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### **Progress in Organic Coatings**

## Bubble entrapment and escape from sprayed paint films Alireza Dalili<sup>a,\*</sup>, Sanjeev Chandra<sup>a</sup>, Javad Mostaghimi<sup>a</sup>, H.T. Charles Fan<sup>b</sup>,

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#### ABSTRACT

A model paint consisting of resin dissolved in butanol was sprayed onto horizontal glass substrates to form films varying from 100 µm to 450 µm in thickness. The substrates were photographed at 5 s intervals and image analysis software used to measure the number, diameter and velocity of air bubbles trapped in the paint layer. Painted substrates were weighed to determine the rate of solvent evaporation. Bubbles escaped from the paint in 100-900 s, the time increasing with paint thickness. The Sauter mean diameter of bubbles in films less than 300 µm in thickness decreased with time because larger bubbles escaped faster than small bubbles, while the mean diameter of bubbles in a 450 µm thick layer increased due to bubble coalescence. Bubble velocities due to movement of the liquid increased with paint thickness and reached 30 µm/s. Bubbles were observed to escape from both upward and downwards facing surfaces. Concentration gradients due to solvent evaporation in a paint film create surface tension variations that drive Marangoni flows, which bring bubbles to the paint surface. An analytical one-dimensional model of solvent diffusion was used to calculate solvent concentration variations in the paint film and the Marangoni number.

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### 1. Introduction

When liquid is atomized and sprayed on a surface, as in industrial paint processes, a large number of bubbles are formed in the deposited layer [1]. This is a well-known problem in the automotive paint industry and "defoaming agents", which are typically surfactants, are added to paints to minimize bubble entrapment [2]. Bubbles can create serious defects in car body finishes. After paint is sprayed on automotive components they are baked in an oven to evaporate the solvent and cure the paint. Evaporating solvent diffuses into any bubbles still in the paint, expanding them until they burst through the paint surface and create visible pinholes [3].

A number of studies have been carried out to investigate the formation of air bubbles during droplet impact [4–15]. Chandra and Avedisian [4] photographed the impact of a *n*-heptane droplet onto a stainless steel surface at room temperature and observed the presence of a single bubble at the point of impact. Mehdi-Nejad et al. [5] numerically simulated droplet impact and concluded that as a droplet approaches a solid surface, the air in the gap between them is forced out. Increased air pressure under the droplet leads to the creation of a depression in its surface in which air is trapped, creating a bubble. Ultrafast X-ray [6] and high speed video imaging [7–10] has been used to visualize bubble formation. Bubbles were also observed during impact of micron-sized droplets [11]. Bubbles were seen to form due to air entrainment in free-surface cusps [12] and during droplet impact on liquid pools and films [13–15]. The process of bubble entrapment is fairly well understood.

However, it is not clear how bubbles escape from paint films. It is standard practice in the automotive paint industry to wait for 5-10 min after spraying a component, a period known as the "flashoff" time during which some of the solvent evaporates, before placing it in an oven to cure [2]. This delay is known to minimize the number of bubbles and suppress their growth, but the exact mechanism by which bubbles leave the paint has not been established.

This study was undertaken to determine how air bubbles escape from a spray painted film. We photographed the motion of bubbles in liquid films applied by spraying paint onto a glass substrate. A model paint, whose surface tension and viscosity were measured, was used. All experiments were done at room temperature. The size and number of bubbles were measured as a function of time using image analysis software. Substrates were weighed to deter-

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t	lime (s)
a <sub>32</sub>	Sauter mean diameter (m)
g	Gravitational acceleration (m/s <sup>2</sup> )
r	Bubble radius (m)
U	Bubble velocity (m/s)
J	Volatile mass flux (kg/m <sup>2</sup> s)
L	Paint film thickness (m)
$D_{v}$	Volatile diffusivity in paint $(m^2/s)$
x	Vertical coordinate (m)
α	Coefficient in mass transfer equation at paint surface
	$(kg/m^2s)$
$F_{\sigma}$	Surface tension force (N/m)
$F_{\mu}$	Viscous shear force (N/m)
Μ	Mass (kg)
С	Concentration
$C_i$	Initial volatile concentration
$C_{\infty}$	Volatile concentration in atmosphere
$\Delta C$	Concentration difference across the paint film
Bi	Biot number
Fo	Fourier number
Ма	Marangoni number
Greek letters	
0	Model paint density $(kg/m^3)$
ρ 0	Volatile partial density $(kg/m^3)$
$\rho_v$	Bubble density $(kg/m^3)$
$\rho_p$	Difference between model point and hubble density
$\Delta \rho$	$(kg/m^3)$
$\mu$	Dynamic viscosity (N s/m <sup>2</sup> )
σ	Surface tension (N/m)
$\lambda_n$	Eigenvalues
	-

mine the rate of solvent evaporation. An analytical model of solvent diffusion through the paint layer was used to calculate concentration gradients and thereby to estimate the magnitude of surface tension gradients that drive Marangoni flows in the paint.

#### 2. Experimental system

Experiments were conducted using model paint, prepared by mixing 85 wt% resin with 15 wt% solvent. The resin consisted of 70 wt% CYMEL 1159 (Cytec Industries Inc., Woodland Park, NJ, USA) and 30 wt% PARALOID AT400 (Dow Chemical Co., Midland, Michigan, USA). The solvent used was n-butanol (Caledon Laboratory Chemicals, Georgetown, ON, Canada). Using thermogravimetric analysis (TGA) it was determined that the pure resin initially contained 35.7 wt% solvent. The initial solvent concentration in the model paint was therefore 45 wt%. The surface tension and viscosity of the paint were measured experimentally by Javaheri [16] using a digital viscometer (DV-I, Brookfield Engineering Laboratories, Middleboro, MA, USA). The viscosity was measured for the pure resin mixture (Cymel and Paraloid) and then for solutions of the resin in *n*-butanol in 2 wt% increments up to 20 wt% of solvent (Fig. 1a). The viscosity decreased from approximately 3500 cP for the pure resin to 200 cP for a solution with 20 wt% butanol. The surface tension was measured [16] using a force tensiometer (Sigma KSV, Biolin Scientific, Stockholm, Sweden) with a standard platinum-iridium ring in contact with the model paint. Increasing amounts of the solvent were added to the paint similar to what was done when measuring viscosity. The surface tension decreased from 27.5 mN/m for pure resin to 23.5 mN/m for pure *n*-butanol (Fig. 1b). The model paint, consisting of resin with 15% solvent, had density ( $\rho$ ) of 988 kg/m<sup>3</sup>,

volatile partial density ( $\rho_{\nu}$ ) of 442 kg/m<sup>3</sup>, dynamic viscosity ( $\mu$ ) of 240 cP and surface tension ( $\sigma$ ) of 26 mN/m. As solvent concentration decreased, the viscosity and surface tension of the model paint increased. Properties of *n*-butanol were taken from [17].

A handheld compressed air sprayer (Project Sprayer, Wagner Spray Tech, Plymouth, MN, USA) was used to spray model paint onto the substrate. The substrate was a  $2'' \times 2''$  piece of heatresistant borosilicate glass (8476K121, McMaster-Carr, Elmhurst, IL, USA), which was chosen because it was transparent and allowed bubbles to be clearly seen when illuminated from below. The compressed air sprayer was used to spray model paint onto two glass substrates at the same time: one substrate was used to photograph bubble motion and the other to measure weight losses. The sprayer was moved at a constant speed across the substrate surface, keeping the spray pattern and flow rate constant throughout the experiments. The number of passes of the spray gun over the substrate determined the paint film thickness. A single pass of the sprayer deposited a paint thickness of approximately  $75-100 \,\mu m$ , so 1-6 passes were used in experiments. A balance (AL-204, Acculab, Bradford, MA, USA) was used to measure the weight of the substrate before and after spraying with a resolution of 0.1 mg and the weight of the paint calculated from the difference. The average paint thickness was calculated by dividing the paint mass by the density of the paint and the surface area of the test surface. The uncertainty in average thickness with this method was estimated to be less than  $\pm 1 \,\mu$ m.

After spraying paint, the glass substrate was placed horizontally above a LED light panel (Gagne Inc., Johnson City, NY, USA) that provided backlighting. It took approximately a minute to remove the substrates from the paint booth, position them under the camera and start photographing and as such time t = 0 marks the first photograph taken. Still images were taken at 5 s intervals for 15 min using a digital SLR camera (D90, Nikon Inc., Melville, NY, USA) mounted above the substrate, pointing downward and triggered by a programmable timer (TC-N3, JYC Technology, Hong Kong, China). The images were taken at 4288 × 2848 pixel resolution. The pictures were analyzed using the threshold function in image analysis software (Imagel, National Institute of Health, Bethesda, MD, USA) to count the number of bubbles in each image and the cross-sectional area of each bubble. Thresholding (Binary Contrast Enhancement) sets a grayscale cutoff point and uses it to transform a grayscale image to binary (black and white). Grayscale values below the limit become black and those above become white. Thus after thresholding, bubbles may no longer look perfectly circular and have a rather jagged outline. The figures shown in this study are the experimental photographs after thresholding.

ImageJ software does have limitations when it comes to distinguishing the individual bubbles in the early time intervals where there is clustering and a large number of bubbles are present in the paint film. This issue was mitigated through enhancing the contrast of the experimental images before thresholding and also fine-tuning the grayscale cutoff point to ensure that the figures produced after the threshold process closely resembled the experimental figures. The smallest bubbles resolved through this process had a diameter of  $4 \,\mu$ m which corresponds well with the  $5 \,\mu$ m value seen in literature for bubbles entrapped from viscous droplet impact onto a glass substrate [9].

#### 3. Results & discussion

#### 3.1. Bubble movement and escape

Fig. 2 shows a sequence of images of bubbles in a paint film with average thickness  $260 \,\mu\text{m}$  on a glass substrate. Initially (t=0) there were a large number of bubbles in the paint film, which appear as

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