

# **Bioinspired Surface Treatments for Improved Decontamination: Thermally Grafted Oils**

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## **EXECUTIVE SUMMARY**

The Center for Bio/Molecular Science and Engineering at the Naval Research Laboratory (NRL) initiated a program in January 2015 for evaluation of bioinspired treatments suitable for use as a top coat on painted surfaces with the intention of achieving improved aqueous decontamination of these materials. Funding was provided by the Defense Threat Reduction Agency (DTRA, CB10125). This report details results for evaluation of surface treatments prepared by thermally grafting oils on paint surfaces. The materials were deposited on polyurethane paint coated aluminum coupons. Retention of the simulants paraoxon, methyl salicylate, dimethyl methylphosphonate, diisopropyl fluorophosphate, and 2-chloroethyl ethyl sulfide following treatment of contaminated surfaces with a soapy water solution is reported along with droplet diffusion on the surfaces and wetting angles.

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# BIOINSPIRED SURFACE TREATMENTS FOR IMPROVED DECONTAMINATION: THERMALLY GRAFTED OILS

## INTRODUCTION

The DoD Chemical and Biological Defense Program (CBDP) seeks to provide protection of forces in a contaminated environment including contamination avoidance, individual protection, collective protection, and decontamination. In January 2015, the Center for Bio/Molecular Science and Engineering at the Naval Research Laboratory (NRL) began an effort funded through the Defense Threat Reduction Agency (DTRA, CB10125) with a view toward evaluation and development of top-coat type treatments suitable for application to painted surfaces that would reduce retention of chemical threat agents following standard decontamination approaches. The effort sought to survey relevant and related areas of research and evaluate identified technologies under appropriate methods to determine efficacy, scalability, and durability.

Slippery omniphobic covalently attached liquid (SOCAL) treatments offer liquid-like characteristics but are based on covalently attached flexible groups, generally on a smooth surface. They are not dissolved or displaced by contacting liquids. Many SOCAL-like treatments involve complex deposition methods or lead to nondurable coatings. Three previous reports covered testing applications of SOCAL based coatings for reduction of target retention.[1-3] Direct thermal grafting of oils to surfaces has been an area of ongoing research and can depending on the attachment surface density and molecular weight of the oil be classified as a SOCAL treatment. Recently interest has developed in using thermal grafting techniques to form a complete liquid-like coating for anti-scaling and omniphobic surface applications.[4] The methods available in the open literature were modified to synthesize coatings at NRL.

For the complete system, aluminum coupons painted with a polyurethane paint system were treated with TEOS and subjected to thermal grafting of one silicone and three PDMS oils (Figure 1). The coupons were subjected to standard evaluations including measurement of sessile, sliding, and shedding contact angles and quantification of retention for the simulant compounds.

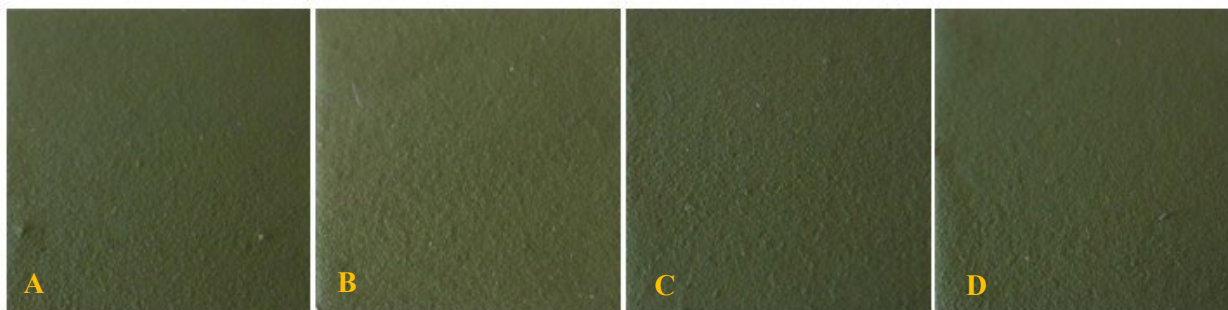


Fig. 1 — Images of painted coupons with (A) Silicone Oil; (B) PDMS 50cST; (C) PDMS 200cST; (D) PDMS 500cSTt.

## METHODS

Sessile contact angles for samples evaluated under this effort used three 3  $\mu$ L droplets per surface with each droplet measured independently three times for each of three targets, water, ethylene glycol, and *n*-heptane. Geometric surface energy was calculated based on the water and ethylene glycol interactions using software designed for the DROPimage goniometer package. Sliding angles were determined using 5  $\mu$ L droplets. The droplet was applied at 0° after which the supporting platform angle was gradually increased up to 60°. Sliding angles for each of the liquids were identified as the angle for which movement

of the droplet was identified. Shedding angles for each liquid were determined using 12  $\mu\text{L}$  droplets initiated 2.5 cm above the coupon surface. Changes in base angle of  $10^\circ$  were utilized to identify the range of droplet shedding angle based on a complete lack of droplet retention by the surface (not sliding). The angle was then reduced in steps of  $1^\circ$  to identify the minimum required angle. Droplet diameters were determined using tools provided by Adobe Photoshop CS3. Droplets of 5  $\mu\text{L}$  were applied to the surfaces and images were collected at 30 s intervals for 5 min followed by images at 5 min intervals for a total of 30 min. DFP samples were kept covered for the duration of the experiment to minimize evaporation. In some cases, reflections from the glass cover can be seen in the images.

Simulant exposure and evaluation methods were based on the tests developed by Edgewood Chemical Biological Center referred to as Chemical Agent Resistance Method (CARM) [5]. Standard target exposures utilized a challenge level of  $10 \text{ g/m}^2$ . The painted coupons were  $0.00101 \text{ m}^2$ ; the  $10 \text{ g/m}^2$  target challenge was applied to the surfaces as two equally sized neat droplets. Following application of the target, coupons were aged 1 h prior to use of a gentle stream of air to expel target from the surface. Samples were then rinsed with soapy water (0.59 g/L Alconox in deionized water). The rinsed coupons were soaked in isopropanol for 30 min to extract remaining target; this isopropanol extract was analyzed by the appropriate chromatography method to determine target retention on the surface.

For analysis of paraoxon, methyl salicylate (MES), diisopropyl fluorophosphate (DFP), dimethyl methylphosphonate (DMMP), and 2-chloroethyl ethyl sulfide (CEES), gas chromatography-mass spectrometry (GC-MS) was accomplished using a Shimadzu GCMS-QP2010 with AOC-20 auto-injector equipped with a Restex Rtx-5 (30 m x 0.25 mm ID x 0.25  $\mu\text{m}$  df) cross bond 5% diphenyl 95% dimethyl polysiloxane column. A GC injection temperature of  $200^\circ\text{C}$  was used with a 1:1 split ratio at a flow rate of 3.6 mL/min at 69.4 kPa. The oven gradient ramped from  $50^\circ\text{C}$  (1 min hold time) to  $180^\circ\text{C}$  at  $15^\circ\text{C}/\text{min}$  and then to  $300^\circ\text{C}$  at  $20^\circ\text{C}/\text{min}$  where it was held for 5 min.

## Coating Synthesis.

### TEOS modification of painted coupons (TEOS/paint)

A sol was prepared from 184 mL 2-propanol, 6 mL tetraethyl orthosilicate (TEOS), and 10 mL  $\text{NH}_4\text{OH}$  (~30% in  $\text{H}_2\text{O}$  reagent) at RT. It was mixed briefly and divided into 2 separate 240 mL PFA jars. 4 painted Al coupons were immersed in each jar, leaning vertically against inside walls, for 10 min at RT. Soaked coupons were then heated in an oven at  $65^\circ\text{C}$  for 30 min. The procedure was repeated twice, for a total of 3 cycles of immersion and heating, with coupons left in the oven at  $65^\circ\text{C}$  over-night for the final curing step. Note: TEOS sol became cloudy the 3rd immersion step; no precipitate or visible discoloration was observed on painted surfaces.

### Thermal grafting of oils on TEOS/paint

The following Dimethylpolysiloxane (PDMS) oils were used in the following grafting procedure; viscosity 50 cSt ( $25^\circ\text{C}$ ) from Sigma-Aldrich (cat. # DMPS5X), MW listed as ~3,780; viscosity 200 cSt ( $25^\circ\text{C}$ ) from Sigma-Aldrich (cat. # DMPS2C), MW listed as ~9,430; and viscosity 500 cSt ( $25^\circ\text{C}$ ) from Sigma-Aldrich (cat. # DMPS5C), MW listed as ~17,250. Also used was the Silicone Oil AR 20, viscosity ~20 mPa.s ( $25^\circ\text{C}$ ) from Sigma-Aldrich (cat. # 10836), MW not found.

TEOS/paint substrates were briefly rinsed with 2-propanol and blown dry with  $\text{N}_2$ . Coupons were immersed in oil (PDMS of viscosity 50 cSt, 200 cSt, or 500 cSt; or silicone oil AR 20) contained in glass culture dishes. Containers were covered with watch glasses and heated in an oven at  $90^\circ\text{C}$  for 1 day. Oil coated substrates were thoroughly rinsed in toluene and 2-propanol. They were initially blown dry with  $\text{N}_2$

and then dried further in an oven at 60 °C. Oil grafted substrates were qualitatively more hydrophobic than “bare” TEOS/paint surfaces.

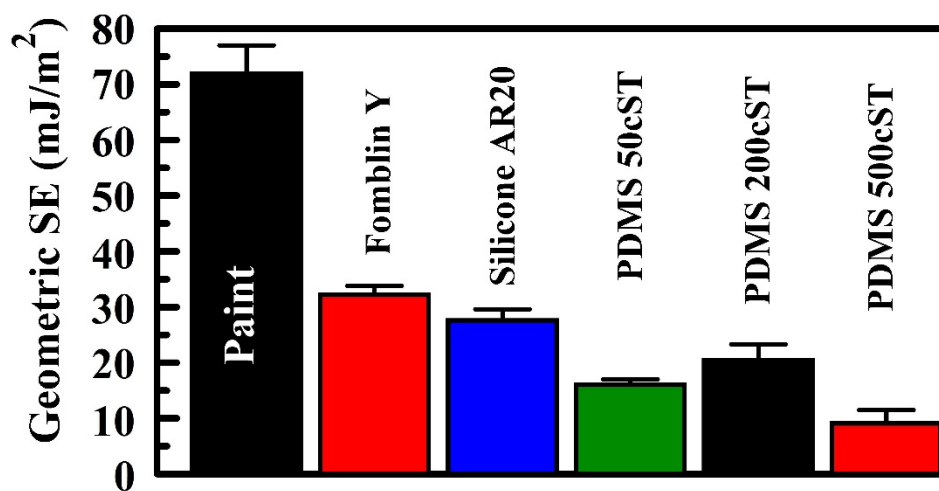


Fig. 2 — Geometric surface energy ( $\text{mJ}/\text{m}^2$ ) for the evaluated coatings. Paint and Fomblin Y results provided for comparison.

## RESULTS

Analysis of the support surface in the absence of additional coatings provides a point of comparison for evaluating the benefits of the surface treatments. Each table includes data on the relevant support material, a painted aluminum coupon and for a Fomblin Y lubricated painted aluminum coupon. Results for four oils thermally grafted directly on painted coupons were considered. Application of coatings considered here reduced the surface energy of the painted surface (Table 1 and Figure 2) to values similar to what is observed when Fomblin Y is used to coat a coupon.

Table 1 – Sessile, Sliding, and Shedding Contact Angles on Aluminum Supports

Coupon	Liquid	Sessile Angle	Sliding Angle	Shedding Angle	Geometric Surface Energy ( $\text{mJ}/\text{m}^2$ )
Paint Only	water	$47.5 \pm 1.1$	>60	>60	$71.9 \pm 5.1$
	ethylene glycol	$55.7 \pm 2.1$	>60	>60	
	n-heptane	--	--	--	
Fomblin Y Oiled Paint	water	$73.1 \pm 2.1$	>60	$46.7 \pm 3.3$	$32.2 \pm 1.6$
	ethylene glycol	$52.5 \pm 0.61$	>60	$49.8 \pm 4.9$	
	n-heptane	$40.1 \pm 2.9$	>60	$36.6 \pm 3.3$	
Silicone AR20 Oil	water	$108.2 \pm 1.1$	>60	>60	$27.6 \pm 2.0$
	ethylene glycol	$79.0 \pm 0.66$	>60	>60	
	n-heptane	--	--	--	
PDMS Oil vis 50cST	water	$107.7 \pm 0.78$	>60	>60	$16.0 \pm 1.0$
	ethylene glycol	$87.0 \pm 0.86$	>60	>60	
	n-heptane	--	--	--	
PDMS Oil vis 200cST	water	$113.8 \pm 1.2$	>60	>60	$20.4 \pm 2.9$
	ethylene glycol	$88.3 \pm 1.6$	>60	>60	
	n-heptane	--	--	--	
PDMS Oil vis 500cST	water	$117.0 \pm 1.2$	>60	>60	$9.1 \pm 2.4$
	ethylene glycol	$101.0 \pm 3.0$	>60	>60	
	n-heptane	--	--	--	

All the thermally grafted oils produced surface energies equivalent to or lower than that observed on the Fomblin Y oiled coupon. The third PDMS oil with the highest molecular weight had the lowest surface energy of any coating in the comparison. All of the surfaces were fully wetted by heptane. No sliding on the surfaces was noted below an incline of 60°. No shedding behavior was noted for these surfaces. None of the coatings induced observable changes in the appearance of the painted coupons.

The tendency of droplets to spread across the surfaces was also evaluated (Figure 3; Appendices). For these studies, droplets of the simulants (5  $\mu\text{L}$ ) were utilized. The spread of the droplets was quantified by measuring the diameter of the droplets in the images over time (Figure 4). For the paint only samples, MES and DFP spread quickly, reaching the edges of the coupon at 10 and 2 min, respectively. DMMP does not spread during the course of the 30 min incubation. The silicone oil show similar behavior with rapid droplet spreading for MES and DFP and none for DMMP. In contrast, the PDMS oils have significantly less spreading of DFP and MES with the amount of spread decreasing as the viscosity and molecular weight of the grafted oil increased.

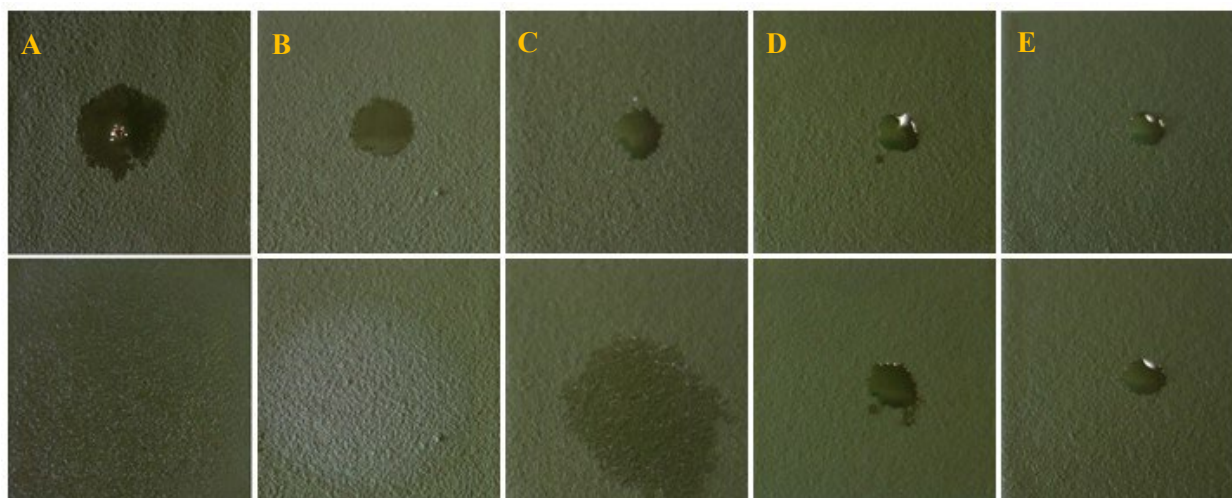


Fig. 3 — Images from initial series of coupons at 0 and 30 min following MES exposure: (A) Paint only; (B) Silicone Oil; (C) PDMS 50cST; (D) PDMS 200cST; and (E) PDMS 500cST.

The coupons were subjected to simulant exposure (10  $\text{g}/\text{m}^2$ ), aging, washing, and drying. These materials showed little change in the appearance or wetting characteristics over these processing steps. When the soapy water process was employed (Figure 5; Table 2), retention of all targets was less for the Fomblin Y lubricated paint than for the paint only surface. Here, the silicone oil coatings tested for agents, had retention reductions greater than Fomblin Y coated coupons except for MES. The retention for all the PDMS oil coatings was significantly lower compared to both paint only and Fomblin Y coated coupons. It also appeared for these samples that retention reduced as the viscosity and molecular weight of oil grafted increased. While these trends are present for this particular polymer using the same coating methodology, different methods of incorporating PDMS into coatings can result in different dependences. For instance, a coating from an earlier study listed in Table 2 that used a PDMS chain of MW  $\sim 2,500$  had performance better than the 50cST sample which used chains with a MW  $\sim 3,780$  demonstrating how alterations to coating assembly can impact performance.[6] Its performance was not as good as the 200cST or 500cST oils suggesting that a longer PDMS chain used for that coating method may improve retention performance.

For comparison, paint only coupons retained significant amounts of target at 5.48, 6.20, 4.28, and 0.52  $\text{g}/\text{m}^2$ . When no rinsing or decontamination steps were used, paint only coupons retained the following:

paraoxon – 9.84 g/m<sup>2</sup>, MES – 9.54 g/m<sup>2</sup>, DMMP – 9.90 g/m<sup>2</sup>, DFP - 7.39 g/m<sup>2</sup>. Though the nominal target application was 10 g/m<sup>2</sup>, recovery from surfaces was always less than this value. Losses due to evaporation would be expected, especially for DFP. Additional losses likely occur during rinse steps due to agent interaction with the untreated region of the coupon; the back of these coupons is unpainted aluminum.

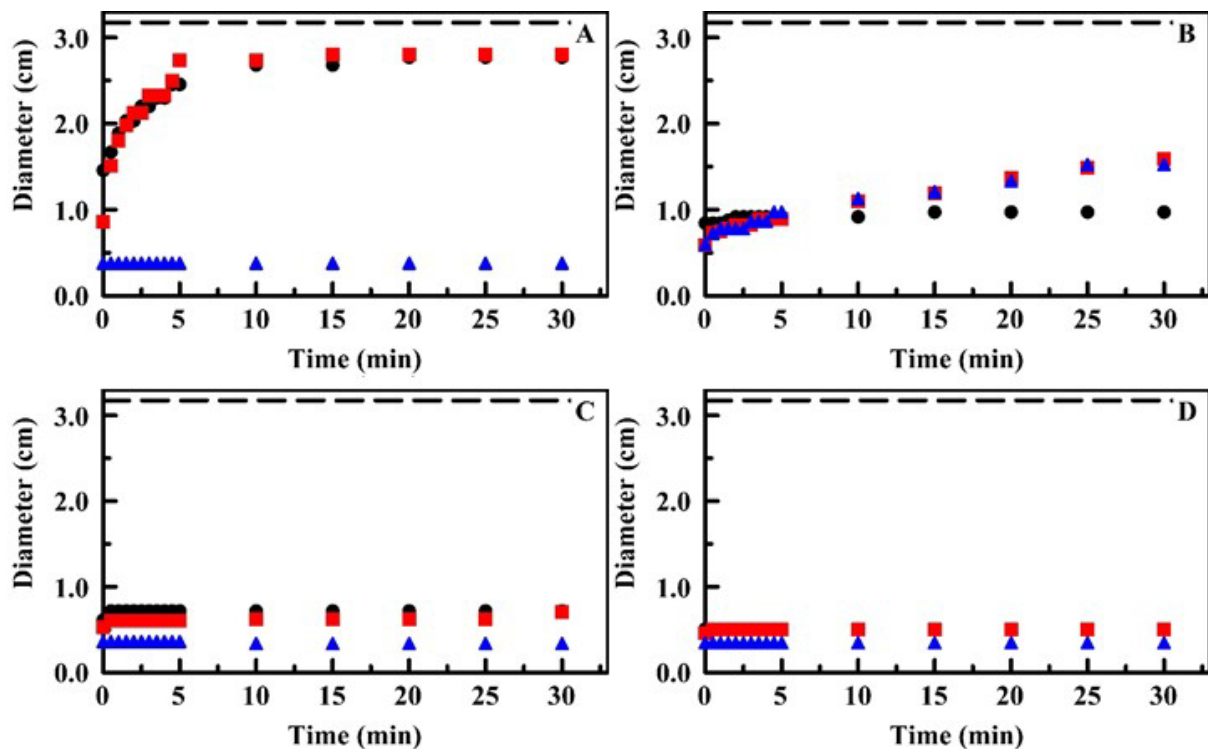


Fig. 4 — Droplet diameters over time following exposure to DFP (black), MES (red), and DMMP (blue) for painted coupons with (A) Silicone oil; (B) PDMS 50cST; (C) PDMS 200cST; (D) PDMS 500cST.

Table 2 – Target Retention (g/m<sup>2</sup>) Following 1 h Aging on Aluminum Supports

Coupon	Paraoxon	MES	DMMP	DFP	CEES
Paint Only	5.48	6.20	4.28	0.52	1.31
Fomblin Y Oiled Paint	1.24	2.85	0.59	0.34	1.36
Silicone AR20 Oil	1.78	4.90	.05	0.54	1.57
PDMS Oil vis 50cST	0.34	2.14	ND	0.56	0.97
PDMS Oil vis 200cST	0.05	0.31	ND	0.04	0.63
PDMS Oil vis 500cST	0.14	0.24	ND	0.02	0.99
PDMS[6]	ND	0.34	ND	0.10	

ND = not detected

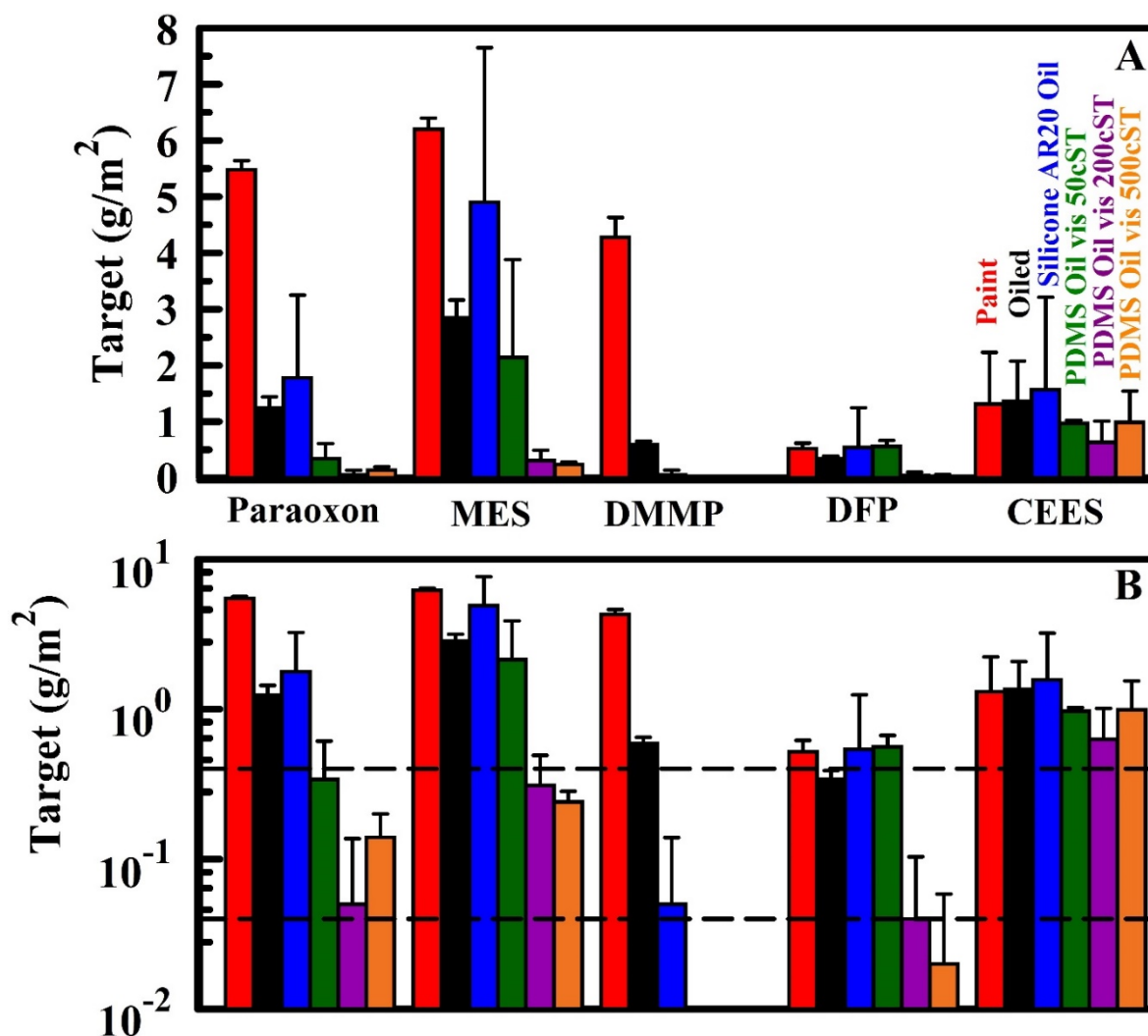


Fig. 5 — Target retention by coupons from initial series following treatment with an air stream and rinsing with soapy water shown to left on a linear scale (A) and (B) on a log scale: (left to right) painted (red), Fomblin Y (black), Silicone AR20 (blue), PDMS 50cST (green), PDMS 200cST (purple), PDMS 500cST (orange).

## CONCLUSIONS

The coatings generated from thermally grafting oils onto TEOS coated surfaces had varying levels of effectiveness. The materials had much lower surface energies than the original painted surface. Compared to Fomblin Y oiled coupons, the values were equivalent or slightly lower. The droplet spreading behavior varied across the oils tested with Silicone AR20 not producing much difference from what is seen for the paint only coupon while the PDMS oils altered the spread of MES and DFP with higher viscosity oils having a greater reduction in droplet spread. For the silicone oil, large reductions were seen for all chemicals except MES which only had a small reduction compared to the paint only coupon. Overall retention of all targets were reduced for the PDMS oils. These results while an improvement over what is observed for the paint only coupon do not approach the values observed for other coatings tested. Thermally grafted oils serve as a useful benchmark and comparison for other coatings that may contain similar building blocks assembled in significantly different ways. The lack of reduction for retention of agents indicates no further testing of these materials is warranted.

## ACKNOWLEDGEMENTS

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## REFERENCES

1. White, B.; Melde, B.; Malanoski, A.; Moore, M. *Bioinspired Surface Treatments for Improved Decontamination: Slippery Omniphobic Covalently Attached Liquid (SOCAL)*; NRL/MR/6930-17,9761; US Naval Research Laboratory, Washington, DC, 2017.
2. White, B.; Malanoski, A.; Moore, M.; Melde, B. *Bioinspired Surface Treatments for Improved Decontamination: Variations on Slippery Omniphobic Covalently Attached Liquid (SOCAL) Coatings*; NRL/MR/6930-19,9847; US Naval Research Laboratory, Washington, DC, 2019.
3. Malanoski, A.; White, B.; Moore, M.; Melde, B. *Bioinspired Surface Treatments for Improved Decontamination: Adaptive Surface Technologies III*; NRL/6306/MR/2022/3; US Naval Research Laboratory, Washington, DC, 2022.
4. Krumpfer, J.W.; McCarthy, T.J. Rediscovering silicones: "unreactive" silicones react with inorganic surfaces. *Langmuir* **2011**, *27*, 11514-11519, doi:10.1021/la202583w.
5. Lalain, T.; Mantooth, B.; Shue, M.; Pusey, S.; Wylie, D. *Chemical Contaminant and Decontaminant Test Methodology Source Document*; ECBC-TR-980; US Army RDEC, Edgewood Chemical Biological Center: US Army RDEC, Edgewood Chemical Biological Center, Aberdeen Proving Ground, MD, 2012.
6. Malanoski, A.; White, B.; Melde, B.; Moore, M. *Bioinspired Surface Treatments for Improved Decontamination: Polyurethane-PDMS Topcoat*; NRL/6306/MR/2022/?; US Naval Research Laboratory, Washington, DC, 2022.

## **Appendix A**

### **Silicone AR20 oil COUPON IMAGES**

Fig. A1 — DFP on Silicone AR20. Images of a film supported by painted coupon before application (A) and at 0 (B), 0.5 (C), 1.0 (D), 1.5 (E), 2.0 (F), 2.5 (G), 3.0 (H), 3.5 (I), 4.0 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target. These images were collected with a glass cover in place to limit evaporation. Reflections from the cover can be seen in some images.

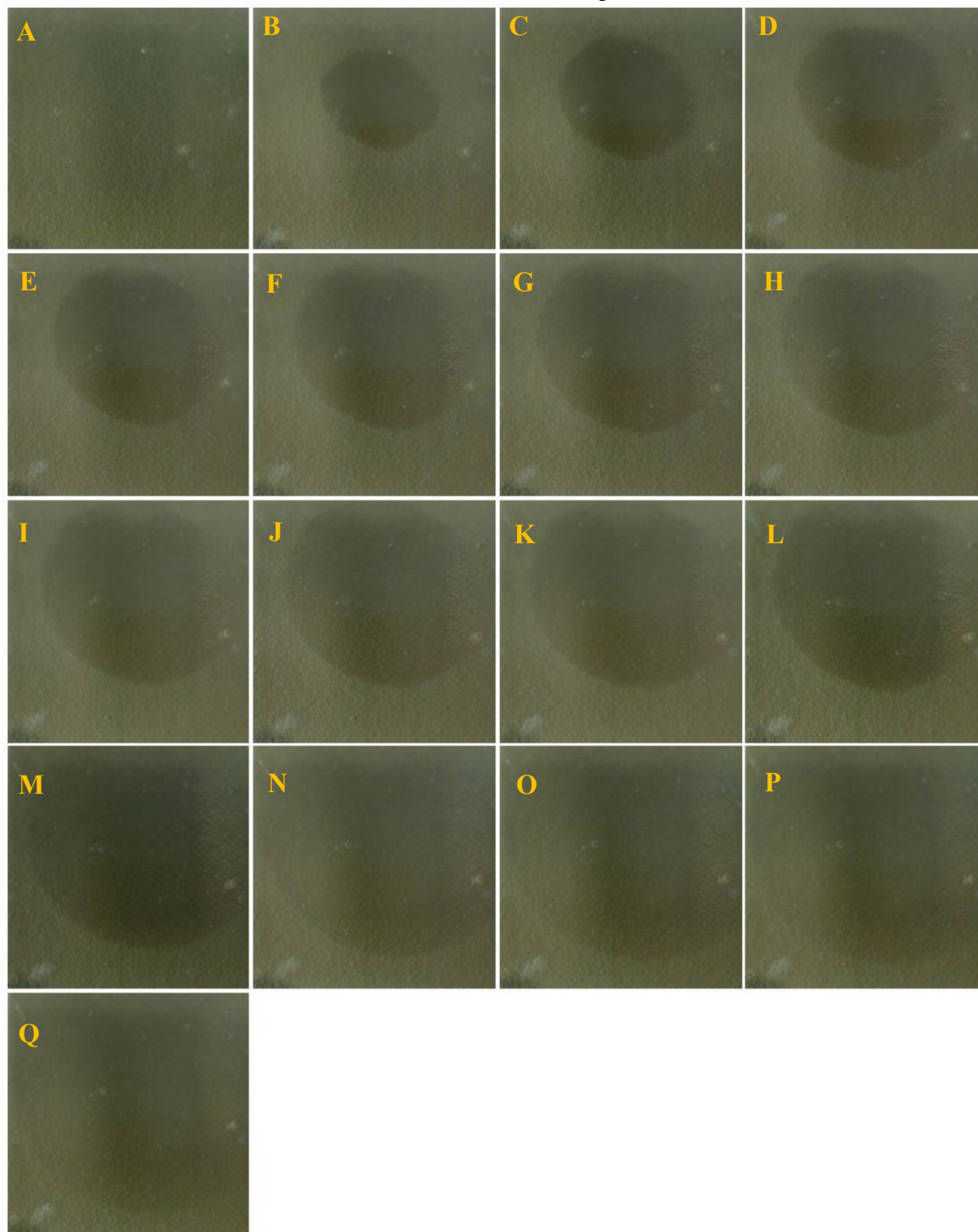


Fig. A2 — MES on Silicone AR20. Images of a film supported by painted coupon before application (A) and at 0 (B), 0.5 (C), 1 (D), 1.5 (E), 2 (F), 2.5 (G), 3 (H), 3.5 (I), 4 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target.

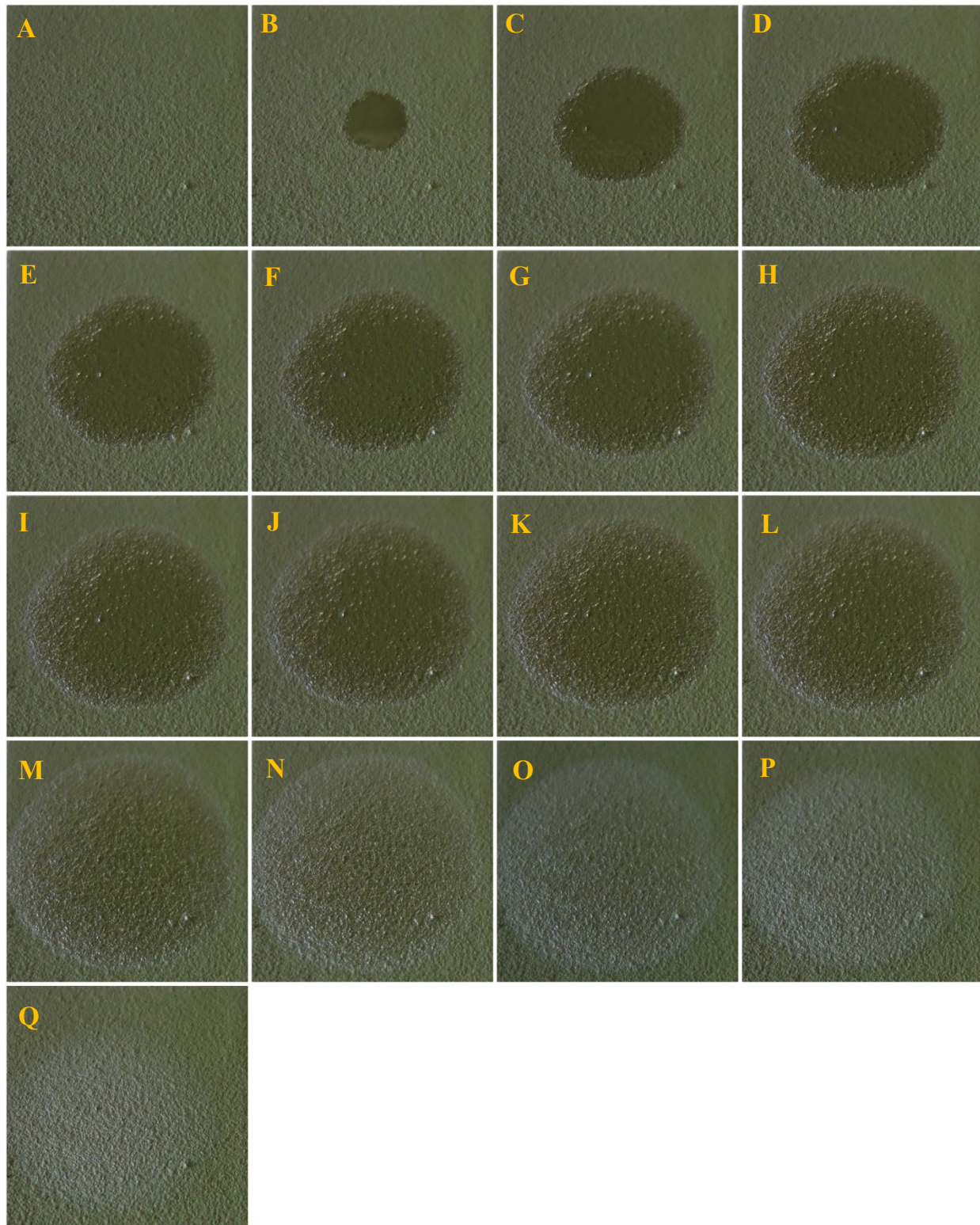
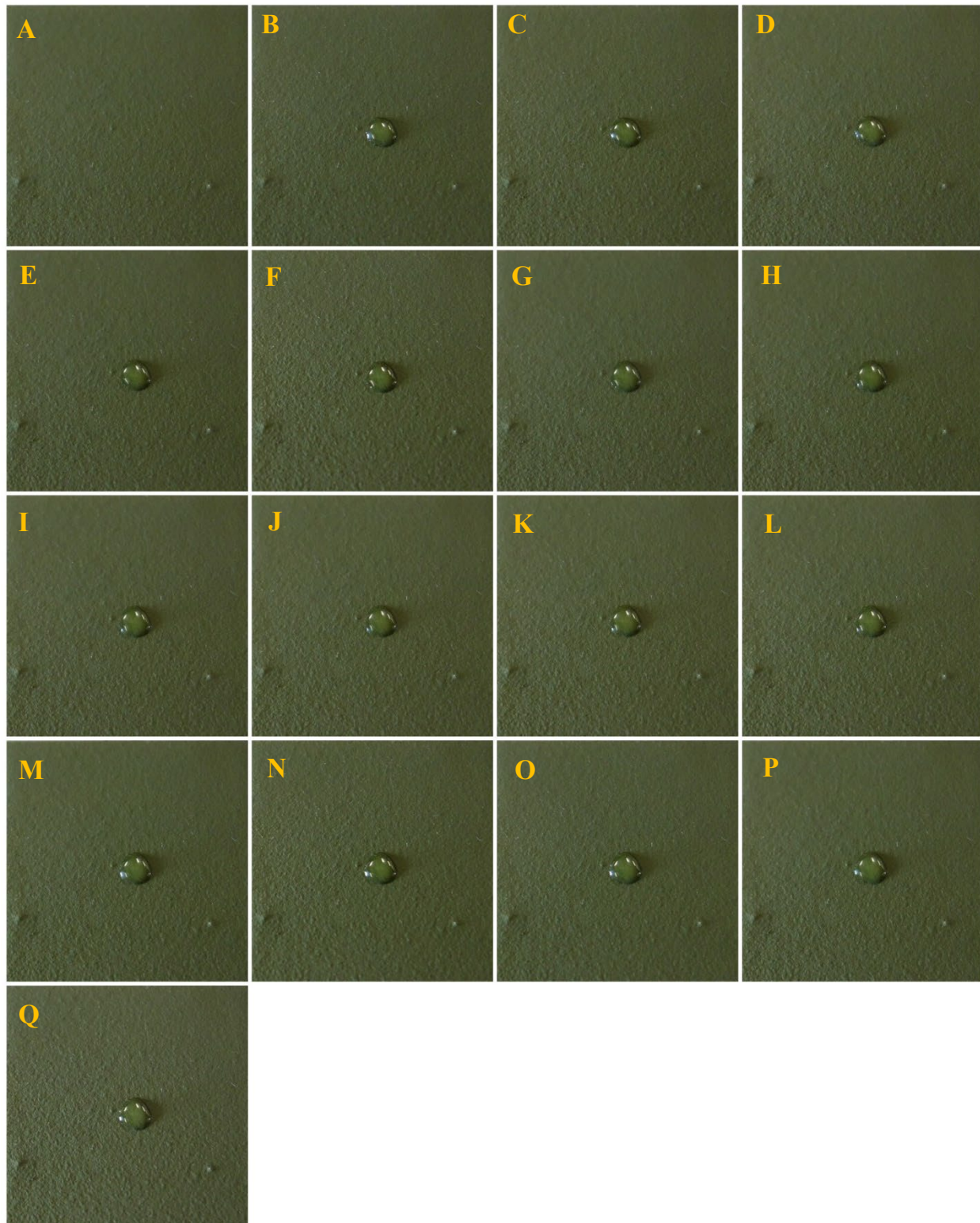


Fig. A3 — DMMP on Silicone AR20. Images of a film supported by painted coupon before application (A) and at 0 (B), 0.5 (C), 1 (D), 1.5 (E), 2 (F), 2.5 (G), 3 (H), 3.5 (I), 4 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target.



**Appendix B****PDMS oil 50cST COUPON IMAGES**

Fig. B1 — DFP on PDMS 50cST. Images of a film supported by painted coupon before application (A) and at 0 (B), 0.5 (C), 1.0 (D), 1.5 (E), 2.0 (F), 2.5 (G), 3.0 (H), 3.5 (I), 4.0 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target. These images were collected with a glass cover in place to limit evaporation. Reflections from the cover can be seen in some images.

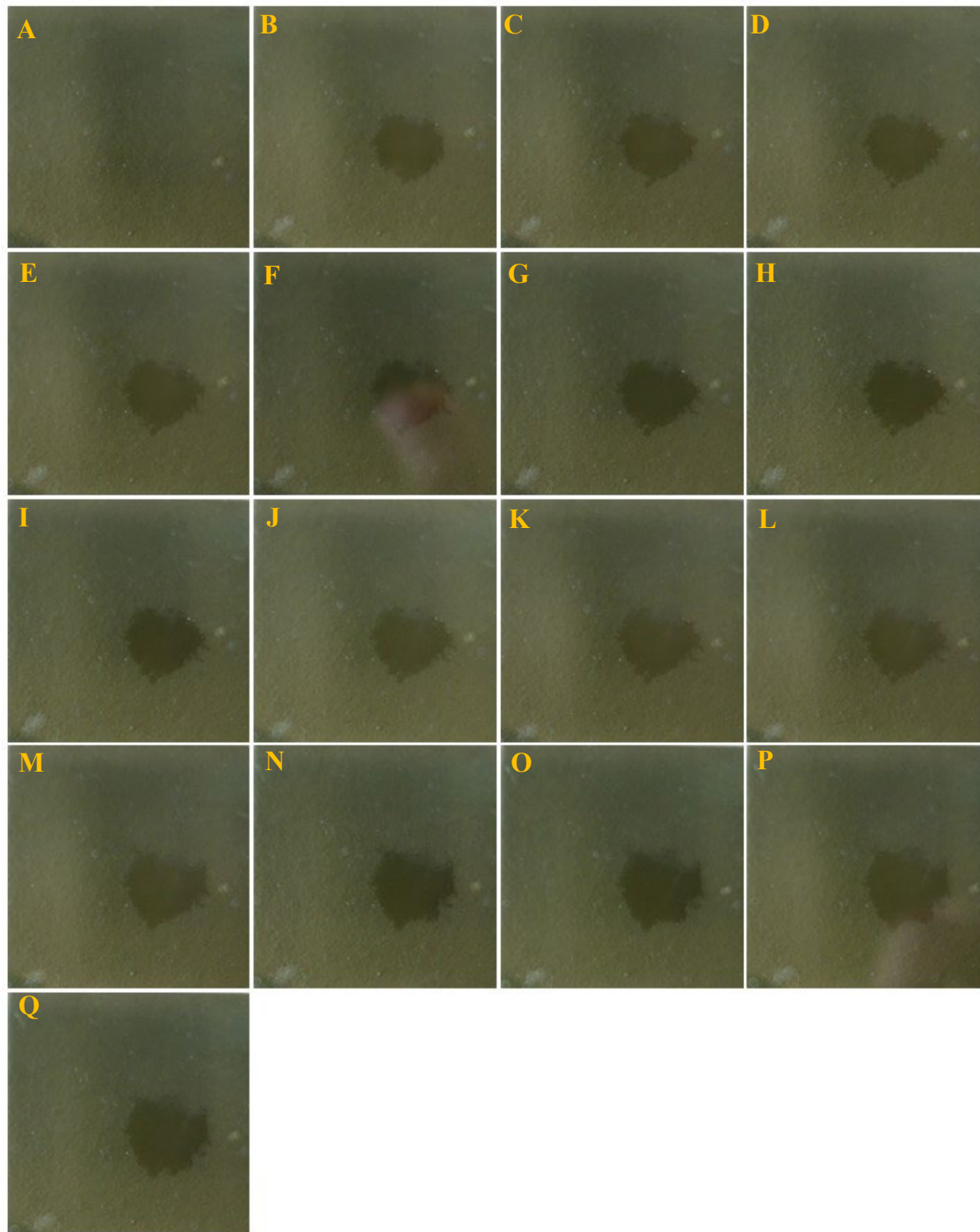


Fig. B2 — MES on PDMS 50cST. Images of a film supported by painted coupon before application (A) and at 0 (B), 0.5 (C), 1 (D), 1.5 (E), 2 (F), 2.5 (G), 3 (H), 3.5 (I), 4 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target.

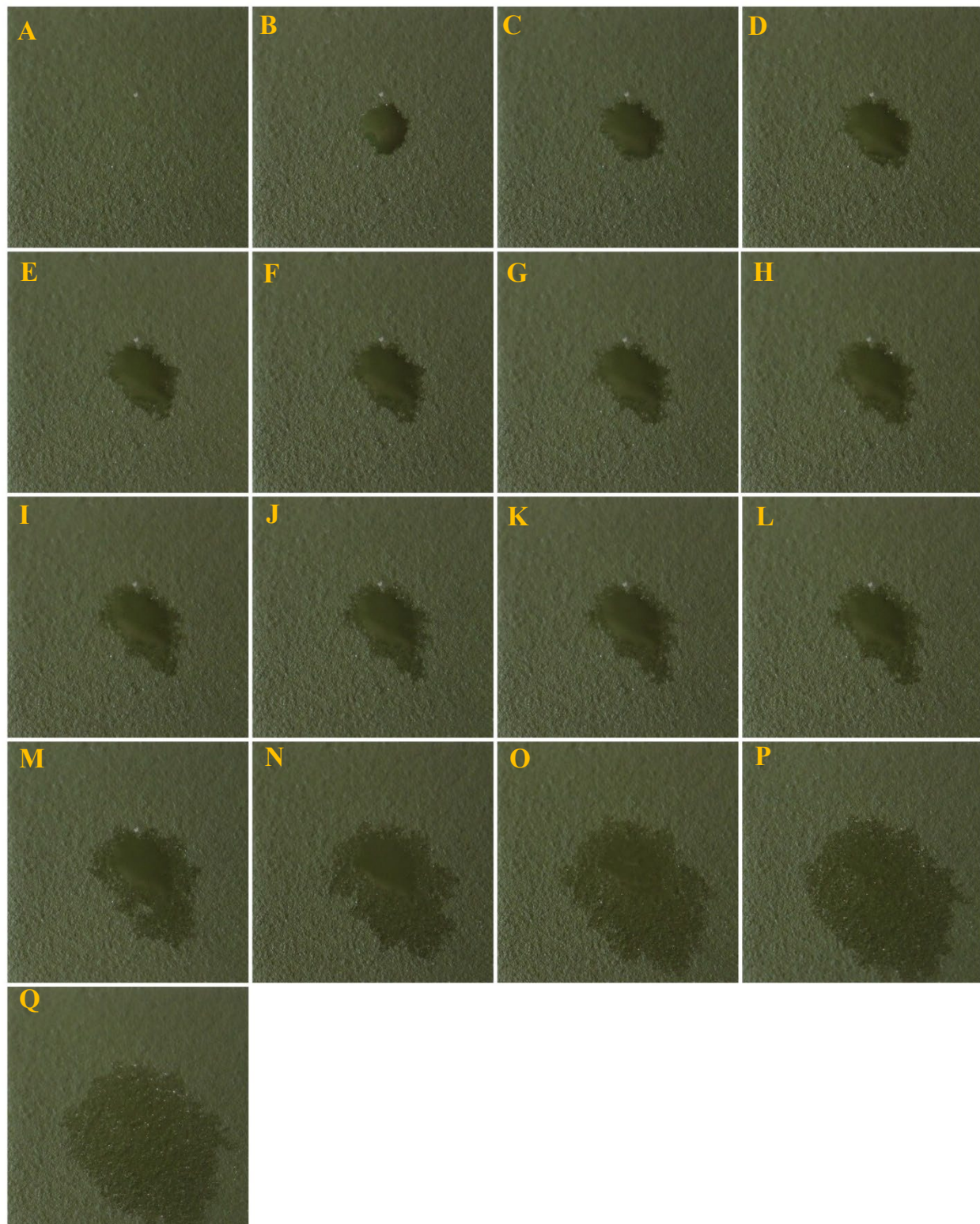
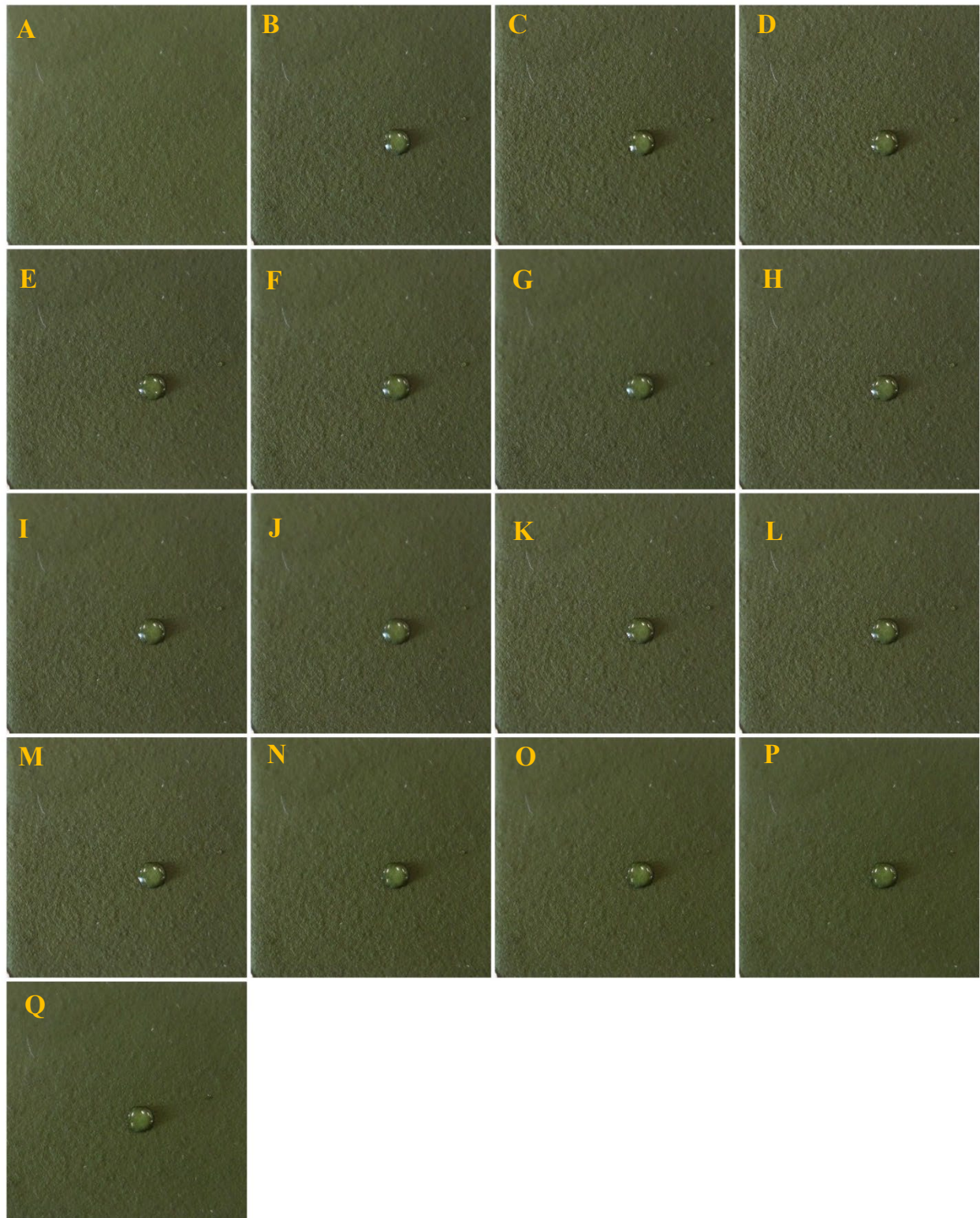


Fig. B3 — DMMP on PDMS 50cST. Images of a film supported by painted coupon before application (A) and at 0 (B), 0.5 (C), 1 (D), 1.5 (E), 2 (F), 2.5 (G), 3 (H), 3.5 (I), 4 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target.



**Appendix C****PDMS 200cST COUPON IMAGES**

Fig. C1 — DFP on PDMS 200cST. Images of a film supported by painted coupon before application (A) and at 0 (B), 0.5 (C), 1.0 (D), 1.5 (E), 2.0 (F), 2.5 (G), 3.0 (H), 3.5 (I), 4.0 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target. These images were collected with a glass cover in place to limit evaporation. Reflections from the cover can be seen in some images.

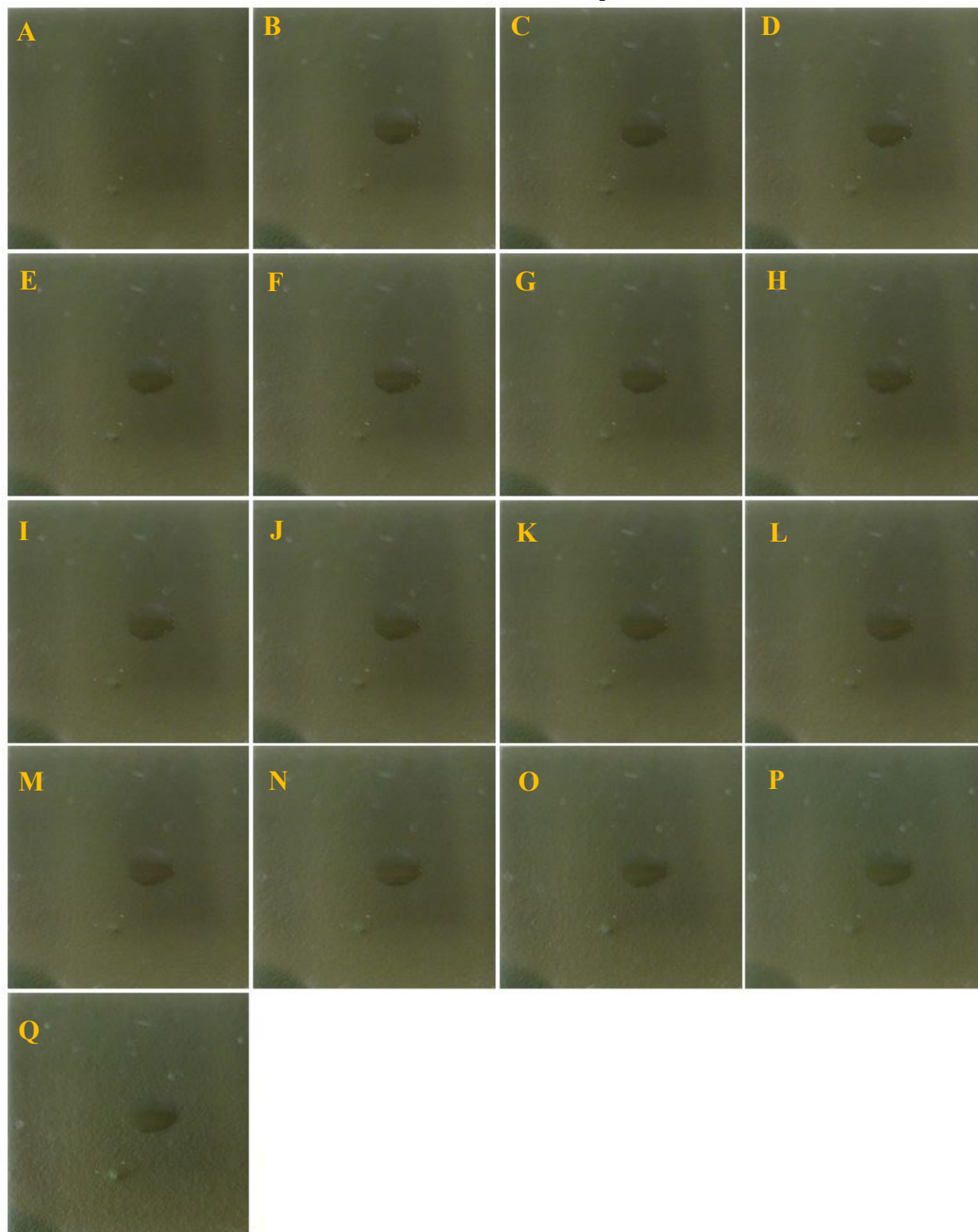


Fig. C2 — MES on PDMS 200cST. Images of a film supported by painted coupon before application (A) and at 0 (B), 0.5 (C), 1 (D), 1.5 (E), 2 (F), 2.5 (G), 3 (H), 3.5 (I), 4 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target.

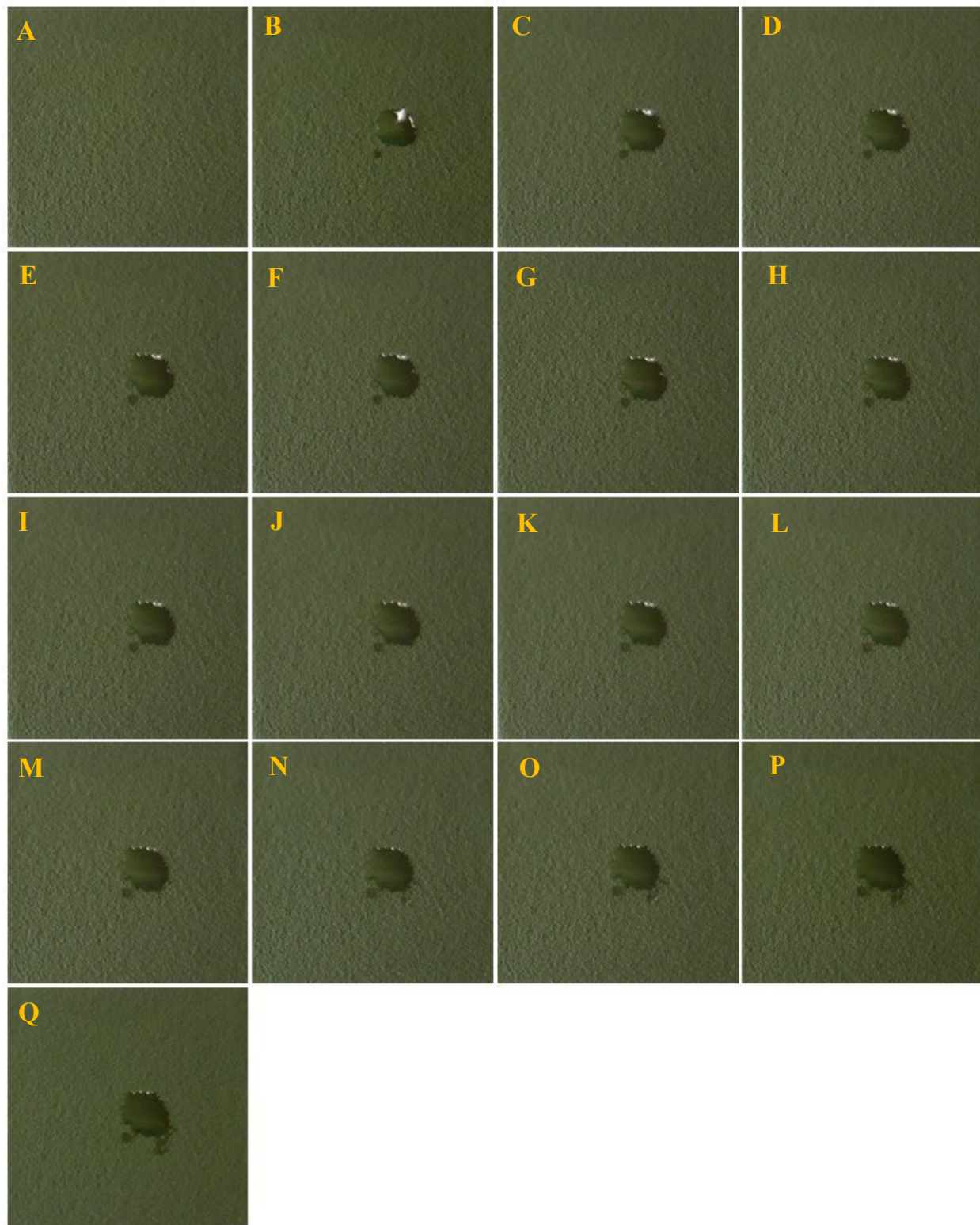
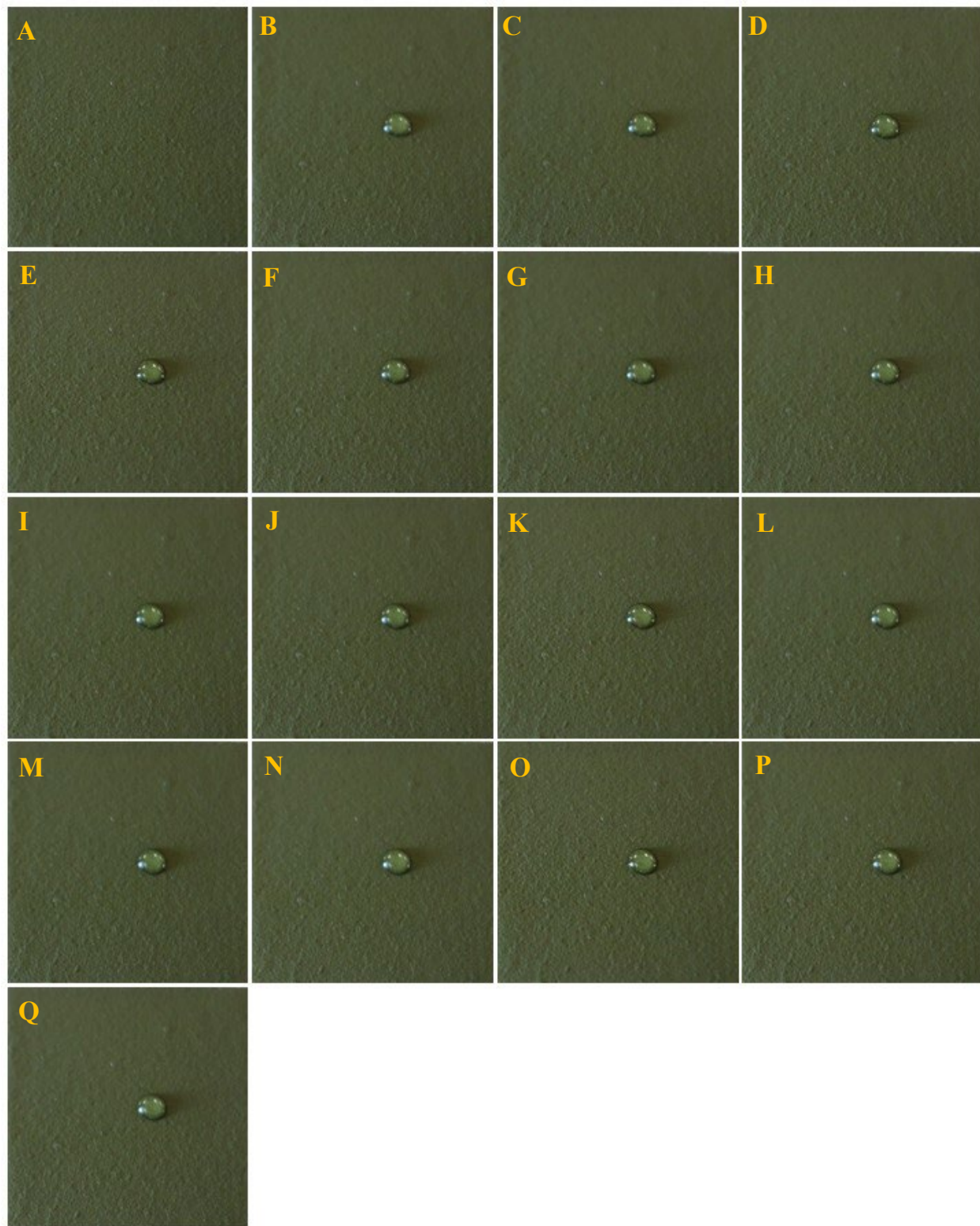


Fig. C3 — DMMP on PDMS 200cST. Images of a film supported by painted coupon before application (A) and at 0 (B), 0.5 (C), 1 (D), 1.5 (E), 2 (F), 2.5 (G), 3 (H), 3.5 (I), 4 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target.



**Appendix D****PDMS 500cST COUPON IMAGES**

Fig. D1 — DFP on PDMS 500cST. Images of a coupon before application (A) and at 0 (B), 0.5 (C), 1.0 (D), 1.5 (E), 2.0 (F), 2.5 (G), 3.0 (H), 3.5 (I), 4.0 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target. These images were collected with a glass cover in place to limit evaporation. Reflections from the cover can be seen in some images.

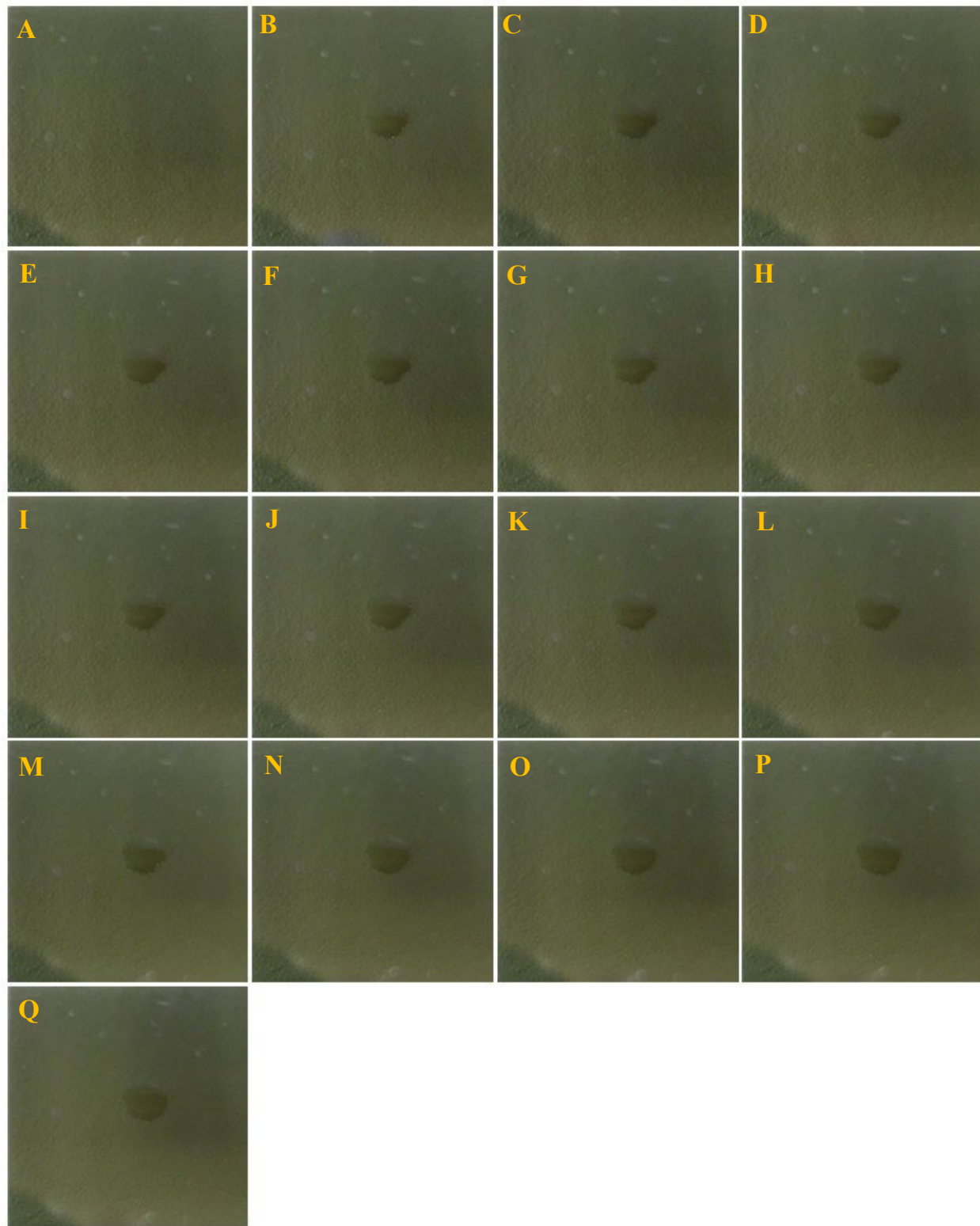


Fig. D2 — MES on PDMS 500cST. Images of a coupon before application (A) and at 0 (B), 0.5 (C), 1 (D), 1.5 (E), 2 (F), 2.5 (G), 3 (H), 3.5 (I), 4 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target.

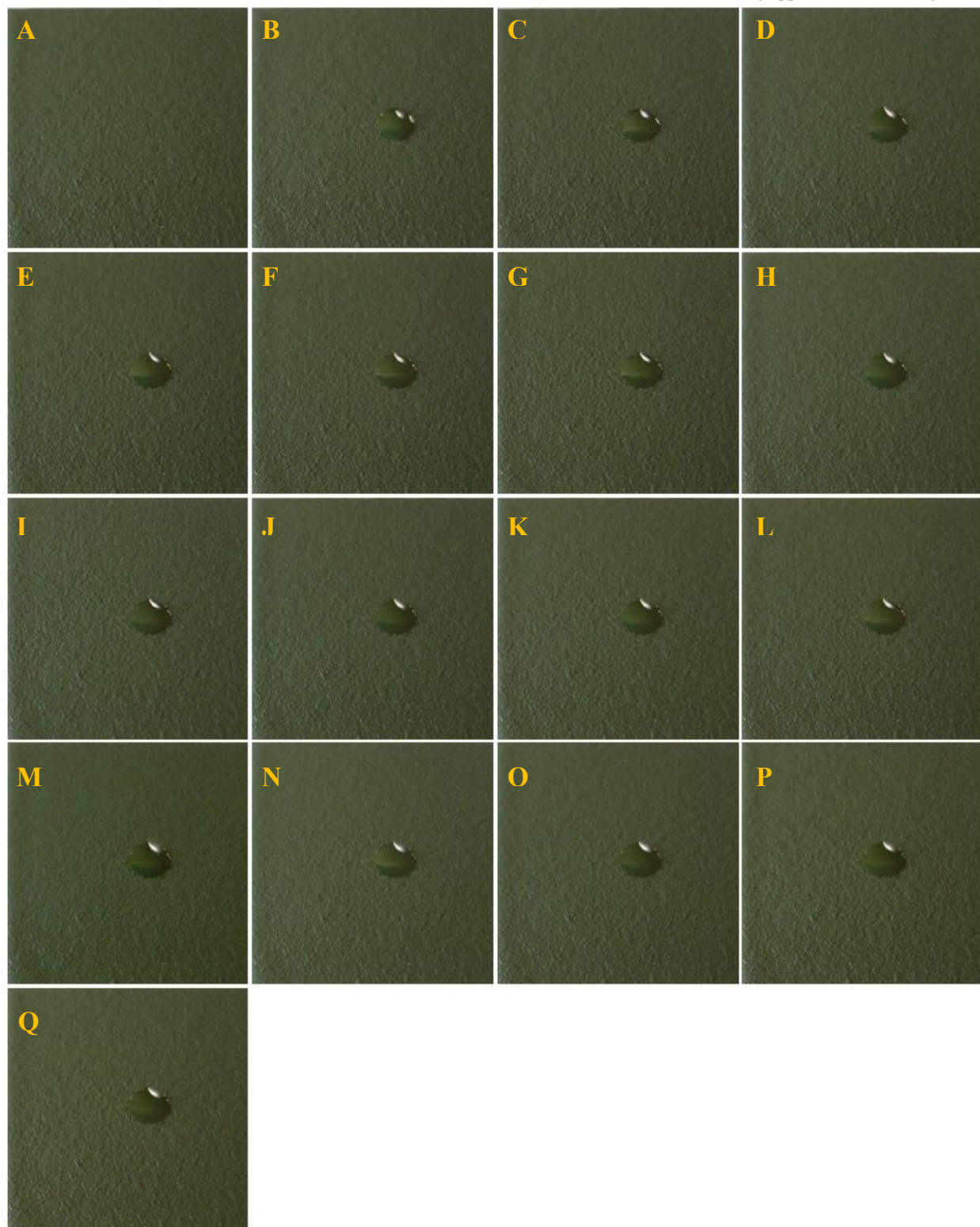


Fig. D3 — DMMP on PDMS 500cST. Images of a coupon before application (A) and at 0 (B), 0.5 (C), 1 (D), 1.5 (E), 2 (F), 2.5 (G), 3 (H), 3.5 (I), 4 (J), 4.5 (K), 5 (L), 10 (M), 15 (N), 20 (O), 25 (P), and 30 (Q) min following application of the target.

