

Analogous Charging Effect of Surfactant-Pesticide Spray Jet on Droplet Characteristics and Deposition on Hydrophobic Leaf Surfaces

Samuel Appah^{1,*}, Eric Amoah Asante², and Christopher Amigangre Ayambire¹

ABSTRACT

An induction charging principle had been applied to enhance spray droplet characteristics and quantity deposition on hydrophobic abaxial-adaxial leaf surfaces from pesticide EC Glyphosate [$C_3H_8NO_5P$] and surfactant Silwet L-77 [$C_{13}H_{34}O_4Si_3$] formulations. A nozzle cap containing two electrodes (spacing at 9 mm apart) was used to superpose charges to spray droplets under applied voltages of 2–12 kV in an electric field (E) of 8.9×10^5 V/m. From a tee-jet flat fan (TP11004VS) nozzle tip fitted into the electrode cap and positioned at 60 cm high above targeted *Brassica campestris* leave surfaces, the spray droplets were directed onto the leaves at a liquid flow pressure of 4 bar and travelling speed of 2 m/s. The measurements were done using Keithsley picoammeter to quantify spray chargeability (CMR), droplet sizes by lesser particle size analyzer (LPSA) and deposition on leaf surfaces by high-speed camera. In effect, droplet sizes of EC, L-77 and EC + L-77 decreased with an increasing applied voltage. The CMR of L-77 was lower than EC and highest for EC + L-77 composite solution. Based on Image analysis of droplets density per leaf area, maximum exposure of adaxial leaf surfaces intercepted many charged spray droplets than abaxial surfaces. As regressed, the quantity of charged spray deposition from EC + L-77 formulation was highest at both adaxial (approx. $27.44 Q_d/cm^2$) and abaxial (approx. $5.57 Q_d/cm^2$) hydrophobic leaf surfaces than EC of 26.12: $3.19 Q_d/cm^2$ and L-77 of 24.80: $2.53 Q_d/cm^2$, respectively. Also, contact angle, Theta E, of charged spray droplets was smaller than Theta C on adaxial than abaxial leave surfaces, a phenomenon attributed to high chemo-electrical properties of formulations that aided the direct flight, coiling and deposition of droplets in order of EC > EC + L-77 > L-77. Generally, there was no observable droplet rebound; hence, surfactant-pesticide composite spraying is considered suitable for electrostatic application in plant protection technology. Therefore, for effective application, charged droplets from glyphosate EC in silwet L-77 solution should be recommended, as it provides optimum droplet sizes, chargeability, contact angle and deposition on hydrophobic leave surfaces.

Keywords: Adaxial-abaxial deposition, charged droplets, droplets density, hydrophobic surfaces.

1. INTRODUCTION

Pesticides are applied to prevent pest infestation, enhance product quality and to control weeds in crop production [1]–[3]. They are formulated at different viscosities and concentrations suitable for effective application. The active ingredients in the chemical are deposited onto the target plants in the form of spray droplets for an interactive effect. The droplets vary in sizes depending on the nozzle tip configuration [4], formulation and application pressure [5]. For a uniform spray, droplet sizes should be in an

atomized form to enable the chemical to penetrate through the plant architecture and spread on the leaf surfaces [6]. The effectiveness of pesticide spraying requires a technology that improves droplets penetration, deposition and retention at reduced bouncing or drifting [7]. Such an objective can be achieved by increasing formulation adhesiveness and spreading properties with surfactant [8], [9] and charging the spray droplets electrostatically [10], [11]. The surfactant helps to reduce the volumetric quantity of toxic and expensive pesticide usage at an improved efficacy

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¹Department of Agricultural Mechanization and Irrigation Technology, University for Development Studies, Ghana.

²Department of Agricultural and Biosystems Engineering, Kwame Nkrumah University of Science and Technology, Ghana.

*Corresponding Author:
e-mail: askappah@yahoo.com

and biological activity [12]. It has a characteristic feature for producing smaller droplet sizes [13] and increasing droplet velocity [14], but these droplet sizes increase when oil-based surfactants are added to a formulation.

The superposition of charges to pesticide spray enhances droplets deposition on leaf surfaces and is significantly controlled by spray liquid properties [10], [15]. Also, droplet charge is enhanced by the addition of surfactants to water solutions [16], hence the need to apply the technology in surfactant-pesticide composite applications. Spray droplet charge has been reported to increase at high applied voltages and lower liquid flow rates during electrostatic induction spraying [17]. From the literature, there is no known electrostatic spray droplets characteristic measurement and impaction behaviour from surfactant cum pesticide spray in plant protection technology. In this paper, we present induction charging effect of surfactant-pesticide composite spray on spray droplets chargeability, sizes and deposition on hydrophobic adaxial-abaxial leaf surfaces. The postulations from the research will provide the significance of charging surfactant in pesticide spray droplets to minimize pesticide wastage, off target deposition and environmental pollution in plant protection.

2. MATERIALS AND METHODS

An electrostatic spraying of surfactant-pesticide composite formulation by induction charging principle is demonstrated to evaluate the effect of applied voltages on droplet characteristics and deposition on hydrophobic leaf surfaces. A factorial design using Minitab Inc software was adopted to generate three factors of pesticide $C_3H_8NO_5P$ glyphosate EC, surfactant $C_{13}H_{34}O_4Si$ silwet L-77 and a composite of EC + L-77 formulations at two replications for the experiment. In every 1 L of tap water, an optimum EC dosage of 12.5 mL and Silwet L-77 of 10 mL were composited for spraying purposes. The solution was continuously stirred with spatula to obtain a uniform emulsion in each case. Accordingly, measurements of solution properties which were significant indicators to spray characteristics and deposition were performed (Table I).

The three formulations were superposed with six applied voltages (V) of 2–12 kV from a 15 kV high voltage capacity generator power supply source. Besides, an induction nozzle cap embedded with two opposite electrodes at distance, $d = 9$ mm apart (Fig. 1) was used to superpose those charges to spray droplets. The positive terminal of the voltage generator was grounded to earth potential to overcome gravitational force $F = m \cdot g$, while the negative terminal was coupled with electrode in the nozzle cap to superpose negative charge to the spray droplets at an electric field, $E = V/d$ of $2.0 \times 10^5 - 13.0 \times 10^5$ V/m suitable for flat fan nozzles configuration.

TABLE I: CHEMO-ELECTRICAL PROPERTIES OF SURFACTANT-PESTICIDE FORMULATIONS

Formulation	pH	°C	(mS/m)	(mN/m)
L-77	6.1	23.5	1.2	36.2
EC	5.3	24.5	4.5	45.1
L-77 + EC	5.7	23.8	6.8	32.4

Spraying was permitted at a constant liquid flow pressure of 4 bar, travel speed of 2 m/s and a 110° tee-jet flat fan (TP11004VS) nozzle tip of 250 mL/min rate, positioned on a custom-built moving equipment at spraying height of 60 cm above target substrates (Fig. 1). The system parameters were chosen based on an initial trials of optimum parameters combination assessment with water solvent, as liquid properties affected spray characteristics. Since droplet charge decreases with spraying height, using 60 cm is ideal to enhance droplets deposition and surface coverage [18], [19]. The experiment was carried out in the Key Laboratory of Plant Protection Engineering, Jiangsu University, China under controlled temperature (25 °C) and humidity (65%). Due to the particulate fine droplet sizes, the room internal wind turbulence was set to still by shutting all openings throughout the experimental process following observation and determination of response variables.

The effectiveness of induction spraying was weighted based on quantity of entrained charges possessed by spray jet. At any given time, t (s), the mass of spray discharge, Q (kg) from nozzle spout containing electric current, I (C/s) to act on leaf surfaces determined its spray chargeability, where the ratio of spray current (I) to rate of discharge (Q/t) is given as charge-mass ratio

$$CMR = I \times [Q/\Delta t]^{-1} \times 10^3 \text{ (mC/kg)}.$$

The rate of spray discharge Q was obtained by collecting spray droplets with a graduated beaker of known weight for 60 s and weighing the amount of solution harvested [11], [20], [21]. Whereas a 60 cm diameter circular (2.829 m²) wire mesh was connected to Keithley picoammeter (Model No. 6485, Keithley A Tektronix Inc. Company, Ohio) by a conducting wire and placed under the spray jet to detect and record spray current from the high voltage generator.

A spray plume from each formulation was directed downward across diffracted rays of Laser Particle Size Analyzer (Winner 318, Shandong, China). At every applied voltage and formulation, particulate droplet sizes were captured, and data was stored in a computer [22]. The classifications of spray droplets were evaluated on the basis of relative span in the range of volume median diameter VDM, $D_{v,1}$, $D_{v,5}$ and $D_{v,9}$ (i.e., estimation of 10%, 50% and 90% proportions of droplets in spray plume). A relative span (RS) = $[D_{v,9} - D_{v,1}]/D_{v,5}$ quantifies the variations of spray droplets in a jet, hence the atomizer and uniformness of droplets from nozzles, the better the relative span [6], [19].

Furthermore, charged spray droplets from each formulation were directed onto leaf surfaces (Fig. 2). A *Brassica campestris* plant with pinnate leaf arrangements at horizontal orientations were erect perpendicular to the nozzle orifice to intercept spray droplets for deposition assessment on both adaxial and abaxial surfaces. The seedlings were pricked out from a commercial vegetable field and potted for 14 days until 5–6 leaves fully emerged on the plant. These live plants were used in such a way to simulate in-situ droplets behaviour on leaf surfaces at field condition. Spraying was done at a single pass across the

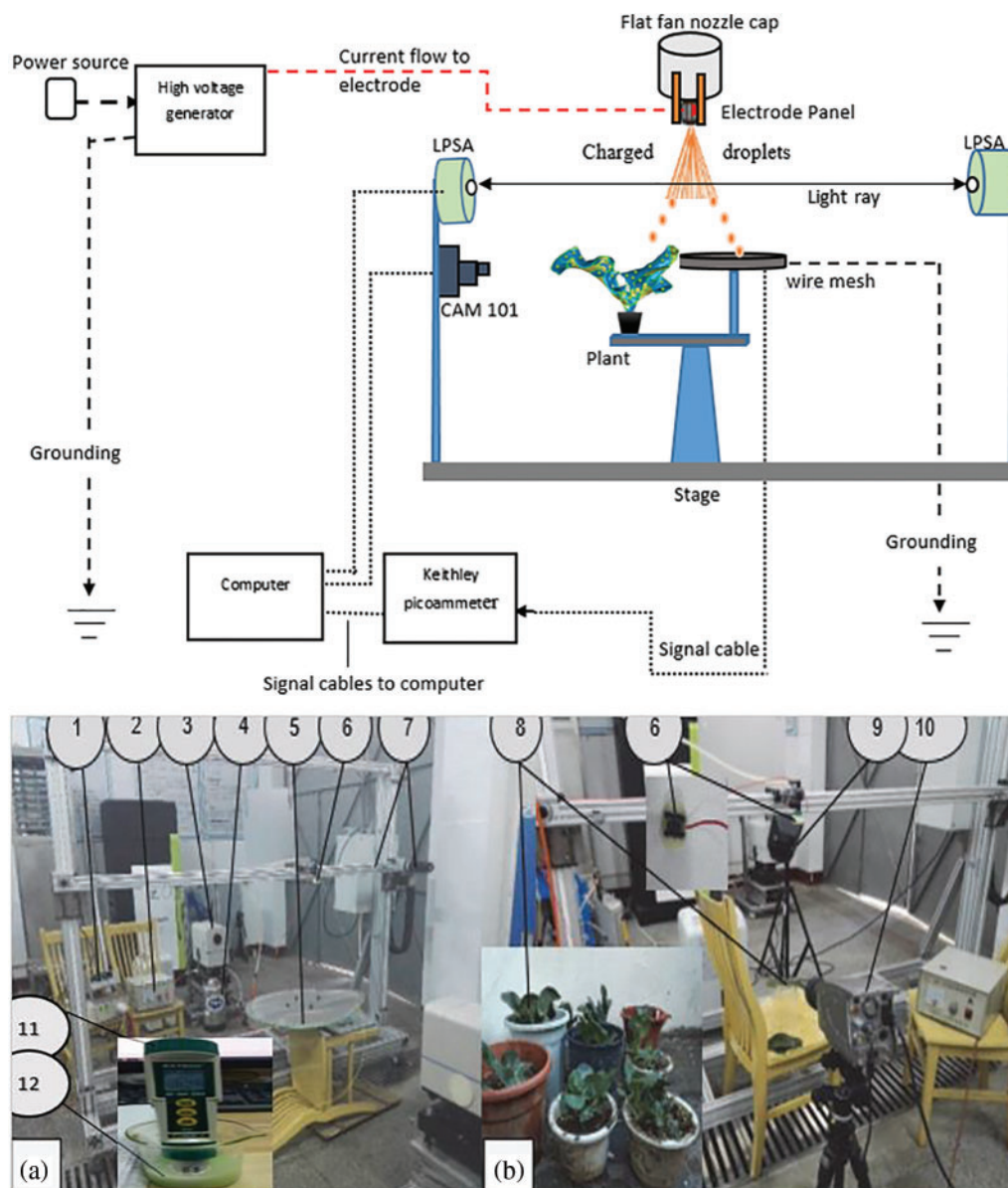


Fig. 1. Experimental scheme and set-up of induction charging of EC, L-77 and EC + L-77 formulation for measurement of (a) CMR and droplet sizes, (b) quantity of droplet deposition on hydrophobic leaf surfaces. 1. Keithley picoammeter, 2. High voltage generator, 3. LPSA, 4. Pesticide formulation tank 5. Circular wire mesh, 6. Induction nozzle cap, 7. Telescopic boom for spraying height and speed adjustment, 8. *Brassica campestris* plant and leaf mount, 9. LED backlight, 10. High speed camera, 11. EC and L-77 formulation, 12. Conductivity meter.

potted plants. From the erect plants, the two most exposed leaves inclined horizontally to the earth plane and perpendicular to the spray plume were carefully picked and photographed adaxially and abaxially after spraying. A high-speed camera (Olympus i-speed 3, Tokyo, Japan) was used to capture the images of droplets. The quantity of charged spray droplets that impinged on both surfaces was estimated from a measured leaf area of 25 cm². The images were then subjected to imageJ software analysis (1.38e/Java 1.5.0_09) as in Martin [23] and Lin *et al.* [24].

Using the ImageJ algorithm, imagery was bandpass filtered, adjusted and threshold to count droplet density (Fig. 6b), while plugin contact angle tool was systematically followed to estimate droplet contact angles as either Elliptical (Theta E) or Circular (Theta C) on the hydrophobic leaf surfaces (Fig. 6a). In both measurements, the images were threshold from RGB to grey at 8 bit to

segregate droplet sizes before quantification of droplets and determination of contact angle.

The quantitative data from droplet sizes, chargeability and density were subjected to statistical analysis using Minitab Inc. 2007 software at a 95% confident interval (CI). The quantity of spray droplets deposited (Q_d) per given area (cm²) on both adaxial and abaxial surfaces of hydrophobic leaf surfaces were extracted from high-speed camera imagery using ImageJ public software tool pack.

3. RESULTS AND DISCUSSION

The nature of spray jet is a characteristic function of surface tension, electrical conductivity, pH and temperature of the formulations. These chemo-electrical properties resulted in variations in droplet sizes, chargeability and droplet density on leaf surfaces. The limit at which breakup of charged droplet occurred is regulated by Rayleigh (Ra)

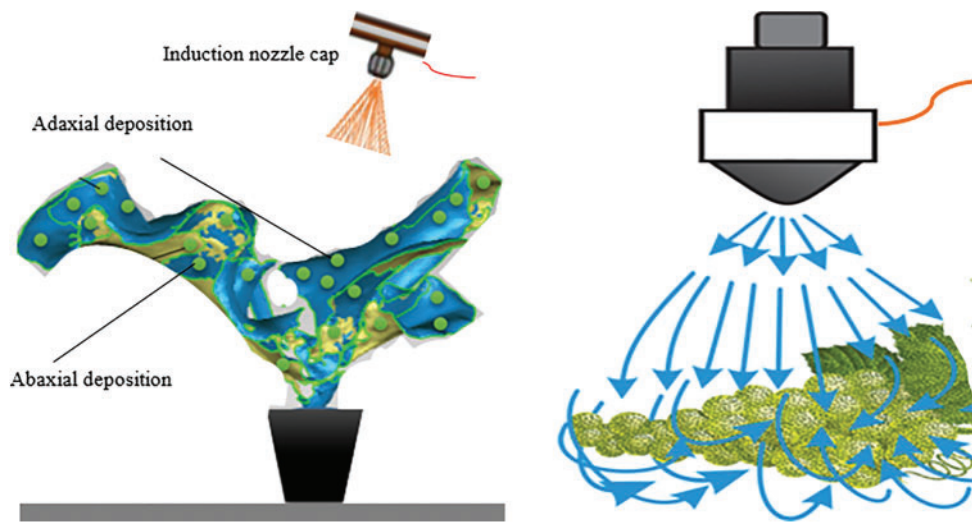
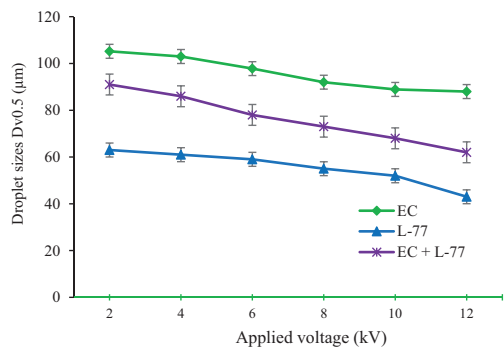


Fig. 2. A model of spray droplets deposition on both adaxial and abaxial leaf surfaces of *Brassica campestris* potted plant.

value of $2\pi\sqrt{2(\epsilon_0\gamma d^3)}$ for $d = (6Vi/\pi)^{1/3}$ at vacuum permittivity constant (ϵ_0), droplet volume (Vi), surface tension (γ) and droplet diameter (d) parameters [25]. Droplet sizes at any level of charged pesticide-surfactant formulations were comparatively small compared to Ra due to variations in electrochemical properties and the breakup of the jetting period. The atomised charged droplet sizes contributed to the maximum attraction and deposition on leaf surfaces in consonance with columbic forces $F_1 = q \times E$ applicable for practical application in plant protection [26]. In all, different applied voltages did not change an average mass flow rate of $EC = 1.62 \times 10^{-2}$ kg/s, $L-77 = 1.60 \times 10^{-2}$ kg/s and $EC + L-77 = 1.66 \times 10^{-2}$ kg/s at all levels.

3.1. Effect of Formulation and Applied Voltage on Charged Spray Droplet Sizes

Fig. 3 presents the nature of droplet sizes from different voltages application on surfactant in pesticide spray jet. There was a general trend of droplet sizes declination from EC, L-77 and EC + L-77 solutions with respect to increase in applied voltages. At higher voltages, there was droplet shrinkage with fast breakup points leading to smaller sizes. The droplet sizes of EC + L-77 solution sandwiched EC



Source of Variation	SS	df	MS	F	P-value	F crit
Formulation	4878	2	2439	32.32	3.65E-06	3.682
Error	1132	15	75.46			
Total	6010	17				

Fig. 3. Charged spray droplets $D_{v0.5}$ diameters from surfactant-pesticide composite spray at different applied voltages.

TABLE II: RELATIVE SPAN OF CHARGED DROPLET SIZES						
Formulation	Applied voltage (kV)					
	2	4	6	8	10	12
EC	0.9438 ^a	0.8263 ^a	0.8728 ^a	0.8752 ^a	0.9250 ^a	0.9434 ^a
EC + L-77	0.8777 ^b	0.8850 ^b	0.8492 ^b	0.8623 ^b	0.9183 ^b	0.8777 ^b
L-77	0.8429 ^c	0.8203 ^c	0.7800 ^c	0.8218 ^c	0.8112 ^c	0.8429 ^c

Note: abc means that present same letters do have no significant difference.

and L-77 solutions with EC values being biggest whereas L-77 recorded the smallest at all levels of applied voltages [13]. Droplet sizes of EC, L-77 and EC + L-77 at 2 kV were 105.2, 63.3 and 91.2 μm which fell at 12 kV to 88.4, 43.1 and 62.3 μm , respectively. An interactive effect of charges on droplet sizes was weighted according to variations in volume median diameter of $D_{v0.9}$, $D_{v0.5}$ and $D_{v0.1}$ droplet segregation in the spray plume (Table II). The level of variability, also known as relative span (RS), was found to be near 1.0 at $p < 0.05$, indicating uniformity of droplet sizes at all formulation treatments. As droplet sizes contribute significantly to the efficiency of pesticide spraying, obtaining optimum sizes that overcome drift at flying time to impact on leaf surfaces is an object of spraying. Yet, with induction charging application, the possessed entrained charges direct spray droplets to target sites no matter the droplet sizes, thereby enhancing surfactant-pesticide spraying efficiency. Applied voltages did not have any significant effect on droplet sizes as compared to formulation treatments.

3.2. Effect of Applied Voltage and Formulation on Spray Chargeability

Spray chargeability (with negative sign) from surfactant-pesticide formulations at different applied voltages is given in Fig. 4. The entrained chargeability of spray was higher upon compositing EC + L-77 during spraying followed by sole L-77 and EC solutions, but with no significant effect within formulations except that of applied voltages. An applied voltage of 2 kV injected -1.01×10^{-8} mC/kg to L-77, -1.13×10^{-8} mC/kg to EC and -1.20×10^{-8}

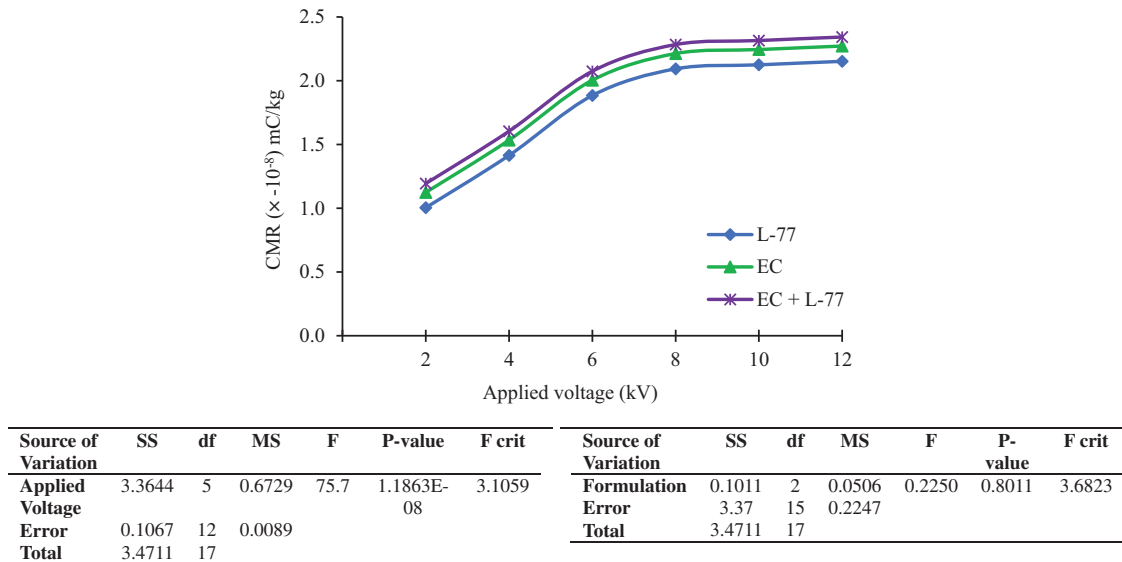


Fig. 4. Effect of pesticide-surfactant formulations and applied voltages on charge-mass ratio (CMR).

mC/kg to EC + L-77 spray droplets, whereas 12 kV superposed -2.15×10^{-8} mC/kg to L-77, -2.27×10^{-8} mC/kg to EC and -2.34×10^{-8} mC/kg to EC + L-77 sprays. The near stability of spray droplets charge occurred at high applied voltages between 8–12 kV which could be attributed to reduction in droplet sizes. This variation in spray chargeability could be as a result of free charge and electric field interference from the surroundings [11]. Though increasing applied voltages resulted in shrinkage of droplet sizes, the entrained charges were maximum at higher voltages as compared to lower voltages that produced larger droplets [11].

3.3. Effect of Applied Voltage on Droplet Deposition and Contact Angle at Adaxial and Abaxial Leaf Surfaces

An in-situ droplet density and contact angle of surfactant-pesticide spray on hydrophobic leaf surfaces revealed the effectiveness of spray chargeability. There were no observable spray droplets rebound on the surfaces of *Brassica campestris* leaves (Fig. 6a). Overall, increasing applied voltages on spray droplets from EC, L-77, and L-77 + EC formulations enhanced droplet density on both adaxial and abaxial leaf surfaces but with no significant effect on interactions (Figs. 5 and 6b). After a single pass application, the adaxial spray deposition per leaf surface area exceeded that of abaxial because of its wider exposure to the spray plume. At minimum applied voltage of 2 kV, the number of adaxial droplet deposition was low, but increasing the applied voltages through to 12 kV significantly increased the number of droplet density deposition across all formulation treatments (Fig. 5a). The least number of spray droplets per given area on adaxial leaf surfaces was observed from L-77, while L-77 + EC yielded the highest spray droplet density deposition with an accompanying coefficient of droplet variability of 91.3%. It is shown that, the combined effect of EC + L-77 solutions was strong enough to direct many charged droplets onto the leaf surfaces. Similarly, on adaxial surfaces, the number (Q_d) of charged spray droplets per given area (cm^2) observed were EC (26.21–29.71 Q_d/cm^2), L-77 (25.02–28.95 Q_d/cm^2) and

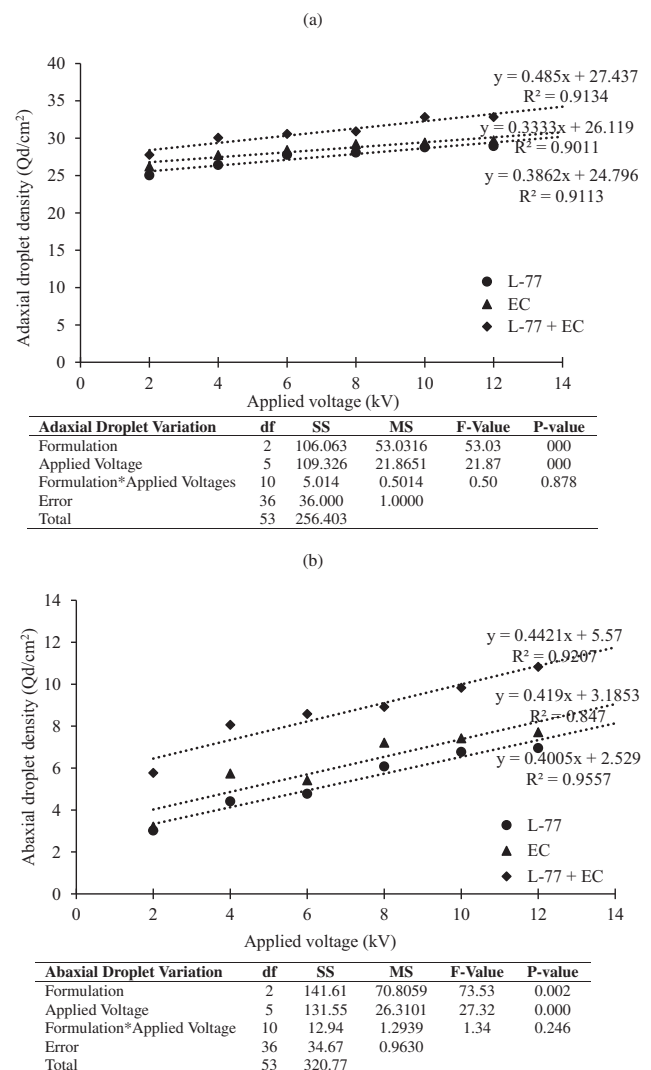


Fig. 5. Droplet density deposition of charged droplets of pesticide-surfactant solutions on: (a) adaxial and (b) abaxial leaf surfaces.

L-77 + EC (27.77–32.83 Q_d/cm^2) at applied voltages of 2–12 kV.

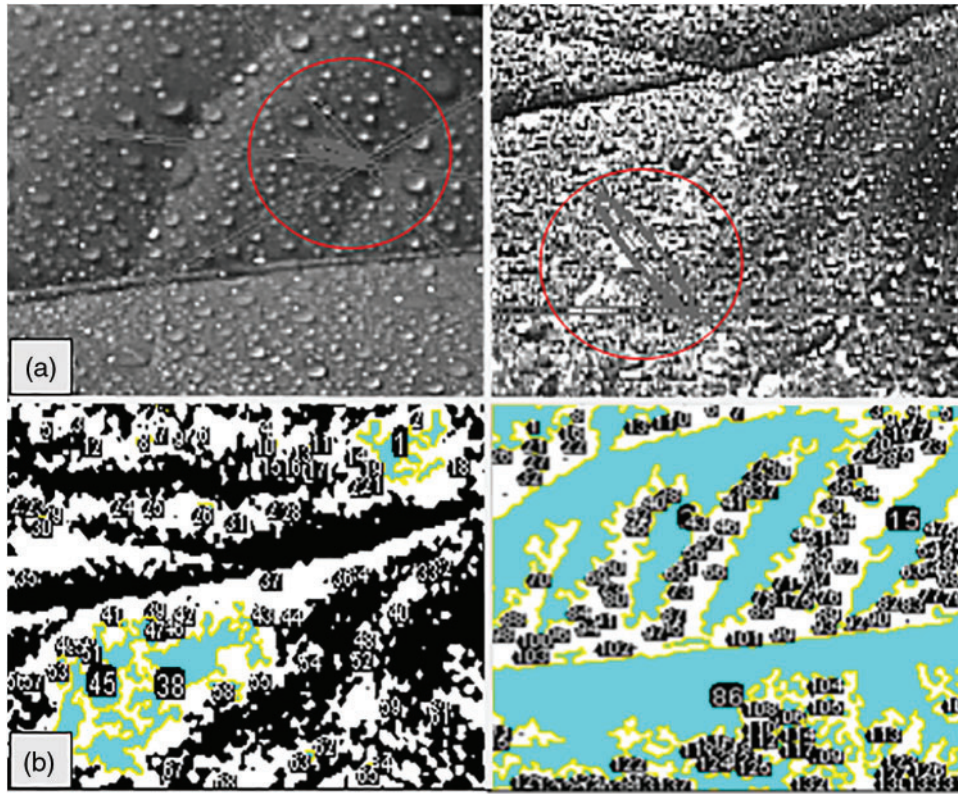


Fig. 6. Sample schematics of ImageJ characterization of (a) spray droplets contact angle (θ) at adaxial-abaxial and (b) quantity of spray droplets deposited per given area (Q_d/cm^2) on adaxial-abaxial at 8 kV applied voltage.

Besides, few droplet density depositions were recorded on abaxial surfaces of *Brassica campestris* leaves due to the awkward and hidden surface exposures to spray jet at all levels of formulation treatments (Fig. 5b). It is by attraction of polarities between charged droplets and leaf ions that enabled droplets to coil and pinned on underside leaf surfaces. Again, least spray droplet density deposition was recorded for L-77 formulation with R^2 of 90%, while maximum coefficient of determination ($R^2 = 92\%$) was produced by L-77 + EC composite spray. However, charged EC produced the lowest droplet variability of approx. 85% in a spray jet. Abaxial droplet density deposition ranged from 3.02–6.95 Q_d/cm^2 for the case of L-77 solution, 3.21–7.71 Q_d/cm^2 with respect to EC solution and 5.77–10.83 Q_d/cm^2 for L-77 + EC solution.

$$\begin{aligned} \text{Adaxial: } & \begin{bmatrix} \text{L-77 + EC} \\ \text{EC} \\ \text{L-77} \end{bmatrix} \\ & \approx \begin{bmatrix} y = 0.4850x + 27.437 \rightarrow R^2 = 91.34\% \\ y = 0.333x + 26.119 \rightarrow R^2 = 90.11\% \\ y = 0.386x + 24.796 \rightarrow R^2 = 91.13\% \end{bmatrix} \end{aligned} \quad (1)$$

$$\begin{aligned} \text{Abaxial: } & \begin{bmatrix} \text{L-77 + EC} \\ \text{EC} \\ \text{L-77} \end{bmatrix} \\ & \approx \begin{bmatrix} y = 0.4421x + 5.57 \rightarrow R^2 = 92.07\% \\ y = 0.419x + 3.1853 \rightarrow R^2 = 84.70\% \\ y = 0.4005x + 2.529 \rightarrow R^2 = 95.57\% \end{bmatrix} \end{aligned} \quad (2)$$

In effect, during spraying, most droplets were deposited on adaxial surfaces than abaxial. Yet, the combined effect of charges on L-77 + EC spray droplets was capable to enhance wraparound deposition for interactive effect. Irrespective of the leaf orientation, there were significant differences among quantity of droplets from the formulations at each applied voltage. An increase in deposition at higher applied voltages was attributed to the production of fine particulate droplet sizes containing high entrained electric charge [27]. Also, the entrained charge of droplets by induction charging inhibited spray drift and confined most of the droplets on leaf surfaces irrespective of the droplet sizes [28]. This behaviour of spray droplet deposition indicates the need to formulate surfactant in pesticide solution [29] and superpose charges by induction principle in plant protection technology [25].

The contact angle of droplets on both adaxial and abaxial surfaces revealed a drastic drop of elliptical (Theta E) as compared to circular (Theta C) angles (Table III). In this case, elliptical contact angle is considered since charged droplets are viewed as oval rather than circular shapes. In all formulation treatments, the effect of high 6.8 mS/m electrical and low 32.4 mN/m surface tension properties (Table I) gave L-77 + EC an intermediate contact angles

TABLE III: CONTACT ANGLE OF CHARGED DROPLET SIZES				
Formulation	Adaxial droplets		Abaxial droplets	
	Theta E	Theta C	Theta E	Theta C
L-77	21.3 ± 15.06	58.3 ± 10.20	33.8 ± 5.66	45 ± 13.15
EC	42.7 ± 8.53	87.7 ± 13.78	65.8 ± 12.11	90.1 ± 9.33
L-77 + EC	34.8 ± 10.08	67.6 ± 6.51	43.4 ± 8.72	56.3 ± 12.14

between sole L-77 and EC solutions (Fig. 6a), suitable for pesticide application in plant protection. The contact angle was smaller on adaxial than abaxial due to high impaction force possessed by droplets in direct flight than those coiling and pinning at underside leaf surfaces. Therefore, for effective application in plant protection, charged droplets from L-77 + EC is ideal to produce a contact angle of approx. 35°–43° Theta E, otherwise EC of 42°–65° and L-77 of 21°–33° could be considered on hydrophobic leaf surfaces.

4. CONCLUSION

Charged droplets characteristics and deposition on hydrophobic adaxial and abaxial leaf surfaces of *Brassica campestris* from pesticide EC, surfactant L-77 and L-77 + EC spray were investigated in-situ at applied voltages of 2–12 kV. The superposition of charges to spray jet decreased droplet sizes of EC, L-77 and EC + L-77 as applied voltages increased, whereas CMR of L-77 was lower than EC and highest for EC + L-77 solution. Also, maximum exposure of adaxial leaf surfaces intercepted more charged spray droplets than abaxial based on ImageJ analysis of droplets density per leaf area. As regressed, charged spray density deposition on adaxial surfaces was highest in the case of EC + L-77 (27.44 Q_d/cm^2) but reduced to EC (26.12 Q_d/cm^2) and L-77 (24.80 Q_d/cm^2) solutions. Similarly, abaxial spray deposition was maximum at EC + L-77 (5.57 Q_d/cm^2) followed by EC (3.19 Q_d/cm^2) and L-77 (2.53 Q_d/cm^2). Also, charged spray droplets produced smaller contact angle on adaxial than abaxial leaf surfaces due to direct flight and high impact force than those droplets coiling and pinning at underside leaf surfaces. There was no noticeable spray droplet rebound, hence, surfactant-pesticide formulation is considered suitable for electrostatic application in plant protection technology.

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CONFLICT OF INTEREST

The authors declare that they do not have any conflict of interest.

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