

UAV spraying on wheat field: how tank-mix adjuvant type and concentration influence the contact angle on wheat leaf surface

Yanhua Meng¹, Qiufang Wu^{1,2}, Hanxue Zhou³, Hongyan Hu^{Corresp. 4}

¹ Anyang Institute of Technology, Anyang, Henan Province, China

² Anyang Wheat Breeding Engineering Research Centre Research Room, Anyang, Henan Province, China

³ Anyang Quanfeng Biotechnology Co., Ltd, Anyang, Henan Province, China

⁴ State Key laboratory of Cotton Biology, Institute of Cotton Research, Chinese Academy of Agricultural Sciences, Anyang, Henan Province, China

Corresponding Author: Hongyan Hu

Email address: huhongyan1986@163.com

Currently, UAV spraying pesticides is a popular issue in Asian countries. How to improve pesticide efficiency of UAV spraying has been concerned by many researchers. The property of spraying solutions is one of the caused concerns due to small droplets and high drift potential of UAV spraying. The wetting property of droplets on crop leaves is a key factor that affects the spraying efficiency of pesticides. Tank-mix adjuvants, which might alter liquid's wetting ability on crop leaves, are coupled with UAVs for foliar application to enhance pesticide efficiency. However, different types and concentrations of adjuvants may have different impacts on the wetting properties of droplets. In this paper, we investigated the effects of four tank-mix adjuvants, BDT, VP, NJF and LY, on the dynamic Contact Angle (CA) values of droplets on the adaxial surface of wheat leaves. We measured the dynamic CA values of various concentrations of each adjuvant solution and determined the optimal concentrations based on the CA values, droplet spreading time and cost. The results indicated that adding any of the four adjuvants significantly reduced the CA values. However, the CA decrease- pattern varied among the four adjuvants. The CAs of BDT and VP solutions showed slight or stable decrease during the observing time (0-8.13 seconds), while these of NJF and LY solutions experienced rapid decrease throughout the observation period. According to the dynamic CA values of different concentrations, the optimal concentrations of BDT, DDE, NJF and LY for field application in wheat were 12%, 16%, 6‰ and 0.3‰, respectively. Alkoxy modified polytrisiloxane adjuvant (LY) could be recommended as an appropriate tank-mix adjuvant for wheat filed application after considering spreading efficiency and cost. This study provides theoretical and practical guidance for selecting and optimizing tank-mix adjuvants for drone spraying.

UAV spraying on wheat field: how tank-mix adjuvant type and concentration influence the contact angle on wheat leaf surface

Yanhua Meng¹, Qiufang Wu^{1,2}, Hanxue Zhou³, Hongyan Hu^{4*}

¹ Anyang Institute of Technology, Anyang, Henan Province, 455000, China

² Anyang Wheat Breeding Engineering Research Centre Research Room, Anyang, Henan Province, 455000, China

³ Anyang Quanfeng Biotechnology Co., Ltd, Anyang, Henan Province, 455000, China

⁴ State Key laboratory of Cotton Biology, Institute of Cotton Research, Chinese Academy of Agricultural Sciences, Anyang, Henan Province, 455000, China

Corresponding Author:

Hongyan Hu*

State Key laboratory of Cotton Biology, Institute of Cotton Research, Chinese Academy of Agricultural Sciences, Anyang, Henan Province, 455000, China

Email address: huhongyan1986@163.com

Abstract:

Currently, UAV spraying pesticides is a popular issue in Asian countries. How to improve pesticide efficiency of UAV spraying has been concerned by many researchers. The property of spraying solutions is one of the caused concerns due to small droplets and high drift potential of UAV spraying. The wetting property of droplets on crop leaves is a key factor that affects the spraying efficiency of pesticides. Tank-mix adjuvants, which might alter liquid's wetting ability on crop leaves, are coupled with UAVs for foliar application to enhance pesticide efficiency. However, different types and concentrations of adjuvants may have different impacts on the wetting properties of droplets. In this paper, we investigated the effects of four tank-mix adjuvants, BDT, VP, NJF and LY, on the dynamic Contact Angle (CA) values of droplets on the adaxial surface of wheat leaves. We measured the dynamic CA values of various concentrations of each adjuvant solution and determined the optimal concentrations based on the CA values, droplet spreading time and cost. The results indicated that adding any of the four adjuvants significantly reduced the CA values. However, the CA decrease- pattern varied among the four adjuvants. The CAs of BDT and VP solutions showed slight or stable decrease during the observing time (0-8.13 seconds), while these of NJF and LY solutions experienced rapid decrease throughout the observation period.

According to the dynamic CA values of different concentrations, the optimal concentrations of BDT, DDE, NJF and LY for field application in wheat were 12%, 16%, 6‰ and 0.3‰, respectively. Alkoxy modified polytrisiloxane adjuvant (LY) could be recommended as an appropriate tank-mix adjuvant for wheat field application after considering spreading efficiency and cost. This study provides theoretical and practical guidance for selecting and optimizing tank-mix adjuvants for drone spraying.

Keywords: contact angle; tank-mix adjuvant; UAV; wheat leaf.

Introduction

Crops are always suffered from the continuous invasion and attacked by pests, diseases and weeds and the application of pesticides are usually adopted to maintain crop output (Matthews & Thomas, 2000; Zhu et al., 2019). Foliage application of pesticides is one of the most efficient approaches to keep arable crops from the harmful damages of pests and diseases (Jensen & Olesen, 2014). During the foliage application, how much a pesticide solution wets a leaf depends on both the properties of the liquid (pesticide solution) and the solid substrate (crop leaf) (Quetzeri-Santiago, et al., 2020). Crop leaves are the main part of plants to receive droplets in the process of foliage application. The ability of crop leaves to retain droplets has been proofed to influence pesticide efficacy significantly (Fountain, Harris & Cross, 2010; Fang et al., 2019). The ability of a crop leaf to retain water on its surface is regarded as leaf wettability (Fernández et al., 2014; Papierowska et al., 2018; Cavallaro et al., 2022). The wettability of a crop leaf could be changed by the physicochemical properties of a liquid (Sanyal, Bhowmik & Reddy, 2006; Nairn, Forster & Van Leeuwen, 2011; Da Silva Santos et al., 2021). Contact angle (CA) is commonly recognized as a key metric to describe the ability of a liquid to wet the crop leaf (Wang et al., 2016; Song et al., 2022). In this study, a CA refers specifically to the angle a liquid form between the interface of leaf surface and liquid and the tangent to the liquid surface (Fig. 1). The larger the wetting tendency is, the smaller the CA is. A liquid that forms a CA smaller than 90° is categorized as a wetting liquid, while a liquid that forms a CA between 90° and 180° is a non-wetting liquid. On the other hand, if a liquid creates a CA between 0° and 90° on a crop leaf, the crop leaf is hydrophilic, otherwise, the crop leaf is hydrophobic (Jeevahan et al., 2018). In the case of a hydrophilic crop leaf, i.e., wettable leaf, the droplet dissipates over its surface quickly and dries faster. However, in the case of a hydrophobic crop leaf, droplet does not spread but retain its shape on the leaf surface, which would result in droplet running off the surface easily. Furthermore, the non-spreading droplets take a longer time to dry, which will create favorable conditions for the growth and spread of plant pathogens (Rowlandson et al., 2018). Therefore, a wetting liquid is required to obtain a satisfactory biological control efficiency during a pesticide spraying application on a hydrophobic crop (Meng et al., 2022).

As mentioned above, the wettability of crop leaf could be changed by the physicochemical properties of spray liquid, which influence the effectiveness of pesticides directly (Zhang et al., 2017; Sobiech et al., 2020). The use of tank-mix adjuvant can alter the physical and chemical

properties of the spray liquid by reducing CA value and surface tension, mitigating the negative effect of PH, enlarging droplet size, and so on, which result in facilitating the spray liquid to spread on crop leaf and improve the efficiency of pesticides(He et al., 2021).

Normally, nozzles of agricultural UAVs are at an altitude of 2 to 3 m above crop canopy, while these of ground-based sprayer is at around 0.5 m above crop canopy. Longer distance between nozzles and target crop canopy and unexpected crosswind could result in higher droplet drift potential (Lou et al., 2018). Furthermore, small droplet size, which are seen in UAV spraying commonly, are another factor to facilitate droplet to be drifted away (Chen et al., 2020). Thus, UAV spraying pesticide are generally combined with the use of tank-mixed adjuvants to improve pesticide efficiency by reducing droplet drift(Wang et al., 2018;Zhang&Xiong, 2021). For a hydrophobic crop, the function of tank-mix adjuvant it is not only to reduce droplet drift, but also to facilitate droplet spread on crop leaf as soon as possible (Peirce et al., 2016).

Wheat is one of the typical hydrophobic crops(Song et al., 2022). CAs on wheat leaves have been measured at 118-152° and 140-146°(Márquez,Stuart-Williams&Farquhar, 2021). Therefore, the wettability of pesticide solution is critical for controlling wheat diseases and pests. Tank-mix adjuvants are employed in pesticide solution when using aerial sprayer to improve pesticide efficiency in several previous literatures. Meng et al.(2018) report that the use of tank-mix adjuvant can reduce imidacloprid dosage by 20% without increase negative effect on wheat aphid control efficacy when using UAV sprayer. Wang et al.(2022) explore that the addition of tank-mix adjuvants to spray solution can improve the control efficacy of wheat aphids and rust significantly and extend the duration of the pesticide. Yan et al. (2021) investigate that the addition of tank-mix adjuvant can improve control effect of prothioconazole on Fusarium head blight in wheat and increase wheat yield. Zhao et al.(2022) report that the use of appropriate tank-mix adjuvants for aerial sprayer on wheat field can significantly improve the performance of pesticide by increasing pesticide dosage delivery efficiency and disease control efficacy. Zhao et al.(2022) also explore that the use of tank-mix adjuvant can also help reduce the pesticide dosage while ensuring their effectiveness, which is similar to the conclusion of Meng et al.(2018) mentioned above. Song et al. (2022) evaluate four types of tank-mix adjuvants on wheat leaf by measuring metrics such as surface tension, CA, and so on and the results indicate that the adjuvant type has great effect on surface tension and CA value.

Although the effect of tank-mix adjuvant on pesticide efficiency of wheat pests and diseases control are explored widely, the measurement of dynamic CA values of different tank-mix adjuvant under a serial of concentration is rarely reported. Normally, only one CA value of a liquid is given but the measuring time of this CA value is unclear. In fact, CA value changes with time after a droplet is deposited on a crop leaf in most cases, especially for a liquid with a tank-mix adjuvant. The main objective of this study was to investigate the influence of tank-mix adjuvant type and concentration on CA values on wheat leaf surface to select appropriate adjuvant type and corresponding concentration for wheat pests and disease control when UAVs are adopted as sprayers.

Materials and methods

Materials

The variety of wheat used in this study is Zhoumai 22, which is planted on campus experimental field of Anyang Institute of Technology. Wheat leaves were collected freshly during the late of flowering period, which is a critical time for wheat pests and diseases control.

Tank-mix adjuvants Beidatong (BDT) (methylated plant oil, Hebei Mingshun Agricultural Co., Ltd, China), Velezia Pro (VP) (mineral oil, TotalEnergies Fluid company, France), Nongjianfei (NJF) (hyperbranched fatty alcohol ether modified polymer, Guilin Jiqi Biochemical Co., Ltd, China), and Lieying (LY) (alkoxy modified polytrisoloxane, Anyang Quanfeng Biotechnology Co., Ltd, China) were used in this study.

CA value measurement

Laboratory experiment was designed to optimize the appropriate concentration of four tank-mix adjuvants (BDT, VP, NJF, and LY) by measuring dynamic CA values on wheat leaf adaxial surface under different concentrations, respectively. The four adjuvants were mixed with tap-water as the tested aqueous solution with different concentrations, respectively. Eight concentrations (2%, 4%, 6%, 8%, 10%, 12%, 14%, 16%) of BDT, eight concentrations (4%, 8%, 12%, 16%, 20%, 24%, 28%, 32%) of VP, fourteen concentrations (0.2‰, 0.4‰, 0.6‰, 0.8‰, 1‰, 2‰, 3‰, 4‰, 5‰, 6‰, 7‰, 8‰, 9‰, 10‰) of NJF, and three concentrations (0.1‰, 0.2‰, 0.3‰) of LY were prepared for dynamic CA value measurements, respectively.

CA value of each concentration was measured on the adaxial surfaces of three freshly undamaged wheat leaves collected from experimental field. Adhesive tape was adopted to fix the tested leaf on the glass slide (25 cm × 76 cm) to facilitate the capture of images for CA measurement. The interval of image capture was 0.07 s, and the dynamic CA value was measured from 0.00 to 8.13 s in most cases. The details of observing time and number of CA values were listed in Table 1. The initial CA ($t=0$ s) was recorded as $CA_{initial}$ and it was compared between solution concentrations of the same tank-mix adjuvant. The final CA (the last measuring time) was recorded as CA_{final} . The change of CA value was used to describe the decrease of CA and it is shown in the following equation.

$$CA_{change} = CA_{initial} - CA_{final} \quad (1)$$

The optical tensiometer Attention Theta Flex (Biolin Scientific) equipped with a high-resolution camera (1984 × 1264 px with a maximum of 3009 FPS) and LED light, is adopted to measure CA by using sessile drop method. The details of measuring process can be found in the previous study ([Meng et al., 2022](#)).

Laboratory measurements were performed at a constant relative humidity of 57% and room temperature of 27 ± 0.4 °C.

Result

Dynamic contact angle on wheat leaves of four aerial adjuvants

As shown in Fig. 3 and Fig. 6, a notable decrease in the CA values is observed after the addition of the four tank-mix adjuvants, respectively. CA values of tap-water on wheat leaf adaxial surface is around 142.89 °, which agrees with the result of the previous study (Márquez, Stuart-Williams&Farquhar,2021). Fig. 2 presents the appearance shape of tap-water droplets on wheat leaf adaxial surface over 8.13 s.

Overall, CA behaviour of BDT and VP are similar (Fig. 3), while these of NJF and LY are alike in most measuring cases in this study (Fig. 6). Fig. 3 and Fig. 6 illustrate the initial CA ($t=0$ s) values ($CA_{initial}$) of each concentration along with CA values of tap-water change during the observing time (0 s - 8.13 s).

In the case of BDT, the highest initial CA value (90.63°) was observed for the concentration of 2% (Fig. 3 a) and the lowest initial CA value (60.04°) was observed for the concentration of 12% (Fig. 3 c). In BDT group of 2%, 4%, 6% and 8%, the initial CA values decrease with the increase of concentration, but the CA values are similar after 0.3 s except for the concentration of 2% (Fig. 3 a). In BDT group of 10%, 12%, 14% and 16%, the highest initial CA value was found in concentration of 16% (83.39°), followed by 10% (81.70°), 14% (69.82°) and 12% (60.04°). It can be seen visually from Fig. 4 that BDT droplets of eight concentrations change over the 8.13 s of the observing time. Therefore, 12% could be the appropriate concentration for BDT adjuvant for wheat field spraying based on the CA values during the observing time.

In the case of VP, the highest initial CA value is 89.99° (32%), the lowest is 60.10° (24%). However, the initial CA values of the other concentrations are approximate, 62.86°-67.63°. CA values of concentration 32% decrease slightly but keep at above 80° over the whole observing time. CA values of concentration 16% drop below 60° after 0.10 s and decrease slightly but stay above 40° during the left observing time (0.10 s – 8.13 s). CA values of concentration 20%, 24 and 28% are kept at around 36°- 45° after 3 s (Fig. 3 d), while the CA values of the remaining concentrations are 58°- 68° after 3 s (Fig. 3b). Fig. 5 shows the appearance shape of VP droplets dissipating on wheat leaf adaxial surface over 8.13 s. Therefore, 16% could be the appropriate concentration for VP adjuvant using on wheat field spraying based on the consideration of CA value during the observing time and crop producing cost.

In the case of NJF adjuvant, the initial CA values of all concentrations are between 41°- 80°. The lowest initial CA value is observed for concentration of 7‰, 39.94°. In the group of 0.2‰, 0.4‰, 0.6‰, 0.8‰ and 1‰, CA values of each concentration decrease slightly during the observing time (Fig. 6 a). It takes around 6 s for the CA Values of concentration 0.2‰ and 0.4‰ to drop below 40 °, but it only takes 0.5 s for CA values of concentration 0.6‰ and 0.8‰ to decrease below 40°. In the group of 2‰, 3‰, 4‰ and 5‰, the initial CA value are similar (48° - 61°) and CA values are below 20° after 1.6 s (Fig. 6 b). In the group of 6‰, 7‰, 8‰, 10 ‰ and 10‰, the initial CA value are between 41° -50° and CA values are below 20 ° in less than 1 s (Fig. 6 c). The lowest initial CA value is 39.94° (7‰) and the highest is 61.31 (2‰) in these two groups. Fig. 7 presents the appearance shape of NJF droplets on wheat leaf adaxial surface over

8.13 s. It can be seen that NJF droplets appearance shape changes notable on the wheat leaf adaxial surface under different concentrations. Therefore, the appropriate NJF concentration for spraying on wheat field could be 6‰ after the comprehensive consideration of initial CA value and the spreading time on wheat leaf.

In the case of LY adjuvant, the initial CA values of concentration 0.1‰, 0.2‰ and 0.3‰ are 68.32°, 54.25° and 57.59°, respectively. It takes around 5 s for CA value of concentration 0.1‰ to decrease below 20°, but it only takes less than 1 s for CA value of concentration 0.3‰ to drop below 20° (Fig. 6 d). Fig. 8 illustrates the appearance shape of LY droplets on wheat leaf adaxial surface under three concentrations. Therefore, the appropriate concentration for LY adjuvant to spray on wheat field could be 0.3‰ based on the analysis of initial CA value and the droplet dissipating time on wheat leaf surface.

Decrease of CA

Fig. 9 and Fig. 10 illustrate the initial CA ($t=0$ s) Values ($CA_{initial}$) and the final CA ($t \leq 8.13$ s, the final observing time is different because of physicochemical properties is different in the four adjuvants) values (CA_{final}) of the four adjuvants. In the case of BDT, the highest $CA_{initial}$ (Fig. 9a) and CA_{final} (Fig. 9b) are observed in the concentration of 2%, 90.63° and 66.53°, respectively. The lowest $CA_{initial}$ (Fig. 9a) and CA_{final} (Fig. 9b) are observed in the concentration of 12%, 69.53° and 44.56°, respectively. Significant differences between the highest and the lowest $CA_{initial}$ and CA_{final} are observed, respectively. However, the remaining $CA_{initial}$ and CA_{final} are not notably difference, respectively. In the case of VP, the highest $CA_{initial}$ (Fig. 9c) is observed in the concentration of 32% (89.99°), while the lowest is found in the concentration of 20% (60.20°). For CA_{final} (Fig. 9d), the highest (82.49°) and the lowest (35.16°) value are observed in the concentration of 32% and 28%, respectively. In the case of NJF $CA_{initial}$ (Fig. 10a), the concentration of 0.4‰ owns the highest value (79.16°), while the concentration of 7‰ has the lowest value 39.94°. For NJF CA_{final} (Fig. 10b), the highest and the lowest are 33.39° (0.2‰) and 6.79° (6‰), respectively. In the case of LY, $CA_{initial}$ (Fig. 10c) and CA_{final} (Fig. 10d) of the three observing concentrations are similar, respectively.

Fig. 11 presents the decrease of CA value ($CA_{initial} - CA_{final}$) between concentrations of the four adjuvants, respectively. Analysis of the case of BDT (Fig. 11a) shows that the top three highest CA decreases are observed in the concentrations of 10% (28.21°), 16% (26.30°), and 4% (26.09°), and the lowest decreases in the concentrations of 12% (15.48°), 8% (16.24°), and 6% (19.45°). Although the decrease of CA values between concentrations are observed in values, the differences between those decrease are not significant in statistically. Thus, the ability of BDT concentrations to reduce CA on wheat leaf adaxial surface is similar based on the difference between the CA decrease. In the case of VP (Fig. 11b), the top three high decreases are seen in the concentrations of 28% (29.66°), 16% (26.88°), and 24% (23.92°), and the top three low decreases are in the concentrations of 4% (1.19°), 8% (6.54°), and 12% (6.84°). The CA decreases of the top three lowest are notably lower than that of the highest. It can be further summarized as that the concentrations of 4%, 8%, and 12% have weak ability to reduce the CA on the wheat adaxial

surface, while the concentration of 28% has stronger ability to reduce the CA. In the case of NJF (Fig. 11c), significant difference of CA decrease is observed between concentrations of 0.4‰ (54.44°) and 7‰ (28.50°), corresponding to the highest and the lowest decrease, respectively. However, the significant difference of CA decrease is not observed in the remaining NJF concentrations. Hence, the concentration of 6‰ has the best performance on reducing CA on wheat leaf adaxial surface in the aspect of the CA decreases value and spreading time on the leaves. In the case of LY (Fig. 11d), the differences of CA decreases between the $CA_{initial}$ and CA_{final} of the three observed concentrations are similar.

Optimal concentration selection

Combined with the results of section 3.1 and 3.2, it could be seen that the optimal concentration for field spraying application might be different because of judgement criterion. $CA_{initial}$, CA_{final} spreading time, and concentration should be under consideration when optimizing the appropriate concentrations for field application.

Discussion

Tank-mix adjuvants can effectively mitigate the evaporation, drift, and rebound of the spray solution (Preftakes et al., 2019; Sijts & Bonn, 2020), when UAVs perform spraying operations, and enhance the retention, diffusion, and wetting effects of droplets on the surface of crop leaves (Klevens, 1948; Donbrow & Jan, 2011; Ryckaert et al., 2008). Wheat, as a superhydrophobic crop (Dorr et al., 2015), has a unique leaf structure that impedes the spreading and retention of droplets on its surface. In this work, we focus on studying the changes of CA on wheat leaf surface after adding different types of tank-mix adjuvants and the variation of CA under different concentrations of the same tank-mix adjuvant. The results indicate that adding tank-mix adjuvants to tap-water significantly reduces the CA of droplets and improves the diffusion performance of droplets. Different types of tank-mix adjuvants have distinct effects on the reduction of droplets' CA and the rate of liquid diffusion. When using the same tank-mix adjuvant, concentration is also a crucial factor affecting the CA values and diffusion of droplets on wheat leaf surfaces.

Tank-mix adjuvants based on surfactants have the ability of lowering droplet surface tension (Hazen, 2000), which is a key parameter to characterize the physicochemical properties of droplets (Arand et al., 2018). The decrease of surface tension results in the reduction of droplets' CA and facilitating the spreading of droplet on solid surfaces. In this study, we measured and analysed the dynamic CA values of droplets on wheat surface after adding adjuvants. The results demonstrate that the addition of tank-mix adjuvants BDT, VP, NJF, and LY reduce the CA of droplets on wheat leaf surface, but the degree of influence vary. LY (alkoxy modified polytrisiloxane) has the most pronounced effect on reducing the CA of droplets. This is in line with previous studies that organosilicon adjuvants can substantially lower the surface tension of pesticide solutions and improve the spreading efficiency of pesticides (Policello & Murphy, 1993). Although NJF (hyperbranched fatty alcohol ether modified polymer) reduce the CAs in a short time as LY does but with a much higher concentrations (6‰). BDT is a plant oil-based adjuvant that can reduce the

CA and augment the wetting property of pesticides by lowering the surface tension of droplets and dissolving the wax layer and cuticle layer of plant leaves. Xiao et al. report that plant oil-based adjuvants can significantly improve the droplet coverage and retention of defoliants in cotton leaves (Xiao et al., 2019). Yuan et al explore that the application of Green-peel orange essential oil (GOEO) as a spray adjuvant has great potential to enhance the deposition and penetration of pesticides on the leaf surface, so that it would increase the pesticide utilization rate (Yuan et al., 2019). VP is a mineral oil-based adjuvant, which has a similar effect as BDT and other plant oil-based adjuvants. A previous study shows that adding mineral oil-based and surfactant to the biopesticide mixture can reduce the CA value and surface tension of the droplet, resulting in greater diffusion of the droplet in leaves (Santos et al., 2019). Our experiments also indicate that plant oil-based and mineral oil-based adjuvants can effectively reduce the CA value of droplets, which would improve the efficiency of pesticide.

As mentioned above, the concentration of tank-mix adjuvant is an important factor that influence the performance of pesticides. For NJF and LY, at different concentrations, the CA value of droplets decreased markedly. For oil-based adjuvants BDT and VP, within a certain concentration range, the CA declined gradually with increasing concentration. The data show that after adding BDT adjuvant, the CA of the droplets decreased significantly firstly, and then gradually decreased and stabilized over time. Within the observation time, the initial CA value reached the lowest when the adjuvant concentration reached 12%. Under the condition of concentration less than 12%, the initial CA value decreased gradually with the increase of the adjuvant concentration. When the concentration exceeded 12%, the initial CA value gradually increased instead of decreasing. Different concentrations of VP also had the effect of reducing CA. The initial CA value reached the lowest when the concentration was 24%. At this point, as the concentration continued to increase, the CA value began to rise, and when the concentration reached 32%, the CA value remained above 80° throughout the observation period, which was significantly different from other concentrations. In addition, during the entire observation period, when the concentration was 16%, 20%, 24%, or 28%, the adjuvant has a similar effect on the initial CA and the final CA of the droplet. Thus, concentration of 16% might be appropriate than the other concentration for VP in practice after consideration of spreading efficiency and cost.

Adjuvants with high concentration may have negative effects on pesticide absorption (Buick, Buchan & Field, 1993). Both BDT and VP tank-mix adjuvants showed the phenomenon that the effect was worse at high concentrations than at lower concentrations. It may be due to the concentration of adjuvant solution reaching Critical Micelle Concentration (CMC), which causes the droplet to produce micelle force and prevents the CA from decreasing (Wang & Liu, 2007). Further experiments on exploring the relationship of CMC of tank-mix adjuvant and CA on wheat leaves are suggested to carry out in the future work, aiming to obtain more reliable and accurate experimental results for practical application.

Conclusions

In this paper, we measured the effect of different concentrations of adjuvants on droplet CA, and obtained the optimal use concentration of BDT, VP, NJF and LY by considering CA changes, droplet diffusion time and other factors comprehensively. Firstly, we investigated dynamic CA of four typical tank-mix adjuvants on wheat leaf adaxial surface under different concentrations to optimize the appropriate concentrations for field application, respectively. We observed that adjuvant concentrations had significant initial CA values and CA changes with time. CA values of BDT and VP changed during the measurement time for all concentrations with a slight CA decrease from 0-8.13 s, while these of NJF and LY with a rapid decrease during the observing time. CA differences were observed among concentrations within the same adjuvant. The appropriate concentrations of the four adjuvants for wheat field application were 12% (BDT), 16%(VP), 6‰ (NJF) and LY (0.3‰) based on the CA dissipation time and values observed from indoor experiments. Alkoxy modified polytrisiloxane adjuvant (LY) could be an appropriate adjuvant for field application on wheat field by considering spreading efficiency and cost. In conclusion, we advise that CA values should be measured to optimize appropriate concentration for field application to obtain satisfactory biological control efficacy. Furthermore, not only the initial CA value is important when assessing the wettability of different liquids and optimizing the appropriate concentration for a specific liquid on the same crop leaf surface, but also what happens with the liquid drops over the observing time.

ACKNOWLEDGEMENTS

We thank Mr. Xiaochao Liu, Mr. yifan zhang, and Mr. Xintao Du for their kind help for this work.

ADDITIONAL INFORMATION AND DECLARATIONS

Funding

This work was funded by National Natural Science Foundation of China (no.32201659). The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

Grant Disclosures

The following grant information was disclosed by the authors: National Natural Science Foundation of China (no.32201659).

Competing Interests

Hanxue Zhou is employed by Anyang Quanfeng Biotechnology Co., Ltd.

Author Contributions :

- Yanhua Meng conceived and designed the experiments, performed the experiments, analyzed the data, prepared figures and/or tables, authored or reviewed drafts of the paper, and approved the final draft.

- 355● Qiufang Wu conceived and designed the experiments, analyzed the data, performed the experiments
356 ,authored or reviewed drafts of the paper, and approved the final draft.
- 357● Hanxue Zhou conceived and designed the experiments ,analyzed the data,performed the computation work,
358 authored orreviewed drafts of the paper, and approved the final draft.
- 359● Hongyan Hu conceived and designed the experiments, analyzed the data, authored or reviewed drafts of
360 the paper, and approved the final draft.

Data Availability Statement:

363 The following information was supplied regarding data availability:The raw measurements are
364 available in the Supplemental Files.

Reference

- 367 **Arand K, Asmus E, Popp C, Schneider D, Riederer M. 2018.** The Mode of Action of
368 Adjuvants—Relevance of Physicochemical Properties for Effects on the Foliar Application,
369 Cuticular Permeability, and Greenhouse Performance of Pinoxaden. *Journal of Agricultural*
370 *and Food Chemistry* 66:5770–5777.. DOI: 10.1021/acs.jafc.8b01102.
- 371
- 372 **Buick RD, Buchan GD, Field RJ. 1993.** The role of surface tension of spreading droplets in
373 absorption of a herbicide formulation via leaf stomata. *Pesticide Science* 38:227–235.. DOI:
374 10.1002/ps.2780380218.
- 375
- 376 **Cavallaro, A., Carbonell-Silletta, L., Burek, A., Goldstein, G., Scholz, F. G., & Bucci, S. J.**
377 **2022.** Leaf surface traits contributing to wettability, water interception and uptake of above-
378 ground water sources in shrubs of Patagonian arid ecosystems. *Annals of botany*, 130(3),
379 409–418.. doi:10.1093/aob/mcac042.
- 380
- 381 **Chen S, Lan Y, Zhou Z, Ouyang F, Wang G, Huang X, Deng X, Cheng S. 2020.** Effect of
382 Droplet Size Parameters on Droplet Deposition and Drift of Aerial Spraying by Using Plant
383 Protection UAV. *Agronomy* 10:195.. DOI: 10.3390/agronomy10020195.
- 384
- 385 **Da Silva Santos RT, Vechia JFD, Dos Santos CAM, Almeida DP, Da Costa Ferreira M.**
386 **2021.** Relationship of contact angle of spray solution on leaf surfaces with weed control.
387 *Scientific Reports* 11.. DOI: 10.1038/s41598-021-89382-2.
- 388 **Donbrow M, Jan ZA. 2011.** Refractometric Determination of the Critical Micelle Concentration
389 of Non-Ionic Surface-Active Agents. *Journal of Pharmacy and Pharmacology* 15:825–830..
390 DOI: 10.1111/j.2042-7158.1963.tb12887.x.
- 391 **Dorr GJ, Wang S, Mayo LC, Mccue SW, Forster WA, Hanan J, He X. 2015.** Impaction of
392 spray droplets on leaves: influence of formulation and leaf character on shatter, bounce and
393 adhesion. *Experiments in Fluids* 56.. DOI: 10.1007/s00348-015-2012-9.
- 394 **Fang, H., Zhang, Z., Xiao, S., & Liu, Y. 2019.** Influence of leaf surface wettability on droplet
395 deposition effect of rape leaves and their correlation.*Journal of Agriculture and Food*
396 *Research* 100011..DOI:10.1016/j.jafr.2019.100011.
- 397 **Fernández V, Sancho-Knapik D, Guzmán P, et al. 2014.**Wettability, polarity, and water
398 absorption of holm oak leaves: effect of leaf side and age. *Plant Physiol.* 166(1):168-180..
399 doi:10.1104/pp.114.242040.

- Fountain, M.T., Harris, A.L., & Cross, J.V. 2010.** The use of surfactants to enhance acaricide control of *Phytonemus pallidus* (Acari: Tarsonemidae) in strawberry. *Crop Protection* 29:1286-1292..DOI:10.1016/J.CROPRO.2010.06.01.
- Hazen, J. L. 2000.** Adjuvants—Terminology, Classification, and Chemistry. *Weed Technology*, 14(4), 773–784. Cambridge University Press..DOI: [10.1614/0890-037x\(2000\)014\[0773:atcac\]2.0.co;2](https://doi.org/10.1614/0890-037x(2000)014[0773:atcac]2.0.co;2).
- He L, Ding L, Zhang P, Li B, Mu W, Liu F. 2021.** Impact of the equilibrium relationship between deposition and wettability behavior on the high-efficiency utilization of pesticides. *Pest Management Science* 77:2485–2493.. DOI: 10.1002/ps.6279.
- Jeevahan J, Chandrasekaran M, Britto Joseph G, Durairaj RB, Mageshwaran G. 2018.** Superhydrophobic surfaces: a review on fundamentals, applications, and challenges. *Journal of Coatings Technology and Research* 15:231–250.. DOI: 10.1007/s11998-017-0011-x.
- Jensen, P.K., & Olesen, M.H. 2014.** Spray mass balance in pesticide application:A review. *Crop Protection* 23-31.DOI:10.1016/J.CROPRO.2014.03.006.
- Klevens HB. 1948.** Critical Micelle Concentrations as Determined by Refraction. *The Journal of Physical and Colloid Chemistry* 52:130–148.. DOI: 10.1021/j150457a013.
- Lou Z, Xin F, Han X, Lan Y, Duan T, Fu W. 2018.** Effect of Unmanned Aerial Vehicle Flight Height on Droplet Distribution, Drift and Control of Cotton Aphids and Spider Mites. *Agronomy* 8:187.. DOI: 10.3390/agronomy8090187.
- Márquez DA, Stuart-Williams H, Farquhar GD. 2021.** An improved theory for calculating leaf gas exchange more precisely accounting for small fluxes. *Nature Plants* 7:317–326.. DOI: 10.1038/s41477-021-00861-w.
- Matthews GA, Thomas N. 2000.** Working towards more efficient application of pesticides. *Pest Management Science* 56:974–976.. DOI: 10.1002/1526-4998(200011)56:11<974::aid-ps231>3.0.co;2-4.
- Meng Y, Zhong W, Liu C, Su J, Su J, Lan Y, Wang Z, Wang M. 2022.** UAV spraying on citrus crop: impact of tank-mix adjuvant on the contact angle and droplet distribution. *PeerJ* 10:e13064.. DOI: 10.7717/peerj.13064.
- Meng, Y., Lan, Y., Mei, G., Guo, Y., Song, J., & Wang, Z. 2018.** Effect of aerial spray adjuvant applying on the efficiency of small unmanned aerial vehicle for wheat aphids control. *International Journal of Agricultural and Biological Engineering*, 11, 46-53..DOI:10.25165/IJABE.V11I5.4298.
- Nairn JJ, Forster WA, Van Leeuwen RM. 2011.** Quantification of physical (roughness) and chemical (dielectric constant) leaf surface properties relevant to wettability and adhesion. *Pest Management Science* 67:1562–1570.. DOI: 10.1002/ps.2213.
- Papierowska E, Szporak-Wasilewska S, Szewińska J, Szatyłowicz J, Debaene G, Utratna M. 2018.** Contact angle measurements and water drop behavior on leaf surface for several deciduous shrub and tree species from a temperate zone. *Trees* 32:1253–1266.. DOI: 10.1007/s00468-018-1707-y.
- Peirce CAE, Priest C, Mcbeath TM, Mclaughlin MJ. 2016.** Uptake of phosphorus from surfactant solutions by wheat leaves: spreading kinetics, wetted area, and drying time. *Soft Matter* 12:209–218.. DOI: 10.1039/c5sm01380a.
- Policello GA, Murphy GJ. 1993.** The influence of co-surfactants on the spreading ability of organosilicone wetting agents. *Pesticide Science* 37:228–230.. DOI: 10.1002/ps.2780370226.

- 445 **Preftakes CJ, Schleier JJ, Kruger GR, Weaver DK, Peterson RKD. 2019.** Effect of
446 insecticide formulation and adjuvant combination on agricultural spray drift. *PeerJ*
447 7:e7136.. DOI: 10.7717/peerj.7136.
- 448 **Quetzeri-Santiago MA, Castrejón-Pita JR, Castrejón-Pita AA. 2020.** On the analysis of the
449 contact angle for impacting droplets using a polynomial fitting approach. *Experiments in*
450 *Fluids* 61.. DOI: 10.1007/s00348-020-02971-1.
- 451 **Rowlandson T, Gleason M, Sentelhas P, Gillespie T, Thomas C, Hornbuckle B. 2015.**
452 Reconsidering Leaf Wetness Duration Determination for Plant Disease Management. *Plant*
453 *Disease* 99:310–319.. DOI: 10.1094/pdis-05-14-0529-fe.
- 454 **Ryckaert B, Spanoghe P, Heremans B, Haesaert G, Steurbaut W. 2008.** Possibilities To Use
455 Tank-Mix Adjuvants for Better Fungicide Spreading on Triticale Ears. *Journal of*
456 *Agricultural and Food Chemistry* 56:8041–8044.. DOI: 10.1021/jf8005257.
- 457 **Santos CAMD, Santos RTDS, Della’Vechia JF, Griesang F, Polanczyk RA, Ferreira MDC.**
458 **2019.** Effect of addition of adjuvants on physical and chemical characteristics of Bt
459 bioinsecticide mixture. *Scientific Reports* 9.. DOI: 10.1038/s41598-019-48939-y.
- 460 **Sanyal D, Bhowmik PC, Reddy KN. 2006.** Influence of leaf surface micromorphology, wax
461 content, and surfactant on primisulfuron droplet spread on barnyardgrass (*Echinochloa crus-*
462 *galli*) and green foxtail (*Setaria viridis*) *Weed Science*. 54(4):627-633.. DOI: 10.1614/ws-
463 05-173r.1.
- 464 **Sijs R, Bonn D. 2020.** The effect of adjuvants on spray droplet size from hydraulic nozzles. *Pest*
465 *Management Science* 76:3487–3494.. DOI: 10.1002/ps.5742.
- 466 **Sobiech Ł, Grzanka M, Skrzypczak G, Idziak R, Włodarczyk S, Ochowiak M. 2020.** Effect
467 of Adjuvants and pH Adjuster on the Efficacy of Sulcotrione Herbicide. *Agronomy* 10:530..
468 DOI: 10.3390/agronomy10040530.
- 469 **Song Y, Huang Q, Huang G, Liu M, Cao L, Li F, Zhao P, Cao C. 2022.** The Effects of
470 Adjuvants on the Wetting and Deposition of Insecticide Solutions on Hydrophobic Wheat
471 Leaves. *Agronomy* 12:2148.. DOI: 10.3390/agronomy12092148.
- 472 **Wang, C., & Liu, Z.Q. 2007.** Foliar uptake of pesticides : Present status and future challenge.
473 *Pesticide Biochemistry and Physiology*, 87, 1-8..DOI:10.1016/J.PESTBP.2006.04.004.
- 474 **Wang, S., Wang, H., Tong, L., Chun, L., Zhong, X., & Zhou, Y..2016.** WETTING
475 PROPERTY REPRESENTATION OF PESTICIDES ON THE CROP LEAF SURFACES
476 1027-1033.
- 477 **Wang, X., He, X., Song, J., Wang, Z., Changling, W., Wang, S., Wu, R., & Yanhua, M.**
478 **2018.** Drift potential of UAV with adjuvants in aerial applications. *International Journal of*
479 *Agricultural and Biological Engineering*, 11, 54-58..DOI:10.25165/IJABE.V11I5.3185.
- 480 **Xiao Q, Xin F, Lou Z, Zhou T, Wang G, Han X, Lan Y, Fu W. 2019.** Effect of Aviation
481 Spray Adjuvants on Defoliant Droplet Deposition and Cotton Defoliation Efficacy Sprayed
482 by Unmanned Aerial Vehicles. *Agronomy* 9:217.. DOI: 10.3390/agronomy9050217.
- 483 **Yan X, Wang M, Zhu Y, Shi X, Liu X, Chen Y, Xu J, Yang D, Yuan H. 2021.** Effect of
484 Aviation Spray Adjuvant on Improving Control of Fusarium Head Blight and Reducing
485 Mycotoxin Contamination in Wheat. *Agriculture* 11:1284.. DOI:
486 10.3390/agriculture11121284.
- 487 **Yuan W, Zhao P, Chen H, Wang L, Huang G, Cao L, Huang Q. 2019.** Natural green-peel
488 orange essential oil enhanced the deposition, absorption and permeation of prochloraz in
489 cucumber. *RSC Advances* 9:20395–20401.. DOI: 10.1039/c9ra02809a.

- 490 **Zhang C, Zhao X, Lei J, Ma Y, Du F. 2017.**The wetting behavior of aqueous surfactant
491 solutions on wheat (*Triticum aestivum*) leaf surfaces. *Soft Matter*.13(2):503-513.
492 .doi:10.1039/c6sm02387h.
- 493 **Zhang, X., & Xiong, L. 2021.** Effect of adjuvants on the spray droplet size of pesticide dilute
494 emulsion. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 619,
495 126557..DOI:10.1016/J.COLSURFA.2021.126557.
- 496 **Zhao R, Yu M, Sun Z, Li L, Shang H, Xi W, Li B, Li Y, Xu Y, Wu X. 2022.** Using tank-mix
497 adjuvant improves the physicochemical properties and dosage delivery to reduce the use of
498 pesticides in unmanned aerial vehicles for plant protection in wheat. *Pest Management*
499 *Science* 78:2512–2522.. DOI: 10.1002/ps.6879.
- 500 **Zhu F, Cao C, Cao L, Li F, Du F, Huang Q. 2019.** Wetting Behavior and Maximum Retention
501 of Aqueous Surfactant Solutions on Tea Leaves. *Molecules* 24:2094.. DOI:
502 10.3390/molecules24112094.
503

Figure 1

Sketch map of CA.

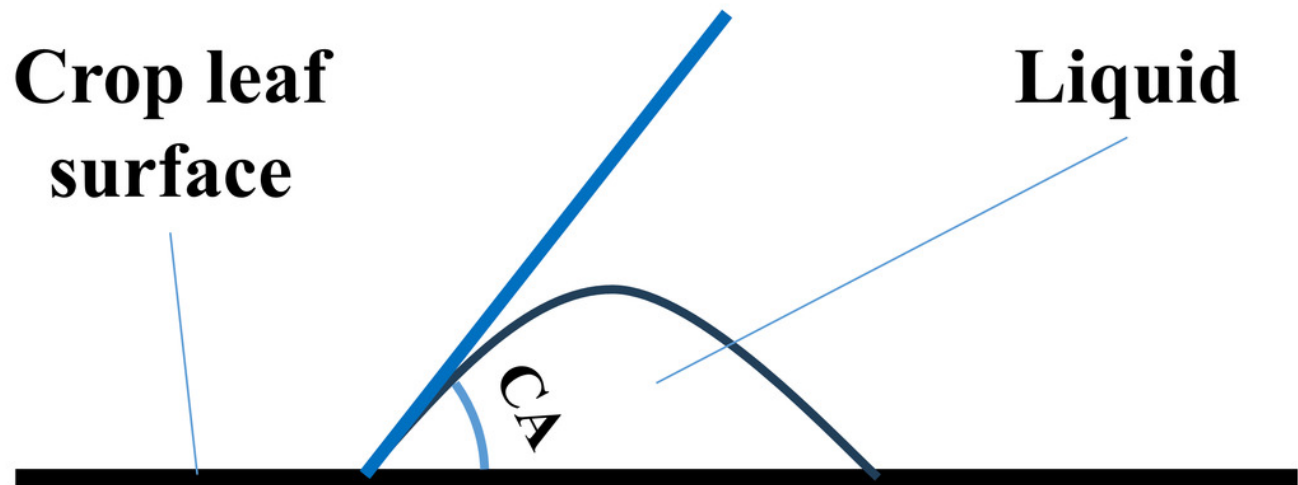


Figure 2

Tap-water droplets spread on wheat leaf adaxial surface during the observing time.



Figure 3

Dynamic CA of BDT and VP under different solution concentration, respectively.

(a): Contact angle changes over time after adding 2% -8% BDT tank-mix adjuvant. (b): Contact angle changes over time after adding 4% -16% VP tank-mix adjuvant. (c): Contact angle changes over time after adding 10% -16% BDT tank-mix adjuvant. (d): Contact angle changes over time after adding 20% -32% VP tank-mix adjuvant.

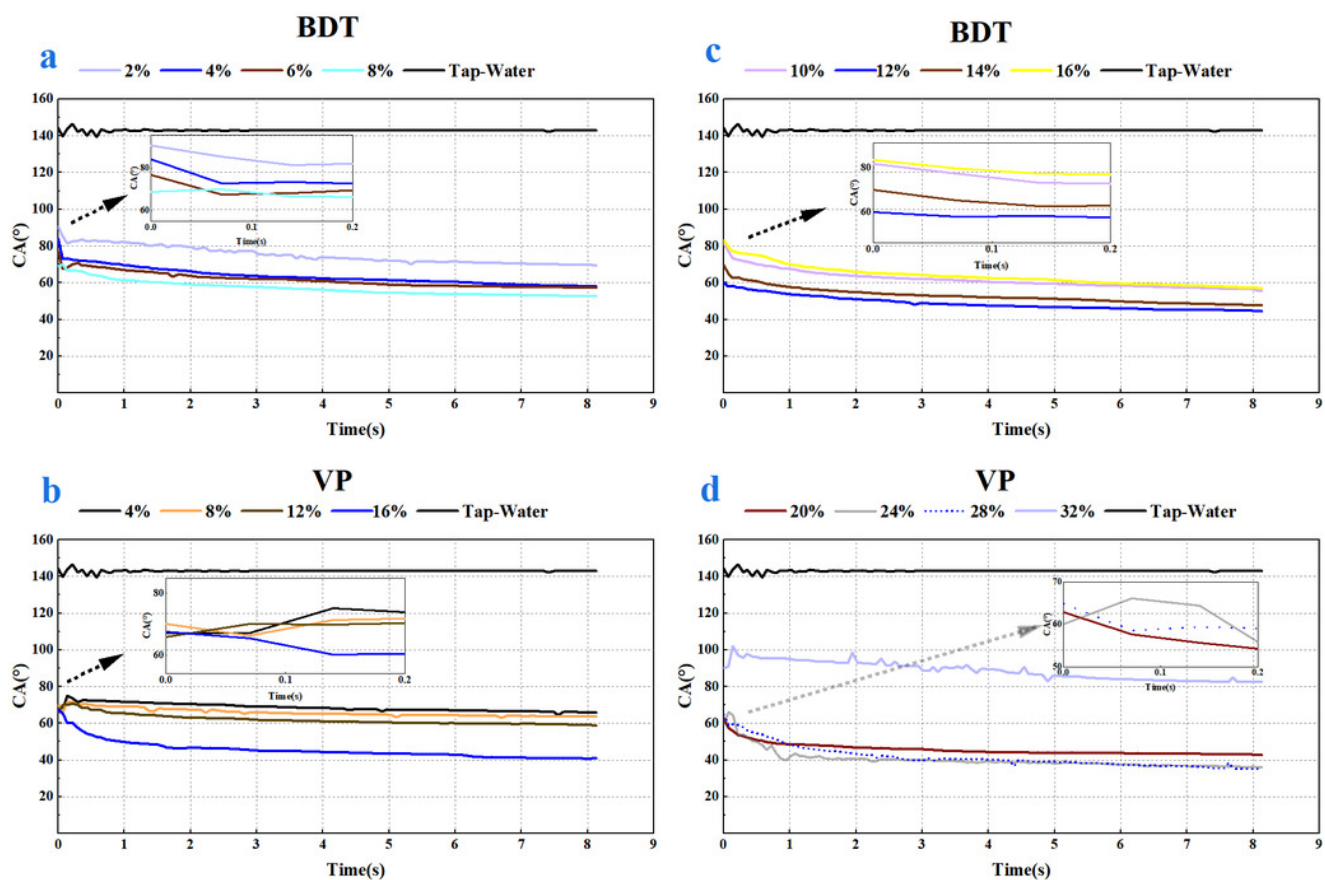


Figure 4

BDT droplets spread on wheat leaf adaxial surface during the observing time.

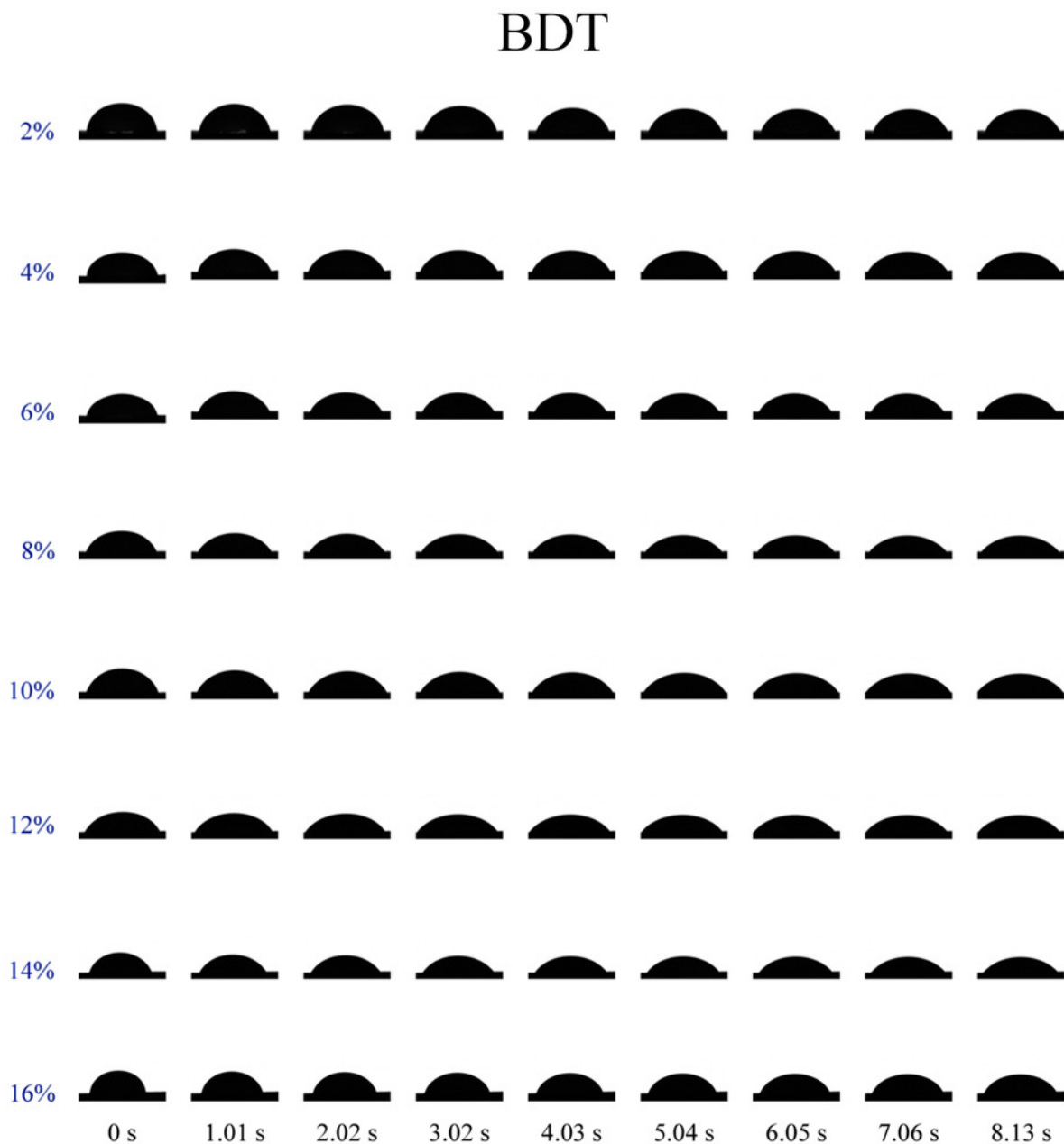


Figure 5

VP droplets spread on wheat leaf adaxial surface during the observing time.

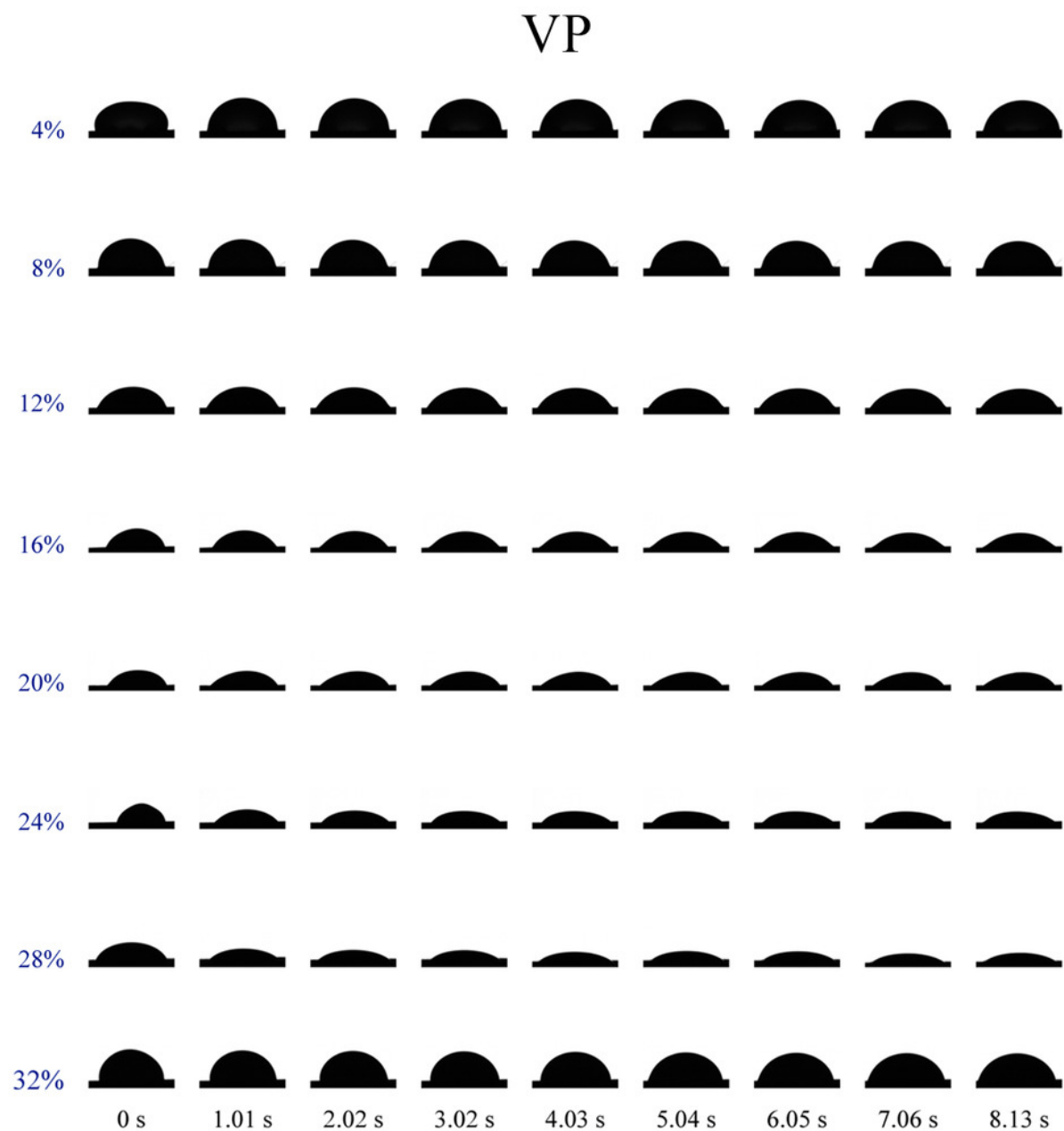


Figure 6

Dynamic CA of NJF (a, b and c) and LY (d) under different solution concentration, respectively.

(a): Contact angle changes over time after adding 0.2‰ -1‰ NJF tank-mix adjuvant. (b): Contact angle changes over time after adding 2‰ -5‰ NJF tank-mix adjuvant. (c): Contact angle changes over time after adding 6‰ -10‰ NJF tank-mix adjuvant. (d): Contact angle changes over time after adding 0.1‰ -0.3‰ LY tank-mix adjuvant.

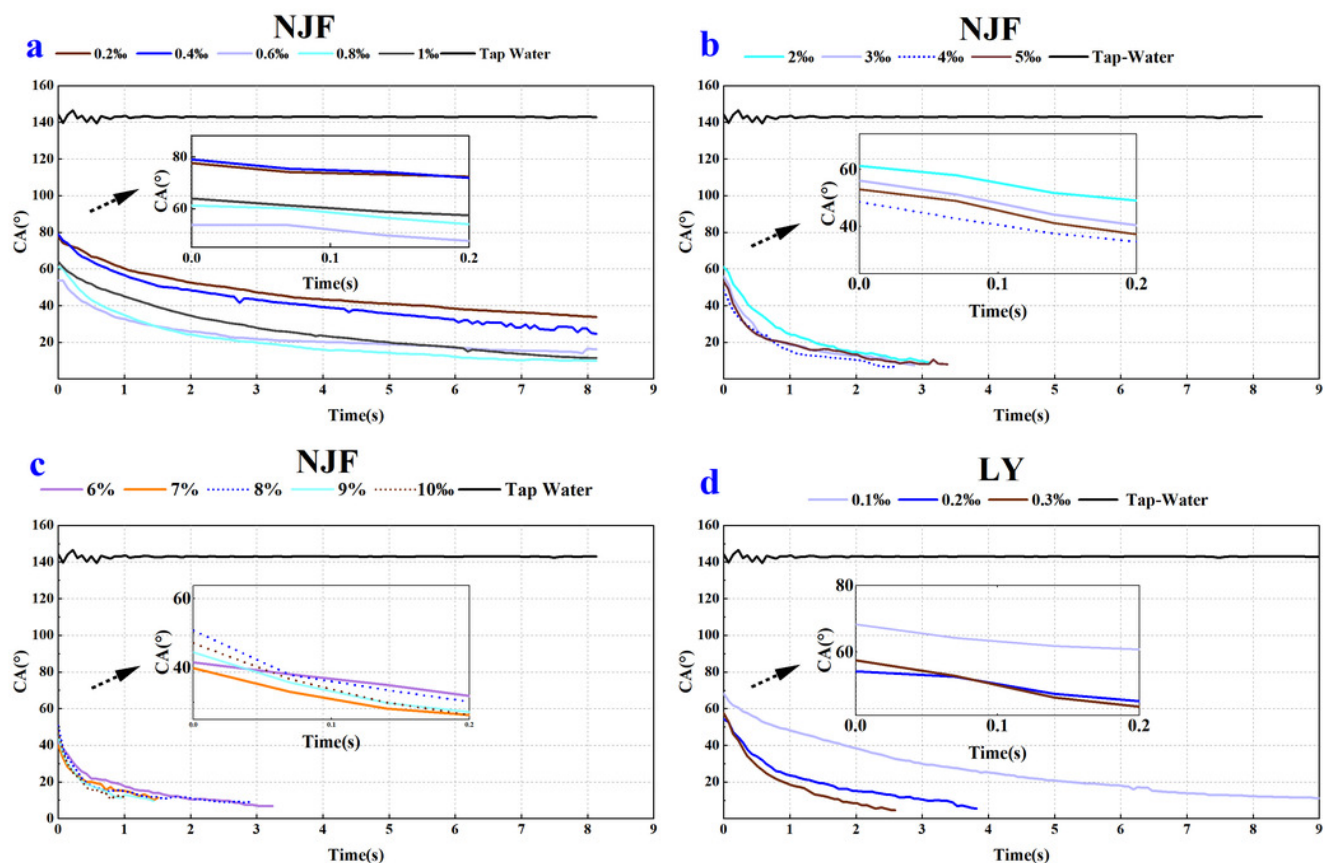


Figure 7

NJF droplets spread on wheat leaf adaxial surface during the observing time.

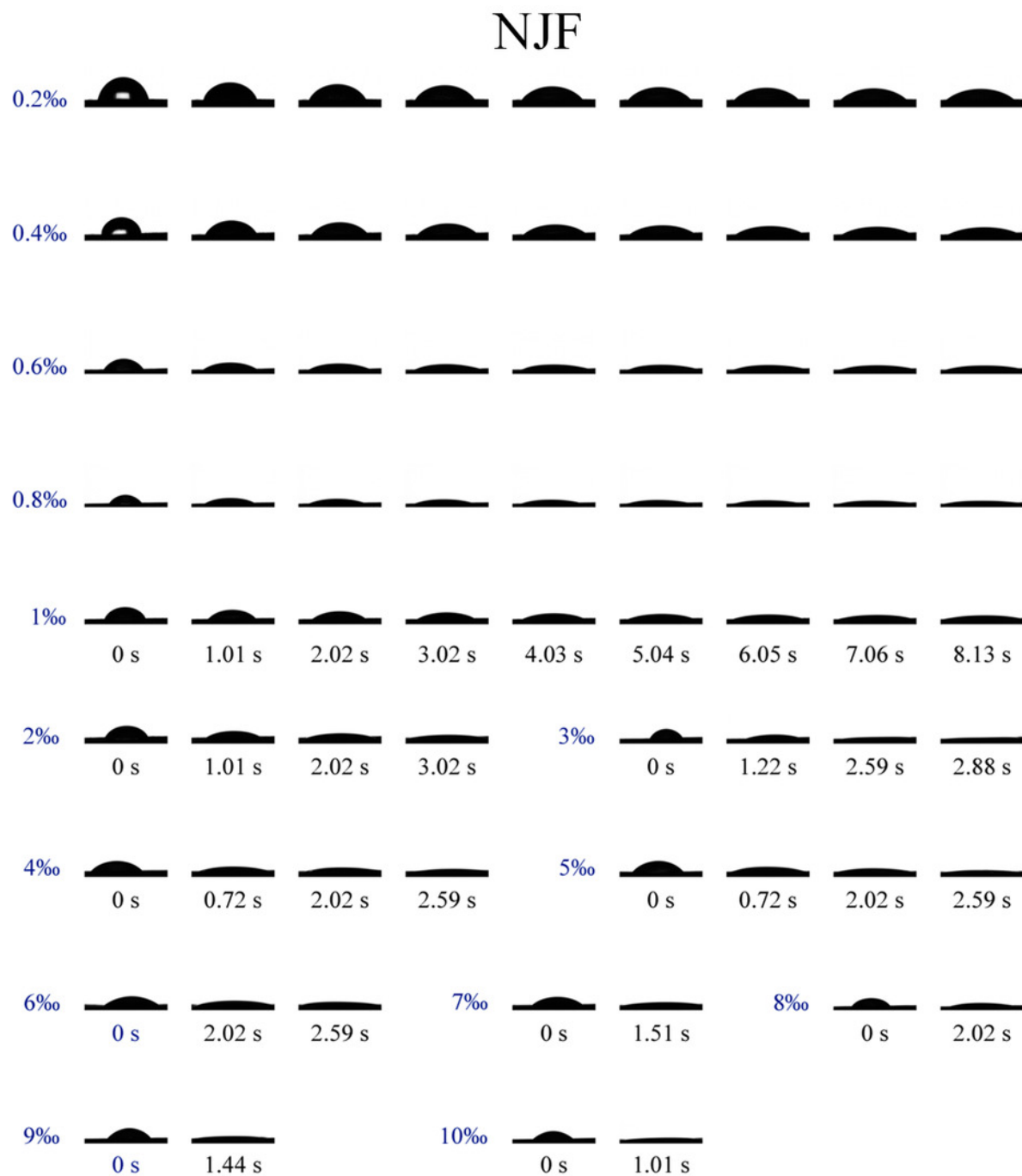


Figure 8

LY droplets spread on wheat leaf adaxial surface during the observing time.

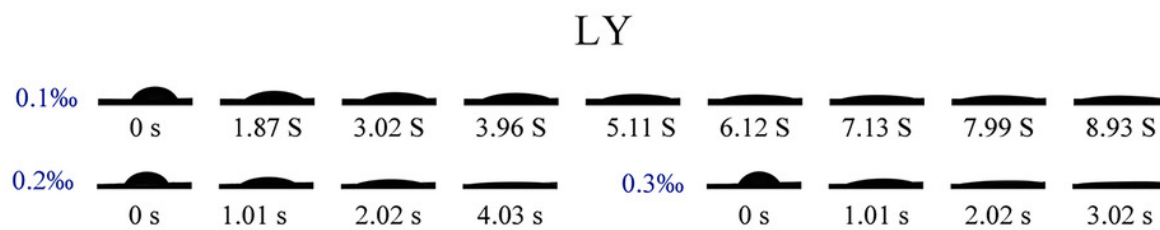


Figure 9

The initial CA and final CA of BDT and VP under different solution concentration

(a) The initial CA after adding 2% -16% BDT tank-mix adjuvant. (b) The final CA after adding 2% -16% BDT tank-mix adjuvant. (c) The initial CA after adding 4% -32% VP tank-mix adjuvant. (d) The final CA after adding 4% -32% VP tank-mix adjuvant.

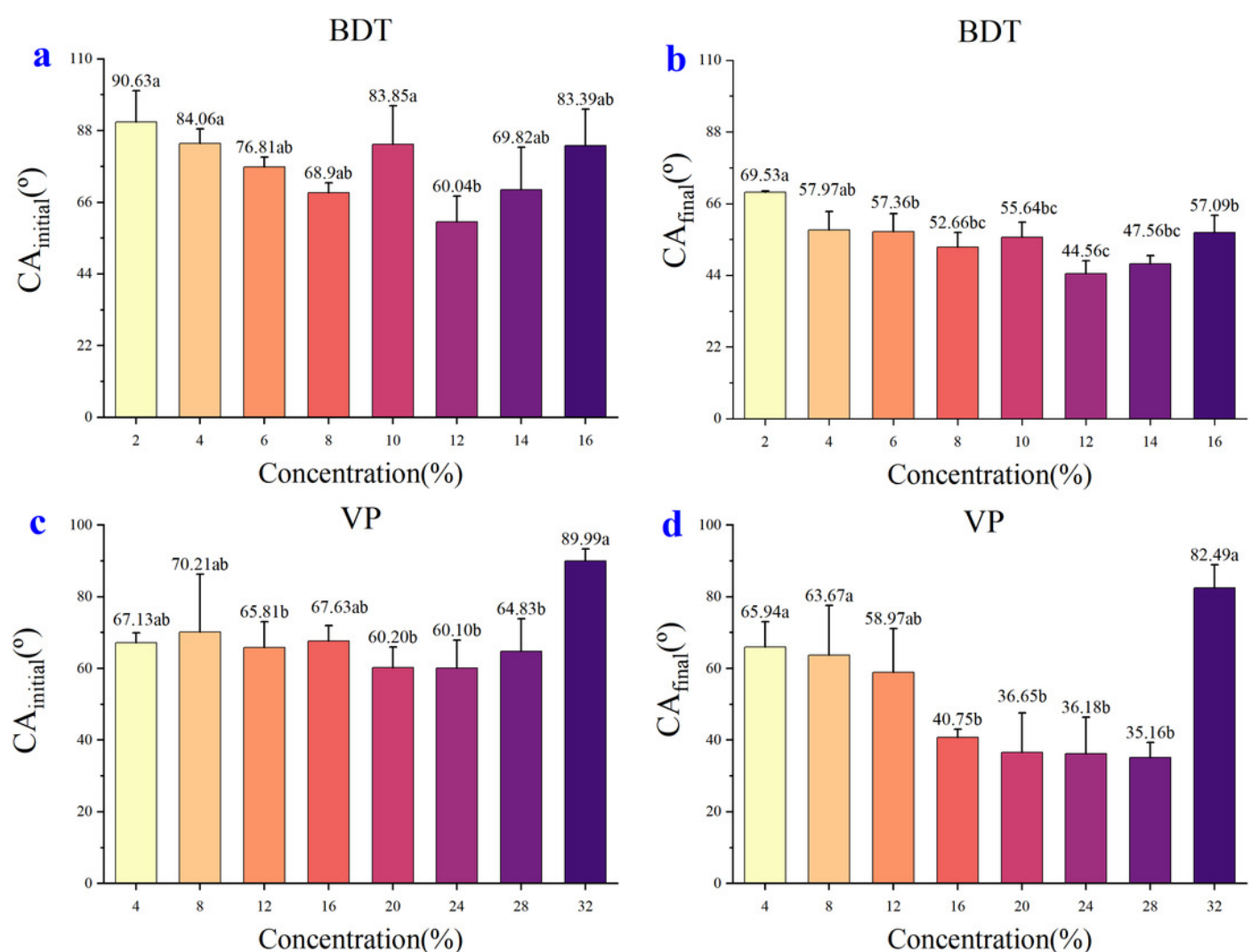


Figure 10

The initial CA and final CA of BDT and VP under different solution concentration

(a) The initial CA after adding 0.2‰ -16‰ NJF tank-mix adjuvant. (b) The final CA after adding 0.2‰ -16‰ NJF tank-mix adjuvant. (c) The initial CA after adding 0.1‰ -0.3‰ LY tank-mix adjuvant. (d) The final CA after adding 0.1‰ -0.3‰ LY tank-mix adjuvant.

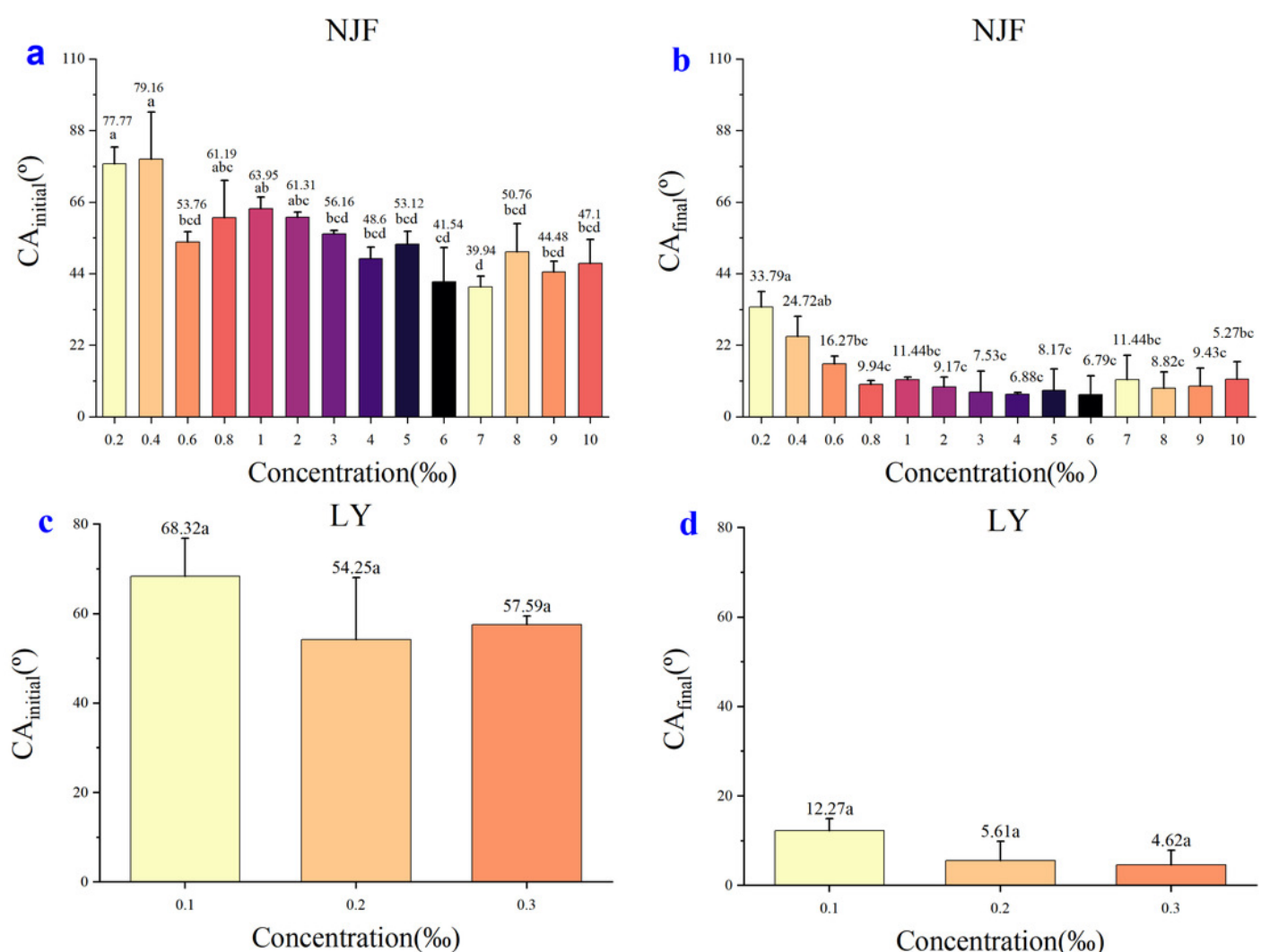


Figure 11

Difference of CA value changes ($CA_{\text{initial}} - CA_{\text{final}}$) from the initial measuring time (t_{initial}) to final measuring time (t_{final}).

(a) Change in CA value after adding 2% -16% BDT tank-mix adjuvant. (b) Change in CA after adding 4% -32% VP tank-mix adjuvant. (c) Change in CA after adding 0.2‰ -10‰ NJF tank-mix adjuvant. (d) Change in CA after adding 0.1‰ -0.3‰ LY tank-mix adjuvant.

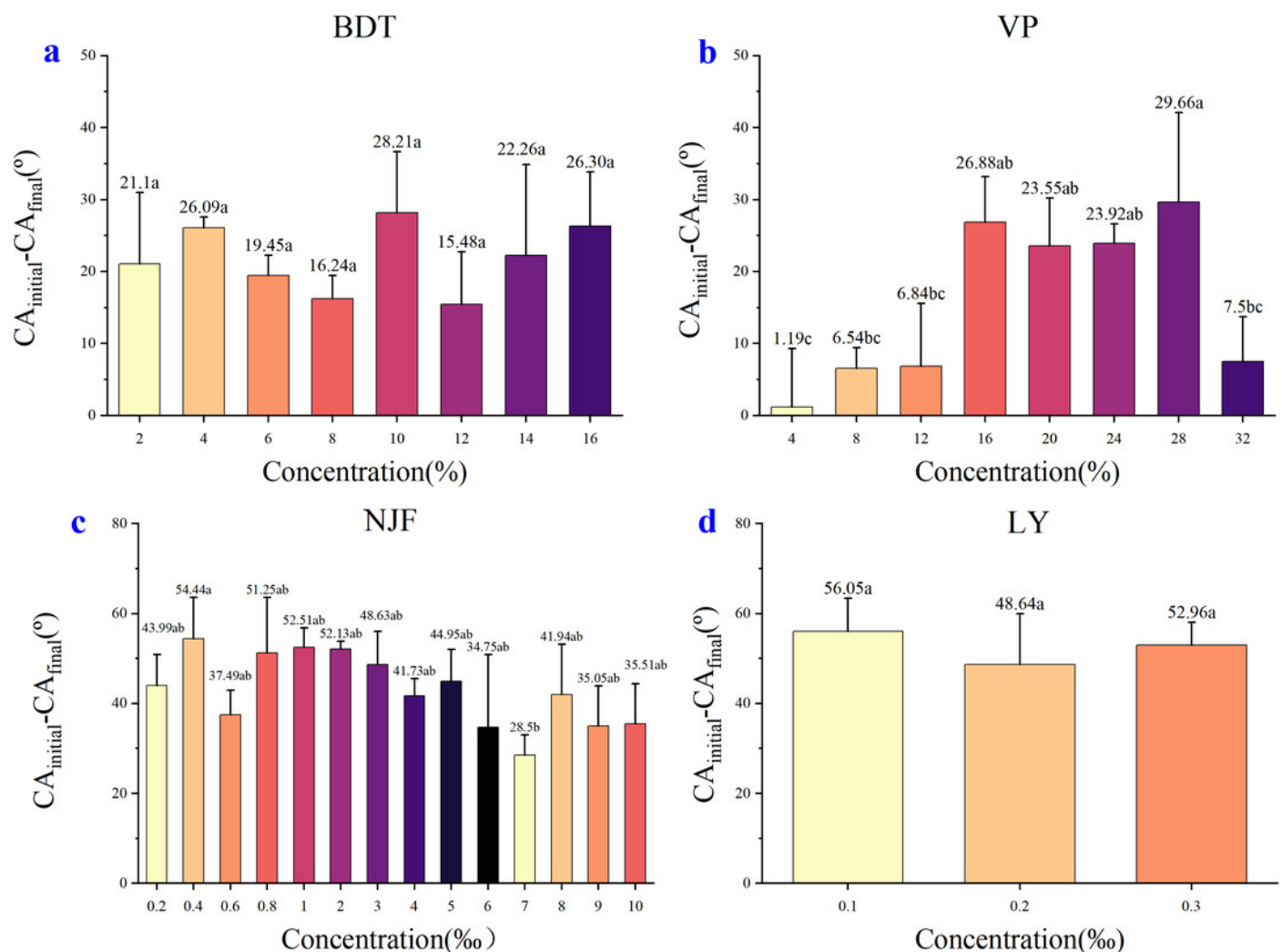


Table 1(on next page)

Solution concentration of the adopted tank-mix adjuvants, and the corresponding observing time and number of CA.

Note: Observing time t_{initial} indicates the first measured CA, while t_{final} is the last measured CA.

| Adjuvant | Solution concentration | Observing time (s) | | Number of measured CAs of each solution concentration |
|----------|--|--------------------|-------------|---|
| | | $t_{initial}$ | t_{final} | |
| BDT | 2%, 4%, 6%, 8%, 10%, 12%, 14%, 16% | 0 | 8.13 | 114 |
| | 4%, 8%, 12%, 16%, 20%, 24%, 28%, 32% | 0 | 8.13 | 114 |
| | 0.2‰, 0.4‰, 0.6‰, 0.8‰, 1‰ 2‰ | 0 | 8.13 | 114 |
| NJF | 3‰ | 0 | 3.10 | 44 |
| | 4‰ | 0 | 2.88 | 37 |
| | 5‰ | 0 | 2.88 | 37 |
| | 6‰ | 0 | 3.38 | 44 |
| | 7‰ | 0 | 2.30 | 33 |
| | 8‰ | 0 | 1.51 | 23 |
| | 9‰ | 0 | 2.95 | 42 |
| | 10‰ | 0 | 1.44 | 21 |
| | 0.1‰ | 0 | 1.01 | 15 |
| | 0.2‰ | 0 | 8.06 | 113 |
| LY | 0.3‰ | 0 | 3.82 | 54 |
| | | 0 | 2.59 | 37 |

1
2

Table 2 (on next page)

The criterion of appropriate concentration optimization.

Note: CA means Contact angle, while CC denotes the corresponding concentration.

$CA_{decrease} = CA_{initial} - CA_{final}$.

1

| Adjuvant | BDT | | VP | | NJF | | LY | |
|--------------------------------|-------|-----|-------|-----|-------|------|-------|------|
| Judgment Criterion | CA(°) | CC | CA(°) | CC | CA(°) | CC | CA(°) | CC |
| Lowest CA _{initial} | 60.04 | 12% | 60.20 | 20% | 39.94 | 7‰ | 54.25 | 0.2‰ |
| Lowest CA _{final} | 44.56 | 12% | 35.16 | 28% | 6.79 | 6‰ | 4.62 | 0.3‰ |
| Maximum CA _{decrease} | 28.21 | 10% | 29.66 | 28% | 54.44 | 0.4‰ | 56.05 | 0.1‰ |

2