

Quantitative Analysis on the Wettability of *Datura Stramonium* Leaves in the Presence of Adjuvants

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Abstract

Datura Stramonium is an indigenous plant in Nepal but is globally cultivated for its wide medicinal applications. The knowledge of the surface properties of leaves and wettability conditions is essential for foliar applications to protect the plants. The wettability of the leaf surface is characterized by the contact angle (CA) measurement, and surface free energy (SFE) directs its wettable interactions. Three different surfactants having different charges; N-Cetyl-N, N, N-Trimethyl Ammonium Bromide (cationic), bis(2-ethylhexyl) sodium sulfosuccinate (anionic) and polyoxyethylene lauryl ether (Brij-L23) (non-ionic) and two commercial bio-wetting agents Gorkha stick more (Nepal) and Keeper (India) were utilized as adjuvants to enhance the wettability of leaves towards super-hydrophilic range (CA < 40°). The secondary parameters like molar free energy (ΔG), wetting free energy (Δg), adhesion tension (T_a), adhesion work (W_a), cohesive energy density (e_c), and solubility parameter difference ($\Delta\delta$) were calculated to describe the surface properties of leaves and efficiency of adjuvants in wettability. The CA value of the adaxial leaf surface was in the hydrophilic range (< 90°) while it was in the hydrophobic range on the abaxial surface (> 90°). The SFE of the adaxial leaf surface was $44.42 \pm 16.52 \text{ mN m}^{-1}$ whereas the abaxial surface had a SFE of $36.82 \pm 1.57 \text{ mN m}^{-1}$. The AOT solution showed better results among all surfactants and commercially available wetting agents by reducing CA value to the super-hydrophilic range on both adaxial and abaxial leaf surfaces below CMC. The effectiveness of adjuvants to lower the CA value follows the pattern of AOT > Gorkha stick more > CTAB > Keeper > Brij-35 solutions. The $\Delta\delta$ value of the abaxial leaf surface was higher than the adaxial leaf surface.

Keywords: *Datura Stramonium*; Contact angle; Adjuvants; Surface free energy; Solubility parameter

Introduction

Datura stramonium is a plant in the Solanaceae family, is native to the Americas and is grown globally from Asia, deserts of Europe to Africa with greater abundance in temperate, tropical, and subtropical climates and are seen in roadsides, wastelands [1,2]. *Datura stramonium* is cultivated in South America, Germany, France, Hungary, and worldwide [3]. Its flower, leaves, seeds, and roots, hold a

prominent place in Ayurvedic medicine due to its medicinal importance [4].

In Nepal, it is commonly known as dhaturu. Indian and Chinese people have used *Datura* for medicine for centuries [5] and traditionally, an oral extract from the leaves is used to treat asthma and sinus infections. At the same time, the bark, when stripped, is applied externally to treat swellings, burns, and ulcers [3]. When mustard oil is mixed with its leaves it is used to

treat skin disorders, and flower petal juice is used to treat ear pain [6]. Dried leaves, petals, and roots were employed as narcotics, bronchodilators, asthmatic treatments, and hallucinogens. The herb was also used to treat diarrhea, skin illnesses, epilepsy, hysteria, regional pain syndrome, hemorrhoids, menstruation cramps, skin ulcers, wounds, burns, rheumatoid arthritis, and gout, treatment of hair fall and dandruff [7–9]. In the Karnali zone, crushed seeds containing rice grains are used to relieve dyspepsia [10]. Datura plants contain significant phytochemicals which include alkaloids, flavonoids, phenols, cardiac glycosides, tannins, saponins, amino acids, and carbohydrates [9]. Their root contains more than fifty percent palmitic acid which is used for making different antimicrobial Anti-inflammatory, analgesic, and antipyretic drugs [7].

Due to its large medicinal uses, it is cultivated throughout the world and it also gets affected by various bacteria, fungi, and viruses [11–15]. To overcome these diseases, one should apply agrochemicals and nutrients either through soil application or through foliar application. Foliar application is better than soil application because it uses a lesser quantity of agrochemical in comparison to soil application that helps to reduce environmental pollution caused by the use of different insecticides and pesticides [16]. Depending on the variety of soil, foliar application might be six to twenty times more effective than soil application [17,18].

For better efficiency of foliar application, it is necessary to understand the wettability character of leaf surfaces which is characterized by contact angle (CA) between the water droplets and the leaf surfaces. The CA ranges from 0° to 180°. In the context of water, a material can be classified as hydrophilic if its CA is less than 90 degrees, hydrophobic if it is between 90 and 150 degrees, and superhydrophobic if it is greater than 150 degrees [19,20]. In the case of leaves, Wang et al. described the super-hydrophilic

range if $CA < 40^\circ$, the wettable range if CA is between 40 to 110° , the non-wettable range if $CA > 110^\circ$ and the superhydrophobic range for $CA > 150^\circ$ [21]. Different contact angle values result from variations in the physical and chemical characteristics of the leaf surface. The density, relative wetness, trichome pattern, and surface roughness are the causes of it [22]. Surfactants are the adjuvants that reduce the surface tension disturbing the cohesive forces of liquid which ultimately enhances the spreading behavior over the surfaces reducing the contact angle and enhancing the wettability [23]. There is no literature known regarding the wettability character of datura leaves in terms of contact angle. So, this is the first attempt to detect the wettability character and the surface property based on dispersive and polar components of the datura leaf surfaces, and solubility parameters. Similarly, the comparative study of three different surfactants and two commercial wetting agents to reduce the CA value on datura leaf surfaces will be the initial work in the climate of Nepal and this work will directly benefit the agro-industries of Nepal for the formulation of wetting agents and other agrochemicals like insecticides, pesticides, micro-nutrients, etc., and benefits to the farmers during foliar applications of those agrochemicals.

Materials and Methods

Materials

A thermostat water bath purchased from Orbit Company, India for temperature management; Sartorius digital balance of four digits after the decimal (capacity = 220 g, accuracy = 0.1 mg), Germany for weighing; Eutech pH meter 2700, Thermo Fisher Scientific for pH measurement; Kruss drop shape analyzer, Germany donated by TWAS for contact angle, surface tension and surface free energy measurement were utilized for this work.

Diiodomethane and Ethylene glycol were of Sigma Aldrich quality, polyoxyethylene lauryl

ether (Brij-L23) was purchased from Merck, India, Dioctyl sodium sulphosuccinate (AOT), and N-Cetyl-N, N, N-Trimethyl Ammonium Bromide (CTAB) were purchased from Loba Chemie, India. Two commercial bio-wetting agents; Keeper, SAGA Biotech Pvt. Ltd., Uttar Pradesh, India, and Gorkha stick more, Gorkha Agro Chemicals Co. Pvt. Ltd., Dang, Nepal were purchased from the local shop, Biratnagar, Nepal.

Methods

Stock solutions of AOT, CTAB, and Brij-35 were prepared both below and above their CMC values using double distilled water. The CMC values of AOT, CTAB, and Brij-35 solutions in water are $0.00235 \text{ mol L}^{-1}$ [24], $0.0009 - 0.001 \text{ mol L}^{-1}$ [25,26] and $40 - 90 \mu \text{ mol L}^{-1}$ [27–29], respectively. The optimum concentrations of Keeper and Gorkha stick more was 1 ml per liter as suggested in the products. So, we have prepared the solutions below and above this optimum concentration.

Both adaxial and abaxial leaf surfaces of *D. stramonium* were cut into small portions avoiding midrib and were placed in the sample chamber using double sided tap. The sessile drop method was utilized to calculate the CA and the OWRK method was adopted using three series of liquids to calculate dispersive, polar components and surface free energy of the leaf surfaces. These data were analyzed through ADVANCE software version 1.9.0.8.

Results and Discussion

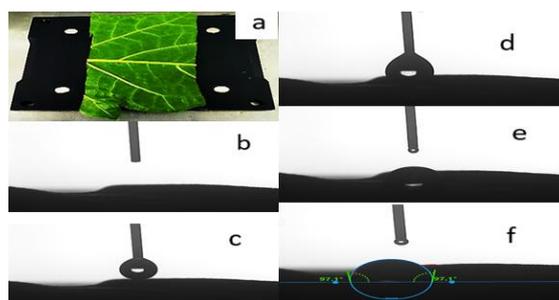
Contact angle measurement

The measurements of CA of water, surfactant, and wetting agent solutions are carried out five times for different leaves and the three best measurements are tabulated in **Tables 1, 2, 3, 4, and 5**. The contact angle of water was detected to be $65 \pm 5^\circ$ on the adaxial leaf surface and $95 \pm 5^\circ$ on the abaxial leaf

surface of *D. stramonium*.

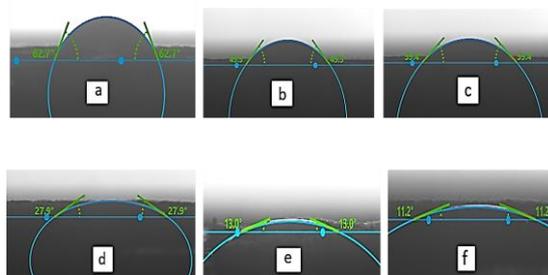
Table 1: CA of AOT solutions on the adaxial and abaxial leaf surfaces of *D. stramonium* at $25 \pm 2^\circ \text{C}$.

[AOT] mol/L	Contact angle ($^\circ$)					
	Adaxial			Abaxial		
	θ_1	θ_2	θ_3	θ_1	θ_2	θ_3
0.00	69.10±1.99	67.81±3	62.81±2.74	91.56±7.16	91.08±5.82	100±4.6
0.0009	47.65±4.21	46.67±7.94	50.48±3	66.67±1.95	63.44±1.80	63.44±1.10
0.001	25.20±5.49	24.52±3.67	25.71±4.75	35.78±7.8	39.71±1.44	37.70±1.87
0.0015	15.54±6.34	15.66±5.56	16.09±5.36	26.65±5.42	28.23±1.90	30.86±2.76
0.002	11.68±2.27	12.17±2.13	10.70±1.64	18.64±1.33	17.78±1.48	20±1.22



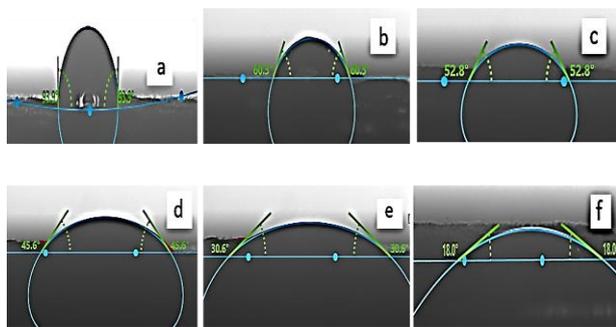
Scheme 1: Picture of undisturbed abaxial leaf surfaces and screenshots of the stepwise dropping of water over that surface taken through a DSA Camera.

The CA value on the large portion of the undisturbed leaf surfaces was detected in the same range as measured on the small strips. The undisturbed abaxial leaf and the screenshots of the stepwise dropping of water droplets on the surface have been depicted in **Scheme 1**. The measurement results showed that the adaxial leaf surfaces lie in the hydrophilic range and the abaxial leaf surface lies in the hydrophobic range [19,20,30,31]. Our aim is to reduce the contact angle of water below 40° so that it will be in the super hydrophilic range [21].



Scheme 2: Screenshot of images of CA of water and minimum CA value of adjuvant solutions on adaxial leaf surface of *D. stramonium* during measurement; (a) water (b) Brij-35, $110 \mu \text{ mol L}^{-1}$ (c) Keeper, 2 ml L^{-1} (d) CTAB, 0.002 mol L^{-1} (e) Gorkha stick more, 2 ml L^{-1}

and (f) AOT, 0.002 mol L⁻¹.



Scheme 3: Screenshot of images of CA of water and minimum CA value of adjuvant solutions on abaxial leaf surface of *D. stramonium* during measurement; (a) water (b) Keeper, 2 ml L⁻¹ (c) Brij-35, 110 μ mol L⁻¹ (d) CTAB, 0.002 mol L⁻¹ (e) Gorkha stick more, 2 ml L⁻¹ and (f) AOT, 0.002 mol L⁻¹.

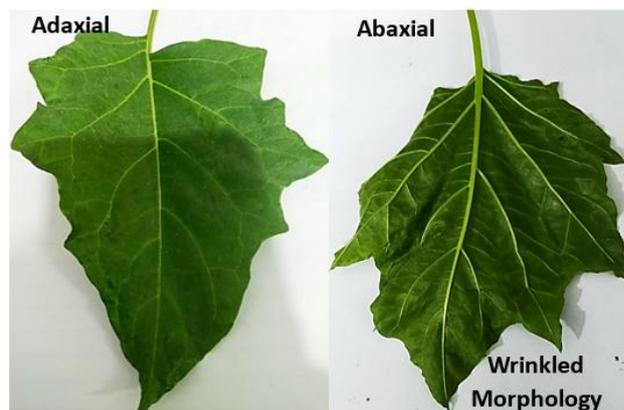
The screenshot images of CA of water and adjuvant solutions with minimum CA value on the adaxial and abaxial leaf surfaces are depicted in **Schemes 2** and **3** where curve fitting was done with the Young-Laplace method. The adaxial surface of *D. stramonium* is smooth while the abaxial surface is rough due to wrinkled morphology as depicted in **Scheme 4**, and such roughness is described in the literature [1].

Table 2: CA of CTAB solutions on the adaxial and abaxial leaf surfaces of *D. stramonium* at 25 ± 2 °C.

[CTAB] mol/L	Contact angle (°)					
	Adaxial			Abaxial		
	01	02	03	01	02	03
0.00	69.10 ±1.99	67.81±3	62.81±2 .74	91.56±7.16	91.08±5.82	100±4.6
0.0006	65.08±4.04	61.23±1.43	62.40±4.56	74.66±2.39	67.31±2.39	73±5.76
0.0007	52.5±1.96	54.51±1.92	53.21±2.48	66.47±2.10	70.76±1.17	68.74±1.4
0.0008	53.82±0.53	50.87±1.33	51.74±1.08	67.38±5.53	61.22±2.98	66.35±1.89
0.0009	46.91±1.39	45.84±1.28	43.06±3.78	59.97±5.13	61.34±1.10	58±2.86
0.0010	41±3.16	40.92±4.29	40.60±0.93	58.06±2.96	55.37±0.89	57.88±1.8
0.0015	38.85±1.49	37.06±2.52	39.53±3.73	55.58±1.07	49.90±4.07	50.65±1.9
0.002	30.53±5.63	26.92±2.57	24.57±2.88	50.86±5.41	53.62±5.12	44.64±5.62

The density of stomata is higher on the abaxial surface than adaxial one [32]. It is seen that both surfaces contained short and small trichomes. However, the glandular trichomes: the unicellular head with bicellular stalk and multicellular head with unicellular stalk, were absent on the adaxial surface but present on the abaxial surface. Similarly, rough non-glandular trichomes: unicellular and tetra-cellular were absent on the adaxial surface but present on the

abaxial surface [33]. It is observed that roughness developed due to heterogeneous surface enhances the CA value [34] as well as higher stomatal density is somewhat responsible for higher CA value [21]. Thus, the heterogeneous roughness due to wrinkled morphology and higher stomatal density account for higher CA values in abaxial surfaces.



Scheme 4: Adaxial and Abaxial leaf surfaces of *Datura stramonium*.

Table 3: CA of Brij-35 solutions on the adaxial and abaxial leaf surfaces of *D. stramonium* at 25 ± 2 °C

[Brij-35] μmol/L	Contact angle (°)					
	Adaxial			Abaxial		
	01	02	03	01	02	03
0	69.10 ±1.99	67.81±3	62.81±2 .74	91.56±7.16	91.08±5.82	100±4.6
20	60.75(±0.55)	63.55(±3.51)	63.45(±1.48)	76.81(±0.6 2)	76.38(±1.3 0)	75.20(±1. 45)
30	55.91(±2.63)	59.69(±3.55)	60.42(±2.06)	68.75(±2.0 3)	67.32(±1.30)	66.32(±1.53)
40	56.73(±1.82)	56.05(±1.76)	55.25(±3.32)	65.46(±2.86)	64.57(±2.11)	64.22(±0.54)
60	54.00(±3.96)	49.07(±3.87)	50.04(±1.79)	63.96(±2.31)	62.82(±1.04)	64.88(±1.20)
80	50.81(±1.10)	49.20(±1.47)	49.51(±3.29)	60.67(±0.3 0)	61.57(±0.60)	61.41(±0.46)
90	47.03(±2.54)	46.69(±2.43)	48.27(±3.84)	59.22(±1.44)	60.12(±0.38)	58.48(±0.75)
100	43.76(±0.41)	49.18(±1.73)	45.81(±1.23)	55.10(±0.32)	58.05(±0.43)	57.03(±0.80)
110	44.08(±0.30)	43.29(±2.10)	47.06(±0.65)	55.39(±1.83)	52.88(±3.67)	56.46(±0.97)

One suitable CA value was taken for each concentration of surfactants and wetting agents from the various measurements and was plotted against concentrations as shown in **Fig.1**. The CA value decreased with increasing concentrations of both surfactants and bio-wetting agent solutions on both adaxial and abaxial leaf surfaces of *D. stramonium*. **Fig.2** depicts the comparative analysis of CA of all surfactants and bio-wetting agent solutions on the adaxial and abaxial surfaces. The AOT solution reduced the CA value in the super-hydrophilic range (< 40°) at 0.001 mol L⁻¹. The CTAB solution reduced the CA in the super-hydrophilic range at 0.0015 mol L⁻¹ in the

adaxial surface but could not reduce below 44° in the abaxial surface up to the concentration of 0.002 mol L⁻¹. It suggested that the CTAB solution should be higher than 0.002 mol L⁻¹ for reducing the CA in the super-hydrophilic range. The contact angle was above 43° in the case of Brij-35 solution in both adaxial and abaxial surfaces at all concentrations below and above CMC. Gorkha stick more solution reduced the CA value in the super-hydrophilic range even at 0.5 ml L⁻¹ at both adaxial and abaxial leaf surfaces while Keeper solution was found to be effective in reducing CA in the super-hydrophilic region in the adaxial part at the concentration of 2.0 ml L⁻¹.

Table 4: CA of Gorkha Stick More (a Nepali sticker) solutions on the adaxial and abaxial leaf surfaces of *D. stramonium* at 25 ± 2 °C.

[Gorkha Stick More]		Contact angle (θ)					
ml/L		Adaxial			Abaxial		
Nepali Sticker		θ1	θ2	θ3	θ1	θ2	θ3
0.00		69.10±1.99	67.81±3	62.81±2.74	91.56±7.16	91.08±5.82	100±4.6
0.5		36.76±1.70	35.76±2.58	35.09±0.66	44.66±5.42	40.96±0.82	40.88±3.09
1.0		28.10±0.27	28.86±2.35	28.95±0.65	38.49±1.08	38.38±1.13	39.05±2.22
1.5		18.16±2.45	19.03±0.88	20.64±1.00	35.73±0.28	37.61±0.23	36.97±0.21
2.0		14.03±0.88	15.16±2.48	13.06±1.45	31.03±0.48	29.61±0.24	31.56±0.23

Table 5: CA of Keeper (an Indian sticker) solutions on the adaxial and abaxial leaf surfaces of *D. stramonium* at 25 ± 2 °C.

[Keeper]		Contact angle (θ)					
ml/L		Adaxial			Abaxial		
		θ1	θ2	θ3	θ1	θ2	θ3
0.00		69.10±1.99	67.81±3	62.81±2.74	91.56±7.16	91.08±5.82	100±4.6
0.5		63.39±1.07	57.59±0.93	64.61±0.77	83.9±1.24	83.90±0.97	82.33±2.33
1.0		51.40±4.11	53.52±2.69	50.67±1.89	71.42±1.00	70.44±1.20	73.3±0.98
1.5		41.32±5.05	44.97±2.33	44.82±0.56	69.79±1.15	65.06±2.34	61.94±4.2
2.0		39.53±1.29	34.83±1.22	36.08±4.5	63.21±1.06	61.39±4.58	62.08±0.77

The pH value of 0.002 mol L⁻¹ AOT solution, 0.002 mol L⁻¹ CTAB solution, 110 μ mol L⁻¹ Brij-35 solution, 2 ml L⁻¹ Gorkha stick more solution, and 2 ml L⁻¹ Keeper solution were found to be 4.4, 7.55, 7.3, 7.00, and 6.72, respectively at 25 °C. AOT solution seems to be more acidic among all adjuvants. Acidic solution disturbs the cohesive force among the molecules in the

water droplet due to dissolved ions [35] thereby enhancing the spreading behavior and reducing CA value.

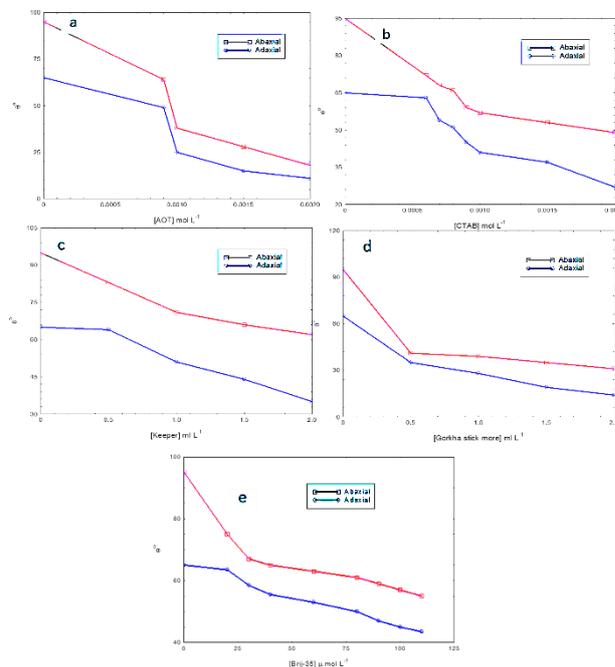


Fig.1. CA vs Concentrations of Adjuvants: (a) AOT, (b) CTAB, (c) Keeper, (d) Gorkha stick more, and (e) Brij-35 solutions on the adaxial and abaxial leaf surfaces of *D. stramonium* at 25 ± 2.

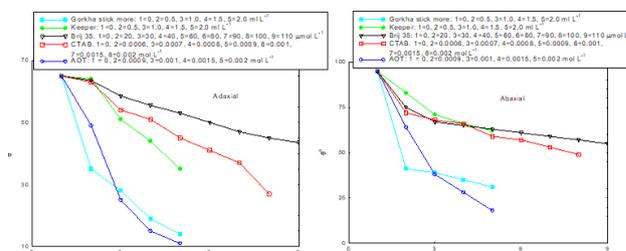


Fig.2. Comparison of CA of Adjuvants at both adaxial and abaxial leaf surface of *D. stramonium*.

The acidic solution also ruptured the waxy composition of the cuticular layer thereby enhancing the absorption of liquid droplets over the surfaces and lowering the CA value of solutions [24].

Molar-free energy and wetting-free energy

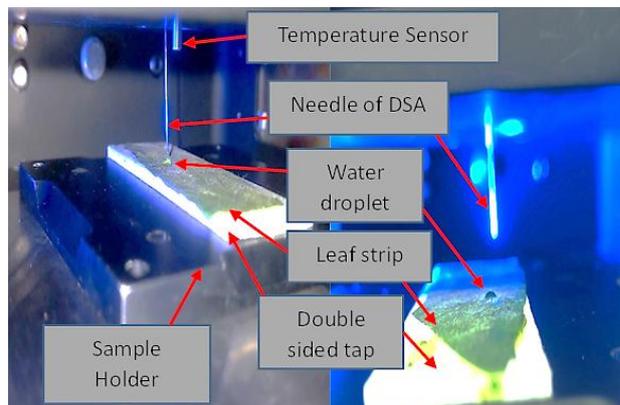
The wetting phenomenon is described in terms of adsorption, and the Gibbs adsorption equation is modified to get molar free energy change (ΔG) with the help of CA (θ) and

temperature (T) as Eq. 1 [36,37]

$$\Delta G = \frac{RT}{3} \ln \frac{(1-\cos \theta)^2(2+\cos \theta)}{4} \quad (1)$$

Where R is a universal gas constant. Eq. 1 is valid for the smooth surface, so we have removed the midrib of the leaf surfaces, avoided the veins as far as possible, and attached over the smooth sample holder with a double-sided tape as shown in **Scheme 5**. Further, the molar surface area of the solid is required to get the wetting free energy (Δg). The Δg value quantifies the intensity of the interactions that draw the liquid downwards onto the surface and is thus regarded as the measure of solid-liquid adhesion. In our experiment, we considered the contact area (A) between the liquid droplet and leaf surface where it was calculated by the diameter of the liquid droplet over the leaf surface. The wetting free energy was calculated as $\Delta g = \Delta G/A$. The negative value of Δg describes the spontaneity of the spreading of liquid and wettability [36]. The literature showed that the wetting free energy is practically zero for CA > 150°, which suggested that CA > 120-130° investigations required the knowledge of geometric structure rather than explanations based on chemical interactions [37,38]. The ΔG inclined towards negative directions with the decreasing CA value as seen in **Fig.3**, and the value of ΔG of the leaf surface is less than 10 kJ mol⁻¹ as tabulated in **Table 6**. The calculated values of ΔG at various CA values (< 120-130°) of surfactant and wetting agent solutions are in the same ranges, and the plot of ΔG vs CA are in a similar pattern as calculated for a smooth surface in the literature [36,37]. This suggests that the estimation of ΔG and Δg are also valid for the leaf surfaces that are prepared to avoid the midribs and veins, which cause major surface roughness. The ΔG value solely depends upon the CA value, so the fluctuation in the CA

value will result in a slight variation in the ΔG value, however, the trend of variation of ΔG with CA will not vary.



Scheme 5: Picture taken from two different angles inside the Sample Chamber of DSA

Table 6: Molar free energy (ΔG) and wetting free energy (Δg) of *D. stramonium* leaf surfaces at various concentrations of AOT, CTAB, Brij-35, Keeper, and Gorkha stick more solutions.

Surfactants	Concentrations	ΔG , kJ mol ⁻¹		Mean diameter of drop (d), mm		Contact surface area (A), mm ²		Δg , J mol ⁻¹ mm ⁻²		
		Adaxial	Abaxial	Adaxial	Abaxial	Adaxial	Abaxial	Adaxial	Abaxial	
[AOT] mol/L	0	-1.32	-0.47	3.25	2.2	8.30	3.80	-159.28	-123.96	
	0.0009	-2.10	-1.36	3.35	2.76	8.81	5.98	-238.37	-227.60	
	0.001	-4.17	-2.86	3.56	3.56	9.95	9.95	-419.40	-287.36	
	0.0015	-5.83	-3.81	3.9	3.86	11.95	11.70	-487.91	-325.89	
	0.002	-6.84	-5.23	4.06	3.94	12.95	12.19	-528.68	-429.33	
	0	-1.32	-0.47	3.25	2.2	8.30	3.80	-159.28	-123.96	
[CTAB] mol/L	0.0006	-1.40	-1.06	3.31	2.34	8.60	4.30	-165.04	-247.49	
	0.0007	-1.82	-1.21	3.45	2.44	9.35	4.68	-195.06	-257.9	
	0.0008	-1.93	-1.28	3.55	2.63	9.90	5.43	-195.01	-255.98	
	0.0009	-2.38	-1.58	3.65	2.78	10.46	6.07	-224.65	-260.06	
	0.001	-2.63	-1.67	3.78	3.02	11.22	7.16	-234.29	-253.84	
	0.0015	-2.94	-1.88	3.89	3.41	11.88	9.13	-247.35	-205.43	
0.002	-3.82	-2.10	3.9	3.52	11.95	9.73	-319.24	-215.90		
[Brij-35] $\mu\text{mol L}^{-1}$	0	-1.32	-0.47	3.25	2.2	8.30	3.80	-159.28	-123.96	
	20	-1.38	-0.97	3.28	2.8	8.45	6.16	-163.58	-186.99	
	30	-1.61	-1.24	3.28	2.9	8.45	6.61	-189.56	-188.26	
	40	-1.75	-1.32	3.29	2.92	8.50	6.70	-208.47	-197.31	
	60	-1.88	-1.40	3.3	2.93	8.55	6.74	-219.36	-208.08	
	80	-2.04	-1.49	3.31	2.94	8.60	6.79	-237.38	-219.28	
	90	-2.22	-1.58	3.32	2.95	8.66	6.83	-256.73	-230.95	
	100	-2.35	-1.67	3.33	2.96	8.71	6.88	-269.90	-243.10	
	110	-2.45	-1.77	3.34	2.97	8.76	6.93	-279.80	-255.78	
	[Gorkha stick more] ml/L	0.5	-3.11	-2.63	3.45	3.17	9.24	7.89	-336.96	-333.13
		1	-3.81	-2.78	3.61	3.2	10.23	8.04	-372.39	-345.78
1.5		-5.06	-3.20	3.84	3.4	11.58	9.08	-436.83	-352.84	
2		-6.05	-3.49	3.96	3.54	12.32	9.84	-491.54	-384.82	
[Keeper] ml/L	0	-1.32	-0.47	3.25	2.2	8.30	3.80	-159.28	-123.96	
	0.5	-1.36	-0.74	3.3	2.5	8.55	4.91	-159.20	-150.39	
	1.0	-1.99	-1.10	3.32	2.72	8.66	5.81	-229.38	-189.04	
	1.5	-2.42	-1.28	3.47	2.84	9.46	6.33	-255.62	-202.37	
	2.0	-3.11	-1.44	3.57	2.99	10.01	7.02	-311.05	-205.84	

The ΔG and Δg values of AOT are highest among all the adjuvants which is followed by Gorkha stick more, CTAB, Keeper, and Brij-35 solutions which validate the CA value seen in the leaf surfaces. The diameter of the liquid drop increased with the increasing concentrations of adjuvants. It is because surfactants reduce the surface tension of the liquid disturbing their

cohesive force which ultimately enhances the surface area or diameter of the droplet.

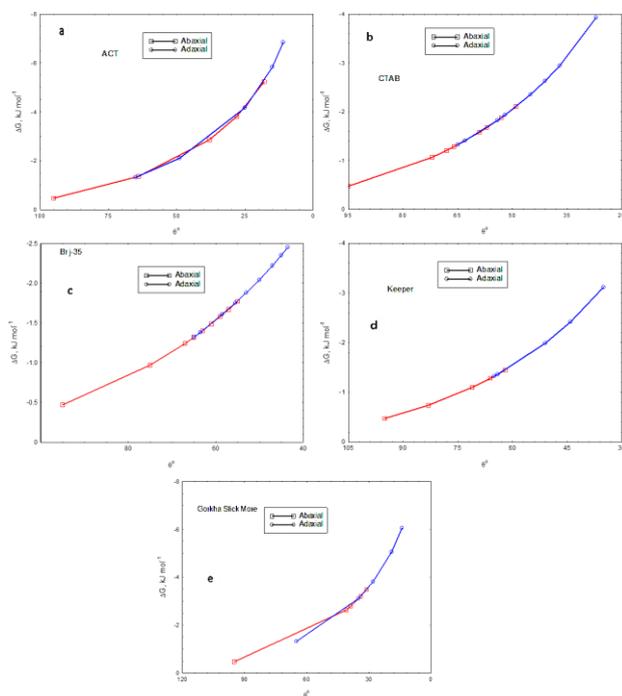


Fig.3. Molar free energy change (ΔG) of *D. stramonium* leaf surfaces vs CA of Adjuvant solutions.

The order of increasing diameter at minimum contact angle of adjuvants follows the order as Brij-35 < Keeper < CTAB < Gorkha stick more < AOT solutions. The negative value of Δg of the abaxial surface is less than that of the adaxial one which suggests that the abaxial surface is less wettable than the adaxial one.

Adhesion Tension (T_a) and Adhesion work (W_a)

The strength of attachment between a liquid droplet and the surface is expressed in terms of surface tension (γ) and contact angle (θ) as $\gamma \cos \theta$, and is known as adhesion tension [39] which is further explained as adhesion work (W_a) as $\gamma(1 + \cos \theta)$ given by Eq.2 [23,40]

$$W_a = \gamma(1 + \cos \theta) \quad (2)$$

Leaf has a unique property where it cleans its surface itself, known as the self-cleaning property and particularly termed as lotus effect where lower adhesion is associated with higher CA value [41–43]. Some leaves oppose this property where higher adhesion is associated with higher CA value and is termed as rose petal

effect [44,45]. Both effects are dependent on the surface composition and structure of the leaves.

Fig.4 depicted that *D. stramonium* possessed only a rose petal effect with all adjuvants at both adaxial and abaxial leaf surfaces. This is practically observed in the environment with dirty leaves even in foggy or highly humid conditions.

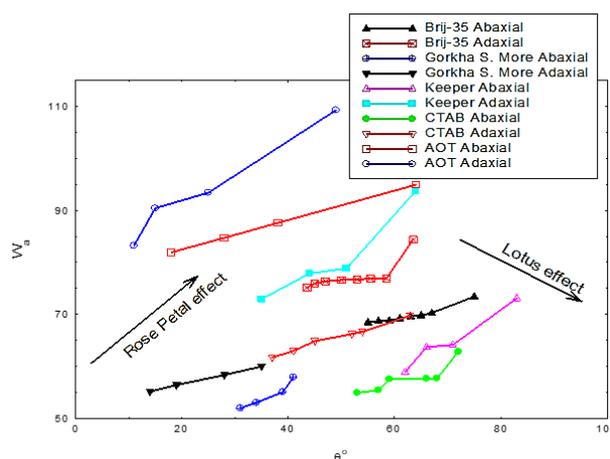


Fig.4. Adhesion work vs CA of Adjuvant solutions at both adaxial and abaxial leaf surfaces of *D. stramonium*.

As the concentrations of adjuvants increased, the number of molecules adsorbed on the leaf surfaces also increased which ultimately enhances the adhesion tension of the liquid which is observed in **Table 7**. Simultaneously, the increasing concentrations of surfactants reduced the surface tension of the liquid. Thus, the combined effect of higher adhesion tension and lower surface tension reduced the CA value which ultimately enhances the wettability of the surface [39].

The adhesion tension values of all adjuvants were higher on the adaxial surface than on the abaxial part. The trichome frequency was higher on the abaxial part than on the adaxial surface [46] which repels the droplets which accounts for lower adhesion on an abaxial portion of the leaves. The adhesion tension values of the AOT solution are higher

among all adjuvants. The leaf mainly contains alkaloids like atropine, scopolamine, and hyoscyamine [47,48] and their structures are depicted in **Fig. 5**.

Table 7. Adhesion Tension and Adhesion Work of different concentrations of Adjuvants on both adaxial and abaxial leaf surfaces.

Adjuvants	Concentrations	Average CA [°]		T _a (mN/m)		W _a (mN/m)	
		Adaxial	Abaxial	Adaxial	Abaxial	Adaxial	Abaxial
[AOT] mol L ⁻¹	0	65	95	30.34	-6.26	102.14	65.54
	0.0009	49	64	43.30	28.93	109.30	94.93
	0.001	25	38	44.41	38.61	93.41	87.61
	0.0015	15	28	44.43	39.73	90.43	84.73
	0.002	11	18	41.23	39.94	83.23	81.94
[CTAB] mol L ⁻¹	0	65	95	30.34	-6.26	102.14	65.54
	0.0006	63	72	21.79	14.83	69.79	62.83
	0.0007	54	68	24.69	15.73	66.69	57.73
	0.0008	52	66	25.24	16.68	66.24	57.68
	0.0009	45	59	26.87	19.57	64.87	57.57
	0.001	41	57	27.09	19.55	62.99	55.45
	0.0015	37	53	27.39	20.64	61.69	54.94
0.002	28	49	30.02	22.30	64.02	56.30	
[Brij-35] μmol L ⁻¹	0	65	95	30.34	-6.26	102.14	65.54
	20	63.5	75	26.04	15.10	84.40	73.46
	30	58.5	67	26.41	19.75	76.95	70.29
	40	55.5	65	27.80	20.75	76.89	69.84
	60	53	63	28.84	21.76	76.76	69.67
	80	50	61	29.99	22.62	76.84	69.27
	90	47	59	30.97	23.39	76.38	68.80
	100	45	57	31.47	24.23	75.96	68.74
	110	43.5	55	31.58	24.97	75.12	68.51
	[Gorkha stick more] ml/L	0	65	95	30.35	-6.26	102.14
0.5		35	41	27.03	24.90	60.032	57.91
1		28	39	27.37	24.09	58.37	55.09
1.5		19	34	27.42	24.04	56.42	53.04
2.0		14	31	27.17	24.00	55.17	52.00
[Kepper] ml/L	0	65	95	30.34	-6.26	102.14	65.54
	0.5	64	83	28.58	7.95	93.78	73.15
	1	51	71	30.46	15.76	78.86	64.16
	1.5	44	66	32.59	18.42	77.89	63.73
	2	35	62	32.85	18.83	72.95	58.93

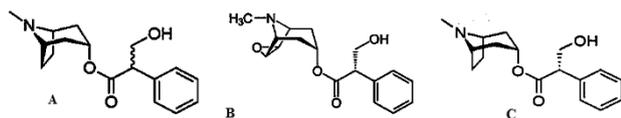


Fig.5. Structure of A) Atropine B) Scopolamine and C) Hyoscyamine [49].

These alkaloids have the sites for the hydrogen-bond formation with surfactants like AOT and Brij-35. The AOT solution has a higher value of adhesion tension due to its acidic nature which ruptures the cuticular wax composition as well as can form the hydrogen bond with the alkaloids present in the leaf surfaces. The Brij-35 solution has a higher value of adhesion tension than the CTAB solution which might be due to the higher number of hydrogen-bond formation in

comparison to CTAB. Thus, the molecular structure and functional group of the surfactants have a significant role in reducing or enhancing the CA that ultimately affects the wettability of leaf surfaces [50].

Surface free energy and Interphase free energy change

Owens, Wendt, Rabel and Kaelble developed a concept to calculate surface free energy (SFE) of the solid surface, popularly known as the OWRK method [51]. This method considered the polar and dispersive parts for the measurement of SFE. Both surface tension of the liquid and surface free energy of the surface are composed of polar and dispersive components (Zhang et al., 2017). Thus, it requires at least two liquids having different polar and dispersive components. However, three liquids ranging from polar to non-polar are best for the estimations of SFE [53]. The surface with higher SFE values is highly wettable [54,55]. In our study, water, ethylene glycol, and di-iodomethane are taken for the measurement of SFE of leaf surfaces. Different variables like surface tension (γ_{LV}), polar component (γ_{LV}^p), and dispersive component (γ_{LV}^d) of test liquids are used to estimate the polar component (γ_{SV}^p) and dispersive component (γ_{SV}^d) of leaf surfaces. The summation of these two components gives SFE of the leaf surfaces. The OWRK method utilizes Eq.3 which is in the form of $y = mx + c$ whose slope and intercept are utilized to get the SFE of leaf surfaces. The equation is automatically solved by the ADVANCE software version 1.9.0.8 to get dispersive and polar components.

$$\frac{(1+\cos\theta)\cdot\gamma_{LV}}{2\sqrt{\gamma_{LV}^d}} = \sqrt{\gamma_{SV}^p} \cdot \sqrt{\frac{\gamma_{LV}^p}{\gamma_{LV}^d}} + \sqrt{\gamma_{SV}^d} \quad (3)$$

Further, interphase surface free energy (γ_{sl}) is calculated by Eq. 4 where Owens and Wendt explained the geometric mean of polar and dispersive components of both liquids and

surfaces as [51]:

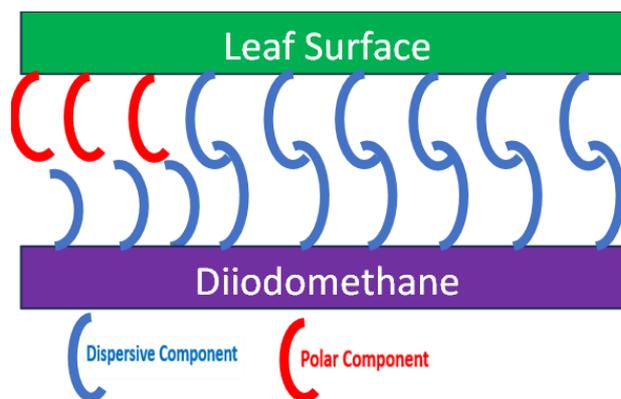
$$\gamma_{sl} = \gamma_{sv} + \gamma_{lv} - 2 \left(\sqrt{\gamma_{sv}^d \gamma_{lv}^d} + \sqrt{\gamma_{sv}^p \gamma_{lv}^p} \right) \quad (4)$$

The lower value of γ_{sl} accounts for better wettability and the higher value accounts for the hydrophobic character of the surfaces. The attachment between two hydrophilic and hydrophobic molecules is due to the interactional behavior of dispersive components among them [51,56].

Table 8. CA [$^\circ$] of water, ethylene glycol, and diiodomethane for three different *D. stramonium* leaves at adaxial and abaxial surfaces and their surface tension value with polar and dispersive parts.

Adaxial Leaf Surface						
Liquid	Mean [$^\circ$] ₁	Mean [$^\circ$] ₂	Mean [$^\circ$] ₃	SFT [mN/m]	Polar [mN/m]	Disperse [mN/m]
Water	50.23 (± 1.76)	59.87 (± 2.74)	59.14 (± 0.99)	72.8	51	21.8
Ethylene glycol	41.40 (± 0.45)	47.42 (± 1.47)	36.99 (± 1.54)	47.7	21.3	26.4
Di iodomethane	38.99 (± 2.95)	40.38 (± 1.02)	31.67 (± 1.21)	50.8	0	50.8
Abaxial Leaf Surface						
Water	83.26 (± 0.41)	83.21 (± 2.72)	83.26 (± 0.41)	72.8	51	21.8
Ethylene glycol	53.89 (± 0.93)	57.15 (± 7.02)	55.20 (± 2.73)	47.7	21.3	26.4
Diiodomethane	52.20 (± 2.30)	49.52 (± 1.70)	51.20 (± 2.30)	50.8	0	50.8

Table 8 suggests that nearly 55.35% part is dispersive in ethylene glycol while diiodomethane is dispersive. In this study, *D. stramonium* leaf surfaces showed nearly 84.5% dispersive part in the adaxial portion and nearly 90.2% dispersive part in the abaxial portion. This dispersive component accounts for lower CA value with ethylene glycol and diiodomethane. Due to the higher percentage of dispersive components in the leaf surface, there exist higher dispersive-dispersive interactions with the dispersive liquid, i.e., diiodomethane which is depicted in **Scheme 6**. The lower percentage of polar component of leaf surfaces showed little interaction with the polar part of the ethylene glycol. There exists adhesion between water and leaf surfaces due to the mutual interactions between the dispersive components of leaf surface and water. The SFE of the adaxial leaf surface is higher than the abaxial one as seen in **Table 9** which accounts for the higher wettability with lower CA value than abaxial surface.



Scheme 6: Schematic Interactions among polar and dispersive components of two substances.

Table 9. Surface free energy (γ_{sv}) of Adaxial and Abaxial surfaces of three different *D. stramonium* leaves with their polar and dispersive parts.

Adaxial Leaf surface				
Results	Value1, mN m ⁻¹	Value2, mN m ⁻¹	Value3, mN m ⁻¹	Average Value, mN m ⁻¹
γ_{sv}	39.64 ± 31.13	43.21 ± 7.34	50.40 ± 11.09	44.42 ± 16.52
Disperse	31.09 ± 19.62	39.08 ± 4.13	42.48 ± 6.43	37.55 ± 10.06
Polar	8.55 ± 11.51	4.13 ± 3.21	7.92 ± 4.66	6.87 ± 6.46
Abaxial Leaf surface				
γ_{sv}	35.97 ± 1.82	37.69 ± 1.29	36.79 ± 1.62	36.82 ± 1.57
Disperse	32.08 ± 1.41	34.50 ± 0.69	33.04 ± 1.30	33.21 ± 1.13
Polar	3.90 ± 0.41	3.19 ± 0.59	3.75 ± 0.32	3.61 ± 0.44

Table 10. Interphase free energy change (γ_{sl}) between leaf surfaces of *D. stramonium* and test liquids; water, ethylene glycol, and diiodomethane, γ_{sv} (adaxial) = 44.42 mN/m and γ_{sv} (abaxial) = 36.82 mN/m.

Surface Parameters	Water	Ethylene glycol	Diiodomethane
γ_{lv} , mN/m	72.8	47.7	50.8
γ_{sl} (Adaxial), mN/m	22.56	4.96	7.87
γ_{sl} (Abaxial), mN/m	28.67	7.76	5.47

There exists the lowest γ_{sl} value with ethylene glycol in the adaxial leaf surface and the lowest γ_{sl} value with diiodomethane in the abaxial leaf surface as shown in **Table 10**. This is because the abaxial portion contains a higher percentage of dispersive components that interact better with the diiodomethane which is dispersive.

Solubility parameter (δ)

The hydrophobic character of the surface is characterized by the solubility parameter (δ) of that surface which depends upon cohesive energy density (e_c) as mentioned in Eq. 5 [57,58]

$$\delta = \sqrt{e_c} \quad (5)$$

where e_c is related to SFE (γ_{sv}) as $e_c = (\gamma_{sv}/0.75)^{3/2}$

The higher the solubility parameter

difference ($\Delta\delta$) between two components, the lower will be the affinity between them [57]. The $\Delta\delta$ value was found to be higher between the abaxial leaf surface and water than that between the adaxial leaf surface and water (**Table 11**) which accounts for a lower affinity of water towards the abaxial leaf surface of *D. stramonium*. The lower affinity prevents the adhesion of water on the leaf surface, thereby reducing the spreading behavior, which ultimately enhances the CA value and reduces the wettability of the surfaces.

Table 11. Cohesive energy density (e_c) and Solubility parameter (δ) of adaxial and abaxial leaf surfaces of *D. stramonium*, $\delta_{\text{water}} = 47.9 \text{ MPa}^{1/2}$ [59].

Adaxial Leaf surface			
γ_{sv} , mJ/m ²	e_c , 10 ⁶ J/m ³	δ_{adaxial} , MPa ^{1/2}	$\Delta\delta = \delta_{\text{water}} - \delta_{\text{adaxial}}$
44.42	455.80	21.35	17.58 MPa ^{1/2}
Abaxial leaf surface			
γ_{sv} , mJ/m ²	e_c , 10 ⁶ J/m ³	δ_{abaxial} , MPa ^{1/2}	$\delta_{\text{water}} - \delta_{\text{abaxial}}$
36.82	343.98	18.55	29.35 MPa ^{1/2}

Conclusions

The AOT solution reduced the CA value in the super-hydrophilic range (< 40°) below the CMC value. The CTAB solution reduced the CA in the super-hydrophilic range in the adaxial surface but could not reduce below 44° in the abaxial surface even at the concentration of 0.002 mol L⁻¹ which is above CMC. The contact angle was above 43° in the case of Brij-35 solution in both adaxial and abaxial surfaces at all concentrations below and above CMC. Gorkha stick more solution reduced the CA value in the super-hydrophilic range even at 0.5 ml L⁻¹ at both adaxial and abaxial leaf surfaces while Keeper solution was found to be effective in reducing CA in the super-hydrophilic region in the adaxial part at the concentration of 2.0 ml L⁻¹. The ΔG and Δg values of AOT are highest among all the adjuvants which is followed by Gorkha stick more, CTAB, Keeper, and Brij-35 solutions. The order of increasing diameter at minimum contact angle of adjuvants follows the

order as Brij-35 < Keeper < CTAB < Gorkha stick more < AOT solutions. The negative value of Δg of the abaxial surface is less than that of the adaxial one. The adhesion tension values of all adjuvants were higher on the adaxial surface than on the abaxial part. The leaf surfaces showed nearly 84.5% dispersive part in the adaxial portion and nearly 90.2% dispersive part in the abaxial portion. The $\Delta\delta$ value was found to be higher between the abaxial leaf surface and water than that between the adaxial leaf surface and water suggesting the abaxial surface is less wettable than the adaxial surface

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Author's Contribution Statement

Pawan Shah: Conceptualization, Sample and Surfactants Selection, Data measurement, Software and graph plotting, Writing: original manuscript, **Diksha Wagle and Roshna Pokhrel:** Measurement of Data **Ajaya Bhattarai:** Conceptualization, Writing: review and editing

Conflict of Interest

The authors do not have any conflict of interest throughout this research work.

Data Availability Statement

The data supporting this study's findings are available from the corresponding authors upon reasonable request.

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