohol has long been utilized as an intermediate in various chemical syntheses. Although allyl compounds are abundant in nature, they are synthesized on a wide scale for commercial use.^{7,8} Methyl methacrylate (MMA) contains two groups such as methyl ester of methacrylic & methacrylate. MMA is easily polymerized in the presence of light and heat to make a large chain of homo & copolymers. MMA has a wide range of applications in dentistry, medicine, paints, coatings, synthetic fingernails, plastics, surgical cement, injection molding, extrusion, and industry.⁹⁻¹³ The radical polymerization of MMA is favored because the ester group can stabilize the radicals.¹⁴ Similarly, Butyl acrylate (BA) is another important acrylic monomer used in the production of nanoemulsion. To achieve the required degree of hardness, it is frequently co-polymerized with MMA.¹⁵ It helps in the copolymerization of reactions because it is hydrophilic in nature.¹⁶ Many reports were available on the synthesis of nanoemulsion via semi-continuous seeded emulsion polymerization of poly butyl acrylate/methyl methacrylate.^{3,17-19} Further, numerous researchers explored AA for the preparation of AA-based nanoemulsions such as AA/allyl acetate, and MMA/allyl alcohol 1,2-butoxylate-block-ethoxylate (AABE).^{19,20} *Aloe Vera* (Family: *Liliaceae*) is widely employed in a variety of sectors, including sunscreens, cosmetics, food, pharmaceuticals, and biopesticides. It shows antioxidant, antibacterial, and anti-inflammatory properties. Vitamins, polysaccharides, amino acids, minerals, hydroxyl anthracene derivatives, and inorganic elements are just a few of the essential components that have been found in *Aloe Vera* in significant quantity.²¹⁻²⁵ Interestingly, it has also been utilized for the preparation of

nanoemulsion via different emulsion polymerization techniques. The *Aloe Vera* nanoemulsions were reported using eugenol, Alginate for oral insulin delivery, carbomer 940/isopropyl alcohol emulgel, and *Aloe Vera* mucilage- ethyl vanillin.²⁶⁻²⁹ Given the above, this study aimed to synthesize sustainable antimicrobial copolymer nanoemulsion of *Aloe Vera* and AA/BA/MMA by emulsion polymerization. Further, its antimicrobial activity was assessed against MTCC 442, MTCC 1144, MTCC 2474 & MTCC 40).

EXPERIMENTAL

Materials

Aloe Vera leaves were collected from Gurukula Kangri (Deemed to be University). Sodium Lauryl Sulfate [SLS], span eighty, and sodium hydrogen carbonates [NaHCO₃] were purchased from E. Merck. Allyl alcohol [AA], butyl acrylate [BA], methyl methacrylate [MMA], acrylic acid & potassium per sulfate were purchased from Sigma-Aldrich for commercial sources.

Pre-emulsification of Monomer

The desired quantity of AA, MMA, concentrated form of *Aloe Vera* gel prepared by Payal *et. al.*, BA, and acrylic acid given in Table-1 were added into the mixture of a precise amount of DI water, SLS, & Span eighty solutions. After that suspension was stirred at 25°C for about 40 min to obtain a stable pre-emulsion.

Synthesis of Sustainable Nanoemulsion

Several batches were carried out using the continuous seeded emulsion polymerization technique for the synthesis of *Aloe Vera* nanoemulsion.³⁻⁵ Table-1 represents the composition of the ingredients used for the preparation of nanoemulsion. The polymerization process was carried out in 1L of borosilicate glass resin reaction kettle. The anionic emulsifier (12.5 g), buffer NaHCO₃ (2.5 g), and water (232.5 g) were added. Initially, 5% of the pre-emulsifier was added for 15 min at 80°C. After that, 2.5 g of the initiator K₂S₂O₈ was added with constant stirring. The 95% of pre-emulsion was continuously added drop by drop to the reaction kettle for 1hr at 80°C temperature. After completion of pre-emulsion, the reactor temperature was kept constant at 85°C, and the reaction was carried out for up to 60 min for termination of all radicals. The resulting reaction mixture was again stirred at fifty degrees Celsius for sixty minutes, then the reaction assembly was cooled to room temperature. A milky-colored nanoemulsion was obtained and fixed the pH value was in the range of 6-9 by ammonia solution.

Table-1: Protocol for the 50:50 (Organic: Aqueous Phase) Ratio Nanoemulsion

Ingredients		Quantity in gram
Pre-emulsion	Conc. <i>Aloe Vera</i>	50.0
	AA	10.0
	MMA	15.0
	BA	50.0
	Acrylic acid	5.0
	Deionized water	105.0
	SLS	12.5
	Span 80	7.5
Reactor	SLS	12.5
	NaHCO ₃	2.5
	Deionized water	232.5
Initiator addition-1	Potassium persulfate	1.25
	Water	5.0
Initiator addition-2	Potassium persulfate	1.25
	Water	5.0
Neutralisation	Ammonia	Variable

Characterization of the Synthesised *Aloe Vera* Nanoemulsion

The functionality of the synthesized nanoemulsion was investigated by FTIR spectrophotometer (Shimadzu, 8400S) under the scanning range 400-4000 cm⁻¹. At 40°C, ¹H-NMR & ¹³C-NMR spectra (Bruker DPX- 400 MHz) were obtained using d₆-DMSO and CDCl₃ as solvents. X-Ray diffractogram of

sustainable nanoemulsion was recorded from 5°-80° on a 2θ scale at a scanning rate of 2°/min. ImageJ software was used to calculate the particle size from the scanning electron microscopic images of the synthesis nanoemulsions. A differential scanning calorimeter (DSC-60 Shimadzu) was used to determine the glass transition temperature (T_g) of the sustainable nanoemulsion. Analysis was performed in the surrounding of N_2 gas at a scanning rate of 10°C per minute from 30–550°C temperature. Thermal analysis (TGA/DTA) was carried out on TA instruments Q500 TGA in a nitrogen atmosphere. The rate of heating was 5°Cmin⁻¹ from 30°C to 550°C. Systronics-335 pH meter was used to examine the pH of sustainable nanoemulsion at ambient temperature.

Viscosity

The viscosities of all the sustainable nanoemulsions were measured at 50 rpm using a digital Brookfield viscometer at room temperature using a suitable spindle.

Solid Content

The solid content was measured by gravimetric analysis. 8.0 g of nanoemulsion was kept on a glass plate and oven dried at 105±5°C for 3 hr.

Electrolytic Stability

The electrolytic stability was examined using NaCl solution (10%) after keeping the synthesized nanoemulsion in 10% sodium chloride solution for 5 days.

Freeze-Thaw Stability

Freshly, prepared nanoemulsions were stored in a beaker at 4°C for 24 h, then the beaker was stored at 40°C for 24 h. The above process was repeated for nanoemulsion stable up to one freeze-thaw cycle (15 cycles). At various intervals, the stability of the nanoemulsion was noted.

Alkali resistance

For Alkali resistance, the 10gm of nanoemulsion was weighed & kept for 10 days in 10 ml of 10% solution of NaOH at room temperature. After ten days, the nanoemulsions were washed with DI water, weighed after drying and the percentage of alkali resistance was calculated. The results of viscosity, alkali resistance, freeze-thaw stability, electrolytic stability, and solid content of sustainable nanoemulsion were given in Table-2.

Antimicrobial Activity of Sustainable Nanoemulsion

Antimicrobial activity of *Aloe Vera* with AA/BA/MMA nanoemulsions was investigated via agar well diffusion method against G (+ive) and G (-ive) pathogenic *Staphylococcus aureus* (MTCC 1144), *Streptococcus pyogenes* (MTCC 442), *Escherichia coli* (MTCC 40) and *Pseudomonas aeruginosa* (MTCC 2474). Before the agar well diffusion experiments, bacteria were freshly prepared in Mullene Hinton agar. All suspensions were mixed at 37°C for 24 h. All bacteria were uniformly spread on agar plates with the help of cotton swabs. 50μL of nanoemulsion samples were put in plates. Plates were incubated for 24 hr at 37°C. All experiments were performed in sterile conditions. The zone of inhibition diameter was measured to evaluate antibacterial activities.^{30, 31}

RESULTS AND DISCUSSION

NMR Spectroscopy Analysis

Figure-1 represents the proton-NMR spectrum of the prepared '*Aloe Vera*' with AA/BA/MMA nanoemulsion. The presence of -CH₃ proton of MMA & -CH₂ protons of BA in nanoemulsion has been ascertained by the chemical shift (δ) = 0.83 to 2.49 ppm. Further, the chemical shift at δ ~3.68 ppm was due to the presence of methyl protons of the ester group in MMA.³²⁻³⁴ The proton signals at δ =5.32 and 1.0-2.0 ppm are attributed to the proton of vinyl and -CH peak of the aliphatic main chain of AA groups, respectively. The -CH₂OH resonances were observed between 3.0-4.0 ppm.³⁵⁻³⁷ Correspondingly, the ¹³C-NMR spectrum of the *Aloe Vera* with AA/BA/MMA nanoemulsion is shown in Fig.-2. Several carbon signals were attained at δ =65.3 ppm and 20-40 ppm, which confirmed the presence of -CH₂OH and the aliphatic main chain of CH₂ of the AA group.⁵ In the ¹³C NMR spectrum, several resonances centered at δ =65.3 ppm, were associated with the pendant methylene carbons of PAA & BA.^{33, 38} The signal at δ ~13.5,

24.6-30.9 and 40.1 ppm, were due to the different carbons of the BA group such as $-\text{CH}_3$, $-\text{CH}_2$ and CH respectively in sustainable nanoemulsion.³⁹ The carbon signal at $\delta=28.5$ ppm represents the $-\text{CH}_2$ backbone carbon in p(MMA-co-BA-co-acrylic acid). Another poor intensity peak $\delta\sim 44$ ppm corresponds to the CH_2-C backbone carbon of PMMA.⁴⁰ No peak between 130 to 140 ppm was found in ^{13}C -NMR spectra, indicating that the synthesis reaction was done without any unreacted double bonds.⁴¹

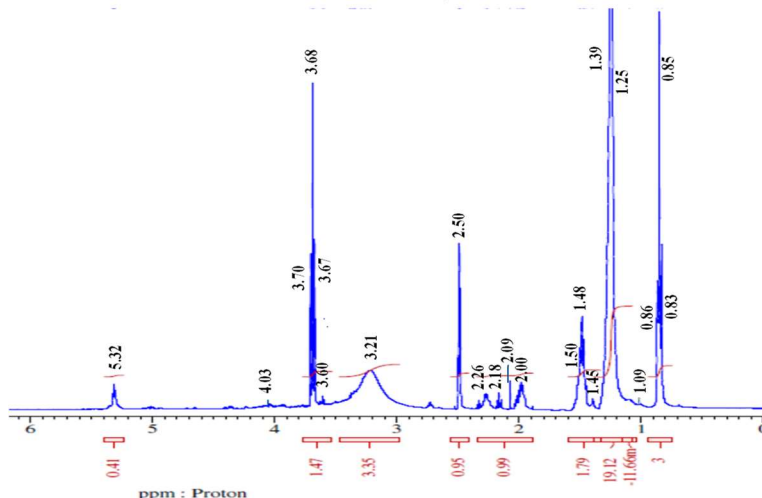


Fig.-1: ^1H -NMR Spectrum of *Aloe Vera* with AA/BA/MMA Nanoemulsion

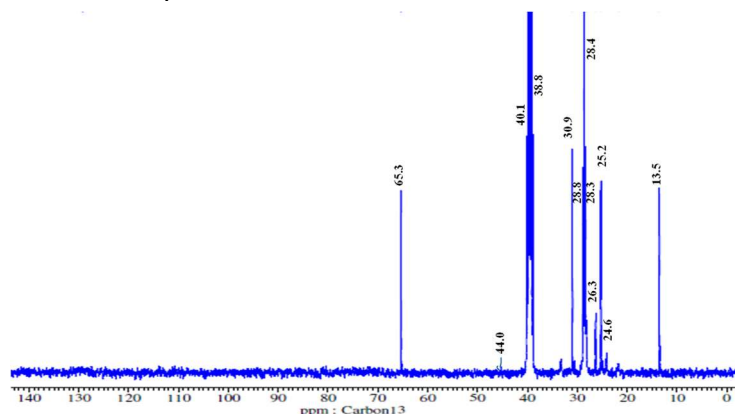


Fig.-2: ^{13}C -NMR Spectrum of *Aloe Vera* with AA/BA/MMA Nanoemulsion

FT-IR Spectroscopy

Different functional groups of *Aloe Vera* with AA/BA/MMA nanoemulsion were further investigated using FT-IR spectroscopy. FTIR spectrum of the *Aloe Vera* nanoemulsion shows in Fig.-3. The FT-IR spectra of *Aloe Vera* with AA/BA/MMA nanoemulsion exhibit the O–H vibrations which are characteristic of AA, such as the O–H stretching at $\sim 3438\text{ cm}^{-1}$ and asymmetric stretching (vibration) of CH_2 at $\sim 2916\text{ cm}^{-1}$. The corresponding $\nu\text{C}-\text{O}$ vibration was observed at $\sim 1049\text{ cm}^{-1}$.⁴² The bending of C–H from the MMA-BA-acrylic acid in copolymer nanoemulsion was attributed to the distinctive peaks at 1365 cm^{-1} .⁴⁰ The C=C stretching causes the strong absorption band at 1596 cm^{-1} , indicating the existence of vinyl ether & *Aloe Vera* extract. The carbon-oxygen vibration of C–O–C linkage from BA, MMA linkage shows a peak at 1049 cm^{-1} and acrylic acid linkage show peaks at 1231 cm^{-1} . The peak at 812 cm^{-1} indicated the presence of =C–H (out-of-plane bending) of the MMA group.⁴³⁻⁴⁵

DSC Analysis

The DSC technique was used to determine the T_g of *Aloe Vera* with AA/BA/MMA nanoemulsion. The thermogram of *Aloe Vera* with AA/BA/MMA nanoemulsion with different ratios has been shown in Fig.-4. The copolymer nanoemulsion revealed more than one T_g . The T_g of p(BA-MMA) in sustainable nanoemulsion was determined by a small endothermic peak at 53°C .^{46,47}

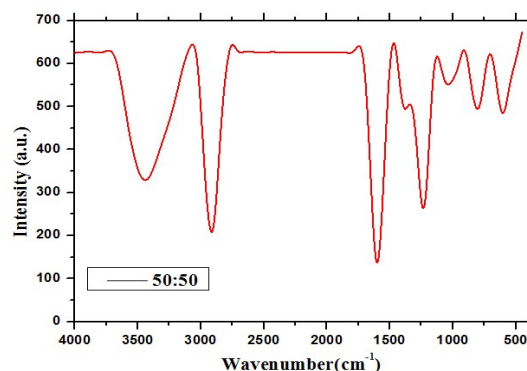


Fig.-3: FT-IR Spectrum of *Aloe Vera* with AA/BA/MMA Nanoemulsion

Similarly, the T_g of MMA from the nanoemulsion was attributed to the broad endothermic peak (119-152°C) centered at 134°C.⁴⁸ In the middle of the graph, the T_g value of *Aloe Vera* allowing from the nanoemulsion was attributed to the sharp peak at 220°C.⁴⁹ Similarly, the broad exothermic peak of (357-427°C) centered at 387°C, which indicates the synthesis of *Aloe Vera* with AA/BA/MMA nanoemulsion.

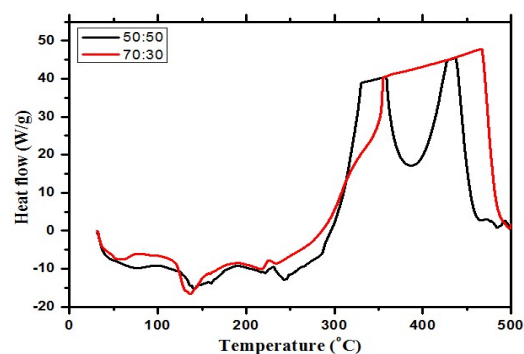


Fig.-4: DSC Thermogram of *Aloe Vera* with AA/BA/MMA Nanoemulsion

Thermal Stability Analysis

TGA & DTA techniques were used to assess the thermal stability of the synthesized *Aloe Vera* with AA/BA/MMA nanoemulsion. The TGA of synthesis of *Aloe Vera* with AA/BA/MMA nanoemulsion is shown in Fig.-5. The thermal degradation of nanoemulsions occurred in three steps over a large temperature range of 30-450°C, demonstrating the copolymer nanoemulsions' remarkable thermal stability. The first step (weight loss) degradation was observed from 30°C to 160°C which shows the breakdown of oxygen, evaporation, and elimination of residual moisture.⁵⁰ The second-step degradation started at 160°C & ended at 320°C. The second phase of degradation was the main stage of degradation. This describes the volatilization of remaining reagents/monomers.⁵¹ The third step of degradation (320-450°C) occurred due to the chain decomposition of PMMA in *Aloe Vera* with AA/BA/MMA nanoemulsion.⁵² Figure-6 shows the DTA thermogram of *Aloe Vera* with AA/BA/MMA nanoemulsions. The endothermic broad peak occurred from 80-140°C with peak maxima at 112°C, attributed to the weak head-to-head links in the main chain. Another exothermic broad peak was observed from 350-480°C. The midpoint of the exothermic peak maxima was observed at 410°C, which goes along with the weight reduction and random scission of the polymer main chain, which started just after the thermal decomposition. Below 200°C, other DTA peaks correspond to the melting and decomposition of non-ionic emulsifiers used in the preparation of nanoemulsions as well as the water absorbed at the surface of the samples.⁵³ Another breakdown peak maximum at 213°C was observed due to the breaking of '*Aloe Vera*', terminal vinyl group decomposition, & linkage of surfactant in sustainable nanoemulsion.⁵⁴

XRD Analysis

The crystalline nature of the *Aloe Vera* with AA/BA/MMA nanoemulsion was examined by XRD. Figure-7 shows the X-Ray diffractogram of the prepared *Aloe Vera* with AA/BA/MMA nanoemulsions. The

diffractogram of PMMA shows a broad peak around $2\theta = 8.01^\circ$ and 14.13° indicating that *Aloe Vera* with AA/BA/MMA nanoemulsion samples is mainly amorphous and low degree of crystallinity in nature.^{55,56} The diffraction peaks of MMA samples^{57,58} was found in the 2θ range of 11.0 – 13.30° . A large peak at $2\theta = 6.53^\circ$ confirmed the p(BA-MMA) in copolymer nanoemulsion. The *Aloe Vera* linkage in nanoemulsion was demonstrated by peaks at $2\theta = 20.19^\circ$, 22.34° , and 24.42° .^{28,59} The diffraction peaks occurring at $2\theta = 7.40^\circ$ in the case of a nanoemulsion with a 70:30 organics: water phase ratio was attributed to the packing of the side and main chains of p(BA).⁶⁰

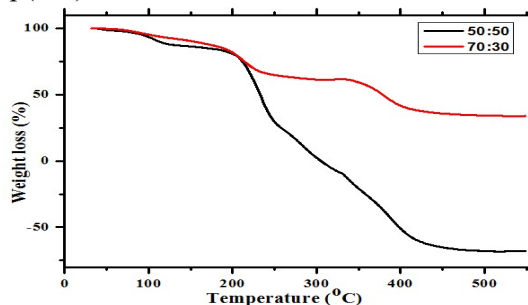


Fig.-5: TGA Thermogram of *Aloe Vera* with AA/BA/MMA Nanoemulsion

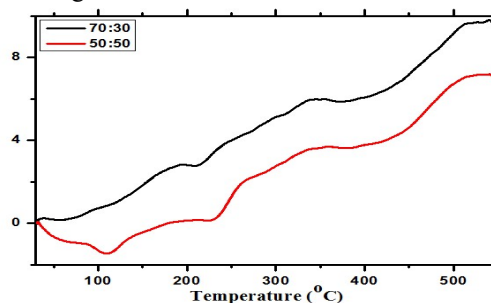


Fig.-6: DTA Thermogram of *Aloe Vera* with AA/BA/MMA Nanoemulsion

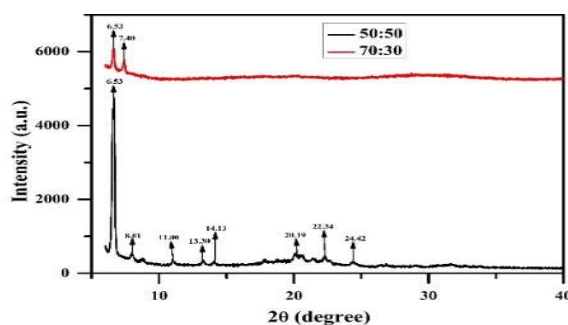


Fig.-7: XRD Pattern of *Aloe Vera* with AA/BA/MMA Nanoemulsion

Surface Morphology and Elemental Composition Analysis

SEM-EDX images were utilized to analyze the surface morphology of synthesized *Aloe Vera* with AA/BA/MMA nanoemulsions at different magnifications, as well as a graph of the weight percentages of various constituents was given in Fig.-8. The *Aloe Vera* with AA/BA/MMA nanoemulsions has a complex morphology, a partially rough outer surface, and irregular, clusters as shown in Fig.-8 (a-c). The prepared nanoemulsions had a size of less than 300 nm, which was recorded to the SEM image with ImageJ software. As a result of the morphological characterization, the oils are more likely to be captured by the inner surface of the nanoemulsion as shown in Fig.-8 (d), and different elemental compositions of synthesized nanoemulsion are displayed, as evidenced by EDX spectra.^{27,61}

Physicochemical Properties of Sustainable Nanoemulsion

Table-2 represents the obtained results of various physicochemical properties like solids content, viscosity, electrolytic stability, alkali resistance, and freeze-thaw stability. It is obvious that the viscosity of the 50:50 organic-aqueous ratio was decreased as the concentration of the aqueous mixed media ratio was also equal.

Coagulation was reduced with the solid content of the 50:50 ratio being higher.^{62,63} Furthermore, the electrolytic stability of the 50:50 proportion is more in comparison to other proportions due to the weaker coagulation power of the synthesized nanoemulsion. The 50:50 ratio also has better freeze-thaw stability, and alkali resistance than other ratios.

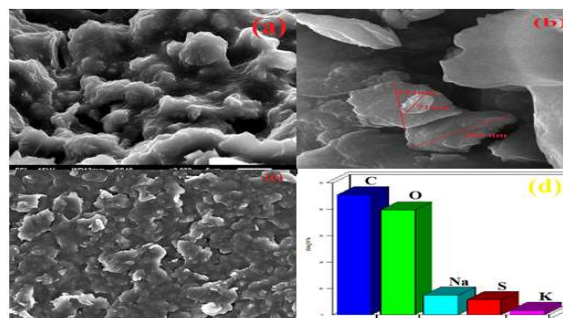


Fig.-8: SEM with EDX of *Aloe Vera* with AA/BA/MMA Nanoemulsion

Table-2: Physicochemical Properties of *Aloe Vera* with AA/BA/MMA Nanoemulsion

Phase ratios	Solid content (%)	pH	Viscosity in Cp	Electrolytic stability	Alkali resistance	Freeze-thaw stability
70:30	45.20	7.20	12.6	No coagulation	01.34g	Stable
60:40	43.90	7.53	18.4	No coagulation	01.60g	Stable
50:50	53.76	7.16	11.2	No coagulation	01.18g	Stable
40:60	18.89	7.51	18.9	Less coagulation	01.36g	moderate Stable
30:70	24.17	8.19	19.6	Less coagulation	01.94g	Less Stable

Antimicrobial Evaluation

Antimicrobial activities of *Aloe Vera* with AA/BA/MMA nanoemulsion were evaluated via agar well diffusion method against G (+ive) and G (-ive) pathogenic. The antimicrobial activity for the different ratios of nanoemulsions was performed by measuring the inhibition zone of diameter. Figure-9 and Table-3 show the inhibition zone images & average inhibition zone respectively. *Aloe Vera* with AA/BA/MMA nanoemulsion was studied for its antimicrobial activity, and average inhibition zones were noted as 20, 20.33, and 22 mm against G (+ive) and G (-ive) bacteria respectively. It is clear that the 50:50 ratio shows high antimicrobial activity towards MTCC 442, MTCC 1144, and MTCC 40.

Table-3: The Zone of Inhibition *Aloe Vera* with AA/BA/MMA Sustainable Nanoemulsion Diameter Against Bacteria's

Bacterial Strains	Zone of inhibition (mm)				
	50:50	70:30	60:40	40:60	30:70
<i>Streptococcus pyogenes</i> (G+) (MTCC 442)	20.00	19.00	18.66	18.33	19.00
<i>Staphylococcus aureus</i> (G+) (MTCC 1144)	20.33	19.33	19.00	19.33	18.33
<i>Pseudomonas aeruginosa</i> (G-) (MTCC 2474)	19.33	19.33	19.00	18.66	19.33
<i>Escherichia coli</i> (G-) (MTCC 40)	22.00	18.66	19.33	18.33	18.66

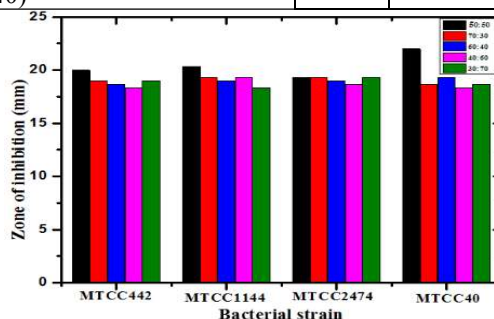


Fig.-9: Antimicrobial Activity of *Aloe Vera* with AA/BA/MMA Sustainable Nanoemulsion

CONCLUSION

The sustainable *Aloe Vera* with AA/BA/MMA nanoemulsions was successfully synthesized. The structural characteristics of the sustainable nanoemulsions were determined using ^1H and ^{13}C NMR, XRD, and FT-IR. Similarly, the surface shape, elemental composition, and thermal properties of nanoemulsion were validated by SEM, EDX, DSC, DTA, and TGA. The SEM images of the *Aloe Vera* with AA/BA/MMA nanoemulsions confirm the complex morphology, the partially rough outer surface, and irregular, clusters. In comparison to others, the 50:50 ratio revealed a significant enhancement in the physicochemical properties of sustainable nanoemulsion. The antimicrobial activity of 50:50 nanoemulsions shows excellent antimicrobial activities against two G (+ive) bacteria: MTCC 442 & MTCC1144 and as well as G (-ive) bacteria: MTCC40.

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CONFLICT OF INTERESTS

The authors declare that there is no conflict of interest.

AUTHOR CONTRIBUTIONS

All the authors contributed significantly to this manuscript, participated in reviewing/editing, and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

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REFERENCES

1. N. Sadurn, C. Solans, N. Azemar, M.J. García-Celma, *European Journal of Pharmaceutical Sciences*, **26**(5), 438(2005), <https://doi.org/10.1016/j.ejps.2005.08.001>
2. J. Singh, R.D. Kaushik, P. Devi, P. Das, *Rasayan Journal of Chemistry*, **13**(1), 761(2020), <http://dx.doi.org/10.31788/RJC.2020.1315563>
3. J. Singh, *Plastic coatings for electronics: Mechanism, Function, Manufacturing technology and Applications*, Lambert Academic Publishing, (2018).
4. J. Singh, M. Chawla, K. Rawat, O. Singh, R.D. Kaushik, *ESSENCE International Journal for Environmental Rehabilitation and Conservation*, **IX**(1), 46(2018), <https://doi.org/10.31786/09756272.18.9.1.107>
5. R.M. Wang, J.F. Wang, X.W. Wang, Y.F. He, Y.F. Zhu, M.L. Jiang, *Progress in Organic Coating*, **71**(4), 369(2011), <https://doi.org/10.1016/j.porgcoat.2011.04.007>
6. D.G.H. Ballard, W.H. Janes, T. Medinger, *Journal of the Chemical Society B: Physical Organic*, 1168(1968), <https://doi.org/10.1039/J29680001168>
7. G. Shao-Hua, *ACS Symposium Series American Chemical. Society*, **755**, 147(2000), <https://doi.org/10.1021/bk-2000-0755.ch011>
8. S.H. Guo, *ACS Symposium Series American Chemical. Society*, **713**, 113(1998), <https://doi.org/10.1021/bk-1998-0713.ch007>
9. H. Soleimani, R. Bagheri, A. Asadinezhad, *Progress in Organic Coating*, **129**, 278(2019), <https://doi.org/10.1016/j.porgcoat.2019.01.019>

10. K. Nagai, *Applied Catalysis A: General*, **221(1-2)**, 367(2001), [https://doi.org/10.1016/S0926-860X\(01\)00810-9](https://doi.org/10.1016/S0926-860X(01)00810-9)
11. P.A. Leggat, U. Kedjarune, *International Dental Journal*, **53(3)**, 126(2003), <https://doi.org/10.1111/j.1875-595X.2003.tb00736.x>
12. P.A. Leggat, D.R. Smith, U. Kedjarune, *Archives of Environmental & Occupational Health*, **64(3)**, 207(2009), <https://doi.org/10.1080/19338240903241291>
13. J. Borak, C. Fields, L.S. Andrews, M.A. Pemberton, *Critical Reviews in Toxicology*, **41(3)**, 230(2011), <https://doi.org/10.3109/10408444.2010.532768>
14. D. Chang, X. Wang, C. Zhu, A. Dong, G. Gao, *Colloids and Surfaces B: Biointerfaces*, **163**, 402(2018), <https://doi.org/10.1016/j.colsurfb.2018.01.013>
15. S. Srivastava, *Designed Monomers and Polymers*, **12(1)**, 1(2009), <https://doi.org/10.1163/156855508X391103>
16. C. Plessis, G. Arzamendi, J.R. Leiza, H.A.S. Schoonbrood, D. Charmot, J.M. Asua, *Macromolecules*, **33(14)**, 5041(2000), <https://doi.org/10.1021/ma992053a>
17. Y.Y. Liu, M.J. Yang, *Chinese Chemical Letters*, **14(8)**, 863(2003).
18. P.L. Tang, E.D. Sudol, M. Adams, M.S. El-Aasser, J.M. Asua, *Journal of Applied Polymer Science*, 42(7), 2019(1991), <https://doi.org/10.1002/app.1991.070420728>
19. M. Eren, H.K. Can, *Progress in Organic Coating*, **135**, 424(2019), <https://doi.org/10.1016/j.porgcoat.2019.06.031>
20. C. Yagci, U. Yildiz, *European Polymer Journal*, **41(1)**, 177(2005), <https://doi.org/10.1016/j.eurpolymj.2004.08.008>
21. F. Habeeb, E. Shakir, F. Bradbury, P. Cameron, M.R. Taravati, A.J. Drummond, A.I. Gray, V.A. Ferro, *Methods*, **42(4)**, 315(2007), <https://doi.org/10.1016/j.ymeth.2007.03.004>
22. Y. Hu, J. Xu, A.Q. Hu, *Journal of Agricultural and Food Chemistry*, **51**, 7788(2003), <https://doi.org/10.1021/jf034255i>
23. E.V. Christaki, P.C. Florou-Paneri, *Journal of Food, Agriculture & Environment*, **8(2)**, 245(2010).
24. M.S. Moghaddasi, S.K. Verma, *International Journal of Biological Sciences*, **2(1)**, 466(2011).
25. T. Mulu, F. Teshale, S. Gemed, O. Sahu, *World Journal of Nutrition and Health*, **3**, 1(2015).
26. S. Sharifimehr, N. Soltanizadeh, S.A.H. Goli, *Journal of the Science of Food and Agriculture*, **99**, 3604(2019), <https://doi.org/10.1002/jsfa.9581>
27. S.K. Basha, M.S. Muzammil, R. Dhandayuthabani, V.S. Kumari, *Materials Today: Proceedings*, **36(2)**, 357 (2021), <https://doi.org/10.1016/j.matpr.2020.04.138>
28. M. Manna, A. Rudra, *GSC Biological and Pharmaceutical Sciences*, **12(2)**, 161(2020), <https://doi.org/10.30574/gscbps.2020.12.2.0262>
29. R. Geetanjali, V. Sreejit, P. Sandip, R. Preetha, *Materials Today: Proceedings*, **43**, 3766(2021), <https://doi.org/10.1016/j.matpr.2020.10.990>
30. S. Anjum, A. Gupta, D. Sharma, D. Gautam, S. Bhan, A. Sharma, B. Gupta, *Materials Science and Engineering: C*, **64**, 157(2016), <https://doi.org/10.1016/j.msec.2016.03.069>
31. J. Singh, M. Chawla and R.D. Kaushik, *Journal of the Indian Chemical Society*, **99(4)**, 1(2022), <https://doi.org/10.1016/j.jics.2022.100408>
32. S. Popa, G. Mosoarca, L. Macarie, N. Plesu, G. Ilia, M. Tara-Lunga-Mihali, *Polymer Bulletin*, **79(2)**, 763(2022), <https://doi.org/10.1007/s00289-020-03502-y>
33. S. Mishra, J. Singh, V. Choudhary, *Journal of Applied Polymer Science*, **115(1)**, 549(2010), <https://doi.org/10.1002/app.30963>
34. P. Devi, J. Singh, H. Rehman, R.D. Kaushik, *Journal of Polymer Research*, **29**, 511(2022), <https://doi.org/10.1007/s10965-022-03339-1>
35. O.S. Joon, D.R. Kinney, W. Wang, P.L. Rinaldi, *Macromolecules*, **35(7)**, 2602(2002), <https://doi.org/10.1021/ma012053m>
36. K. Iio, K. Kobayashi, M. Matsunaga, *Polymers for Advanced Technologies*, **18(12)**, 953(2007), <https://doi.org/10.1002/pat.870>
37. A. Bozzi, C. Perrin, S. Austin, F.A. Vera, *Food Chemistry*, **103(1)**, 22(2007), <https://doi.org/10.1016/j.foodchem.2006.05.061>
38. M.B. Larsen, S.J. Wang, M.A. Hillmyer, *Journal of the American Chemical Society*, **140(38)**, 11911(2018), <https://doi.org/10.1021/jacs.8b07542>

39. N.T. McManus, A. Penlidis, *Journal of Applied Polymer Science*, **103**(4), 2093(2007), <https://doi.org/10.1002/app.24469>
40. R. Shanti, A.N. Hadi, Y.S. Salim, S.Y. Chee, S. Ramesh, K. Ramesh, *RSC advances*, **7**(1), 112(2017), <https://doi.org/10.1039/C6RA25313J>
41. N. Phukon, C.R. Haramagatti, P. Jha, V. Dave, *Journal of Polymer Research*, **20**(10), 1(2013), <https://doi.org/10.1007/s10965-013-0268-4>
42. A. Fahmy, D. Debarnot, J. Friedrich, *Journal of Adhesion Science and Technology*, **29**(10), 965(2015), <https://doi.org/10.1080/01694243.2015.101136>
43. K.S. Joshy, C.P. Sharma, N. Kalarikkal, K. Sandeep, S. Thomas, L.A. Pothen, *Materials Science and Engineering: C*, **66**, 40(2016), <https://doi.org/10.1016/j.msec.2016.03.031>
44. P. Pi, W. Wang, X. Wen, S. Xu, J. Cheng, *Progress in Organic Coating*, **81**, 66(2015), <https://doi.org/10.1016/j.porgcoat.2014.12.006>
45. A. Ratcha, B. Yoosuk, S. Kongparakul, *Advanced Materials Research*, **844**, 385(2014), <https://doi.org/10.4028/www.scientific.net/AMR.844.385>
46. H. Yu, J. Peng, M. Zhai, J. Li, G. Wei, J. Qiao, *Radiation Physics and Chemistry*, **76**(11-12), 1746(2007), <https://doi.org/10.1016/j.radphyschem.2007.02.088>
47. Z.A. Merih, A.S. Sarac, *Journal of Applied Polymer Science*, **130**(6), 4264(2013), <https://doi.org/10.1002/app.39705>
48. M. Monroy-Barreto, N. Esturau-Escofet, M. Briseño-Terán, M.C. Pérez-Vázquez, *International Journal of Polymer Analysis and Characterization*, **17**(7), 515(2012), <https://doi.org/10.1080/1023666X.2012.704556>
49. A. Ahmad-Sekak, M.R. Ahmad, T.J. Bustami Effendi, *Proceedings of the International Colloquium in Textile Engineering, Fashion, Apparel and Design 2014 (ICTEFAD 2014)*, 7-11(2014), https://doi.org/10.1007/978-981-287-011-7_2
50. S. Sharma, M.K. Poddar, V.S. Moholkar, *Ultrasonics Sonochemistry*, **36**, 212(2017), <https://doi.org/10.1016/j.ultsonch.2016.11.029>
51. Z.H. Chen, F. Yu, X.R. Zeng, Z.G. Zhang, *Applied Energy*, **91**(1), 7(2012), <https://doi.org/10.1016/j.apenergy.2011.08.041>
52. F. Wang, Y. Zhang, X. Li, B. Wang, X. Feng, H. Xu, Z. Mao, X. Sui, *Carbohydrate Polymer*, **234**, 115934(2020), <https://doi.org/10.1016/j.carbpol.2020.115934>
53. M. Pishvaei, T.F. Farshchi, *Iranian Polymer Journal*, **19**(9), 707(2010).
54. M. Abbasian, N.K. Aali, S.E. Shoja, *Journal of Macromolecular Science, Part A*, **50**(9), 966(2013), <https://doi.org/10.1080/10601325.2013.813814>
55. M.S. Gaur, P.K. Singh, A.P. Indolia, P.K. Yadav, A.A. Rogachev, A.V. Rogachev, *Ferroelectrics*, **510**, 56(2017), <https://doi.org/10.1080/00150193.2017.1327280>
56. R. Kumar, S.A. Ali, P. Singh, U. De, H.S. Virk, R. Prasad, *Nuclear Instruments and Methods in Physics Research*, **269**, 1755(2011), <https://doi.org/10.1016/j.nimb.2010.12.025>
57. S. Ahmad, S. Ahmad, S.A. Agnihotry, *Bulletin of Materials Science*, **30**, 31(2007), <https://doi.org/10.1007/s12034-007-0006-9>
58. S. Husseinsyah, N.L.I. Zailuddin, A.F. Osman, C.L. Li, A.A. Alrashdi, A. Alakrach, *Polymer*, **12**, 2618(2020), <https://doi.org/10.3390/polym12112618>
59. Y.S. Choi, I.J. Chung, *Macromolecular Research*, **11**, 425(2003), <https://doi.org/10.1007/BF03218971>
60. K. Matsuura, K. Kuboyama, T. Ougizawa, *Journal of Applied Polymer Science*, **138**, 50268 (2021), <https://doi.org/10.1002/app.50268>
61. X. Wu, J. Li, J. Wang, L. Cao, *International Journal of Industrial Chemistry*, **11**, 91(2020), <https://doi.org/10.1007/s40090-020-00204-2>
62. R.D. Kaushik, J. Singh, M. Chawla, *Emulsion of some monomers for application in paint technology*, Lambert Academic Publishing, 1-76 (2018).
63. J. Singh, M. Chawla and R.D. Kaushik, *ESSENCE International Journal for Environmental Rehabilitation and Conservation*, **9**(1), 55(2018), <https://doi.org/10.31786/09756272.18.9.1.108>

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