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# Following the Drying of Spray Paints Using Space and Time Contrast of Dynamic Speckle

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*The short drying times of spray paints are of great benefit in some applications, and this property allows for painting over almost immediately. However, this fast-drying process precludes following it using conventional techniques. In a previous article, we used the dynamic speckle to follow the drying process of solvent-based and water-based paints with relatively long drying times, and recently we developed alternative speckle contrast methods to characterize faster processes. This article presents the application of these methods using dynamic speckle techniques to study the drying of a spray paint. Activity image display is also included.*

*Keywords: Dynamic speckle techniques, drying of spray paints*

Many industrial processes involve the coating of substrates with thin layers of paint to protect and/or decorate them and to impart desirable properties such as gloss, adhesion, magnetic properties, etc., and new coating products are continually being developed for other specific uses. The coating process involves the application of paint using a wide variety of applicators such as brushes, spray guns, roll coaters, extrusion dies, etc. Drying is part of the coating process and, therefore, the knowledge of the drying process has practical applications. For example, it is very important to establish the shortest time necessary for drying of the first layer before the application of the second layer.

After applying paint, the initial drying process consists of a constant slope falling period of loss of mass.[1] In that period, the coating behaves as if it were a pool of solvent. There is always solvent on the surface of the coating and the rate of evaporation of drying is controlled solely by factors external to the coating. The geometry, air velocity, air temperature, and solvent level in the drying air and any source of heat are the most important factors. In some cases, the constant rate period may be absent and all the drying occurs in the falling rate period. Different paint formulations behave in different ways. For example, in waterborne coatings most of the drying takes place in the constant rate period, and in solventborne coatings the constant rate period is short or even nonexistent. Spray paint is a paint product in an aerosol. There are different types including general spray paint, car enamel, vehicle rim spray paint, galvanizing spray paint, hobby and decorative spray paint, marking spray paint, sealant spray, and radiator paint. Spray paint is, therefore, a very useful type of paint application and the drying process is an important issue. In a previous article[2] we used dynamic speckle interferometry (DSI) to study the drying process of waterborne and solventborne paints, where the drying times were long enough to follow the process properly. However, faster drying processes like those found in spray paint applications were not possible to follow. In this article, we used a similar laser speckle technique to characterize faster drying processes. Before describing the new method used here, we will briefly discuss the origin of the speckle phenomenon.

When a coherent beam coming from a laser illuminates an object, a typical "speckle" pattern is observed.[3] If the

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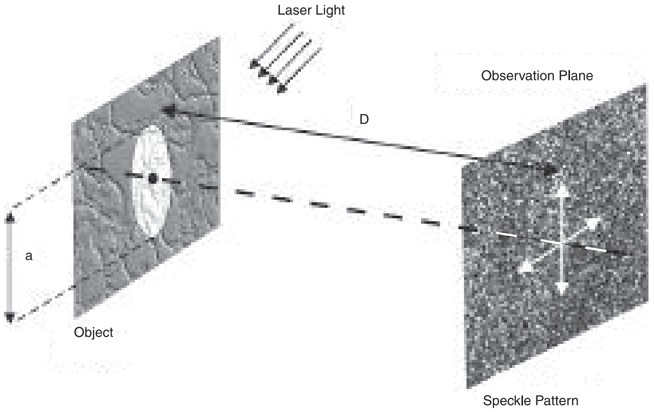


Figure 1—Schematic physical principle of speckle pattern.

surface of the object does not remain rigid, but rather presents some type of local movement, then the intensity diagram of the observed speckle evolves in time. This phenomenon, well known as "dynamic speckle," is characteristic of biological samples.[4–6] This behavior of the speckle pattern can also be observed in nonbiological industrial processes, including the drying of paint, corrosion, and heat exchange.[7,8] The visual appearance of the speckle diagram is similar to that of a boiling liquid.

This activity takes place when the sample changes its properties due to movement of the scattering centers, changes in the optical path due to variations of refractive index, configuration changes, or a combination of these situations.

Many efforts have been made to assign numbers that characterize this activity and that correlate favorably with alternative measurement methods of interest for the experimenter. The study of the temporary evolution of the speckle patterns may provide an interesting tool to characterize the parameters involved in these processes.

In a recent article,[2] we compared the results from speckle experiments with those obtained from conventional gravimetric measurements. There, the second order moment of the co-occurrence matrix of the speckle time history was used to characterize the evolution of the process. In that case, the tested paints dried in about 90 minutes. As stated above, using the same technique for faster drying paints, like traffic road, spray, and nail paints, results in sampling times that are longer than suitable intervals. The method is therefore inadequate.

In order to solve this problem, two alternative methods for characterizing the temporal evolution of faster processes were developed and used in this article. One of the proposed methods is based on the use of the "time contrast" of the speckle pattern history. In using this method, we describe an activity image that displays the case when nonuniform activity is presented across the surface. The other technique, based on "space contrast," uses a similar measurement but in the space domain. Basically, it measures the blurring of the speckle, and, in some cases, allows this approach in faster processes where the use of the other methods are not adequate. Methods using this concept have been developed by Briers.[9,10] The two methods are used here to follow the drying process after application of a spray paint but they might be used for other applications like those devoted to the characterization of dynamic processes in biological and industrial cases.

## THEORY

The surfaces of most of the materials are rough in the scale of optical wavelength ($\lambda \approx 6 \times 10^{-7}$ m). When laser light illuminates these surfaces, the scattered light acquires a granular appearance known as "laser speckle." This phenomenon is observed in a distant point from the surface, either in free propagation ("Fresnel speckle") or through an optical system ("Fraunhofer speckle"). The speckle pattern is produced by the interference between the coherent waves coming from the different microscopic elements of the surface. *Figure* 1 shows a typical speckle pattern in a free propagation configuration.

The random nature of the speckle diagrams requires the use of statistical tools for studying this phenomenon. The statistical properties depend on the coherence of the incident light and of the characteristics of the diffusing surface.

Using statistical analysis of the speckle patterns produced by a Gaussian laser beam,[3] the probability density function for the intensity distribution $I(x,y)$ of fully developed speckle patterns generated by a rigid diffuser is:

$$P\,(I) = (1/2\sigma^2)\ exp\ (-\ 1/2\sigma^2) \qquad (1)$$

where $\sigma$ is the standard deviation of the intensity.

Also, the second order of statistics[3] provides an estimation of the average size of the speckle grains. For example, if the illuminated area of the sample is a circular region with diameter $a$, the radius of the Fresnel speckle observed at a distance $D$ from the sample is:

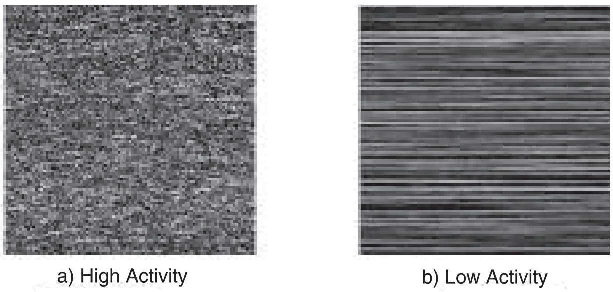
$$r = 1.22\ (\lambda\ D/a) \qquad (2)$$

where $\lambda$ is the wavelength of the laser light; $D$ and $a$ are shown in *Figure* 1.

A useful parameter for measuring the fluctuations of intensity is the speckle contrast. The contrast is defined statistically as:

$$C = \sigma \,/ < I > \qquad (3)$$

For the case of a single pattern of polarized speckle, the contrast is maximum and close to 1. We are going to call this magnitude the "space contrast" (SC) of the speckle pattern. In general, for the case of a sum of speckle patterns, the contrast diminishes. For example, for a sum of



a) High Activity b) Low Activity

Figure 2—Typical cases of THSP for (a) high activity and (b) low activity.

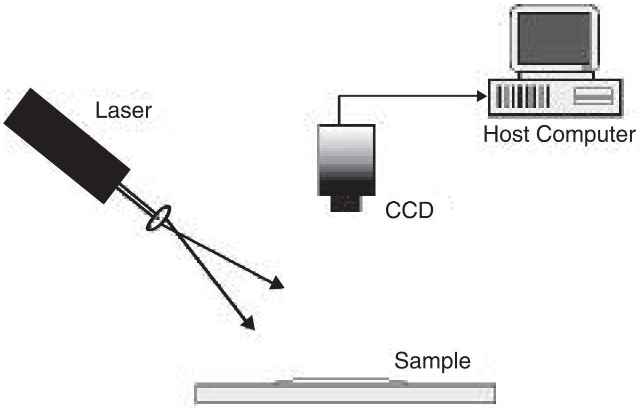


Figure 3—Experimental set-up showing the applied spray spot on the glass substrate.

N independent different speckle patterns, the associated contrast has a minimum value close to $1/\sqrt{N}$

As stated above, this concept has been applied to the analysis of speckle diagrams that evolve in time for the case of the drying of paint, but it might also be useful for other dynamic phenomena.

To understand how this concept is used, let us suppose that a layer of paint is extended on a surface. At the beginning the paint is "fresh" (just-applied), and therefore the observed speckle pattern presents a high local activity. To characterize that state, *N* images are registered and the contrast of the sum of the speckle diagrams is calculated. The resulting contrast of the speckle pattern is low.

On the other hand, when the paint is completely "dry," the speckle diagram does not reveal any activity and all *N* images are identical and equivalent to a single speckle pattern with maximum contrast (that is, the statistical independence condition between the patterns is not fulfilled). Between both extreme states, the images are partially correlated and the evolution of the process can be followed by means of the contrast for the averaged *N* registered images.

To illustrate the use of the second technique we are going to use the display method proposed by Oulamara et al.[11] For every state of the phenomenon being assessed, 512 successive images of the dynamic speckle patterns are registered using a CCD camera. Each image is digitized (512 rows × 512 columns measured in pixels) by a frame grabber at 8 bits gray levels and stored in the memory of the computer. A certain column (for example, the middle one) is selected in each of them. Then, a new image, named the "Temporary History of the Speckle Pattern (THSP)," is constructed by setting, side by side, the chosen column of the first image, the same column of the second image, and so on, 512 times. The vertical direction corresponds to the spatial direction of the speckle pattern and the horizontal direction represents the time evolution of this speckle pattern. So, the temporal activity of the sample appears as intensity changes in the horizontal direction. The rows represent different points of the object and the columns represent its time intensity variations. Every row of the THSP images was used to calculate a value of the contrast using equation (3). All the obtained values were averaged and we call this result the temporal contrast (TC) of the dynamic speckle pattern.

So, when a phenomenon shows low temporal activity, time variations of the speckle pattern are slow and small. In each row of THSP, the speckle appears "stretched" in the time direction showing, horizontally, an elongated shape. In the limit, when there is no activity at all, the THSP shows no variation in the horizontal direction. In this case, the TC is of low value or zero. Conversely, when the phenomenon is very active, the THSP shows fast intensity variations that resemble an ordinary spatial speckle pattern. The intensity variations are caused by changes of phase in the wave-front (also called phase excursions) and when these changes are important, the time contrast is greater than in the former case and eventually approaches the value 1. *Figure* 2 shows two typical cases of THSP for (a) high activity and (b) low activity.

Briers[12] theoretically studied the concepts of space and time contrast in a case where the dynamic speckle pattern was produced by a mixture of moving and stationary scatterers. He found that TC and SC could be related to the ratio of the mean intensity of the light from the moving scatterers to the total intensity of the scattered light.

## EXPERIMENTAL RESULTS

Experiments were carried out by spraying paint on a glass substrate. The spray paint was a commercial product (synthetic alkyd-based white enamel in aerosol) with about 44 wt% solids content. The experimental set-up is shown in *Figure* 3. An attenuated 10 mW He–Ne laser was used to illuminate the sample. A CCD camera connected to a personal computer with a frame grabber was used as a register medium to record the images. Fresnel speckle size was adjusted so that it was well resolved by the sensor by changing the distance D according to equation (2). The images of the diagram of dynamic speckle were taken every 0.08 sec (the fastest available time resolution). Each image was composed of 512 × 512 pixels and digitized to 256 intensity levels (8 bits). The average intensity of the laser was maintained constant in all the measurements. In our previous work, a gravimetric comparison was made, but in this case no such direct comparison was possible,

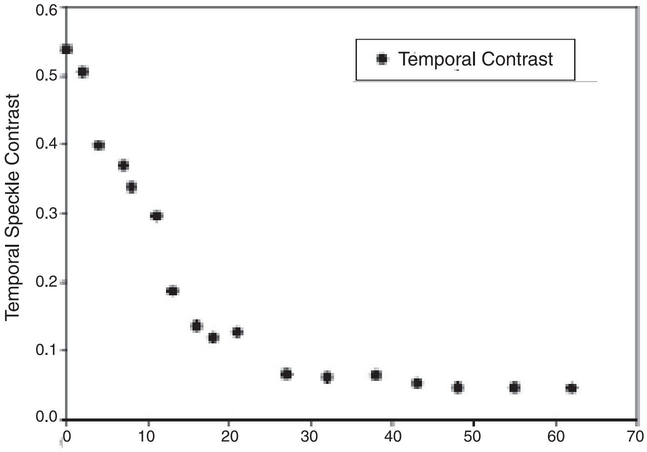


Figure 4—Experimental curve of drying of paint process using the temporal contrast method.

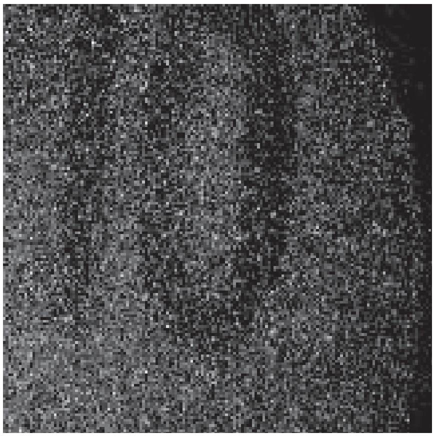


Figure 5—Activity image display of a spray-painted coin.

due to the way in which the spray paint was applied. When applying liquid paint with a film-applicator it is possible to obtain an almost constant thickness film and edge effects; that is, the changes in the thickness of the films at the periphery of the sample on glass can be ignored and a negligible influence on the gravimetric data is expected. No edge effects are expected in the optical data collections in all cases, because the laser beam is focused in the central zone of the sample. However, in spray applications, a nonuniform sample is applied and edge effects are expected to be important. Bearing this situation in mind, some gravimetric curves were obtained by spraying the paint on glass substrates and starting the data collection (weight changes) as soon as possible. The drying curves, though not expected to match the "speckle" curves, followed the general pattern observed previously, i.e., three zones where the rate of weight change was different in each of them.

### *Temporal Contrast Method*

The state of the sample was controlled every two minutes at the beginning of the process and then every five minutes in the final part. The evolution was followed for one hour.

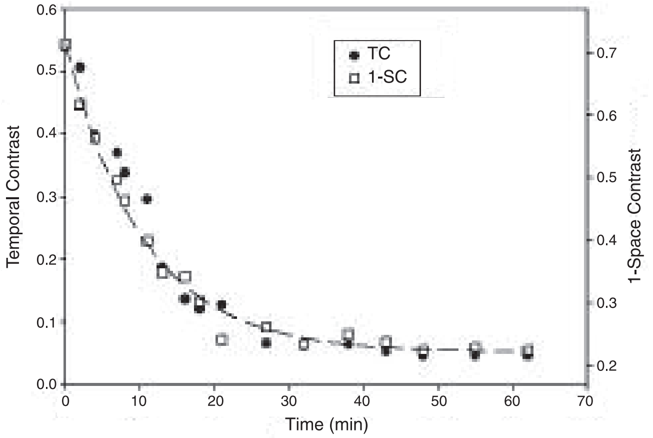


Figure 6—Experimental curves of drying of paint process using the temporal (TC) and the space contrast (SC) methods (a dashed line was included as a guide for the eyes).

Using the Oulamara technique,[11] a THSP image was obtained for each sampling drying state in approximately 40 sec. For this THSP image the temporal contrast was calculated and averaged.

*Figure* 4 shows an example of the application of the method of temporal contrast (TC) for the spray paint. In this case, the curve fell because the activity of the sample appeared as changes of intensity in the horizontal (time) direction. When it was very active ("fresh" or just-applied paint), the THSP resembled a diagram of ordinary speckle, and, therefore, with high speckle contrast. When the activity diminished, the THSP showed elongated forms and the mean speckle contrast was smaller. It was observed here that a stationary state corresponding to a "dry" paint state was reached in approximately 30 min. The intersection of slopes of the fast rate and the stationary state was about 20 min and should coincide with the "set-touch" time of the state of dryness of the paint, where almost all the solvents were evaporated. The set-touch time provided from the manufacturer was 15 min in an ordinary application, i.e., a thin layer. In our case, for experimental convenience, a larger amount of paint was applied, so larger times for set-touch were expected. This curve monotonically decreased as expected for a drying process of paints.[2] However, it was not regular. This irregular behavior can be attributed to the fact that aerosol paints dry faster than liquid paints and since each THSP takes about 40 sec to be registered, the sampling rate may not be adequate. Anyway, the typical features of the drying process were observed, i.e., the three zones with variable rate of changing: the initial state of the process with a constant rate of change, the falling rate period, and the almost zero rate of change state.

To verify the reproducibility of the results, we performed the same experiments in different replicate films with the same ambient conditions. It is important to note that there are always stray currents of air in a room, so perfectly "same ambient conditions" are rarely achieved, but for our purposes we can consider them as acceptable.

### *Activity Images*

The concept of temporal contrast can be applied to construct images, named "activity images," depicting the loci of points with the same activity.[13] In this case, a set of $N$ images is recorded by using an optical system, so a Fraunhofer speckle pattern is formed. These $N$ images are stored in the frame grabber and the TC is calculated for the same pixel along the $N$ images. Then, a new image is constructed by assigning each pixel a gray level proportional to its local TC.

*Figure* 5 shows an example where an inhomogeneous paint drying pattern was deliberately induced using a coin as substrate. By applying enough paint to cover the surface of the coin, the pattern of the coin becomes "invisible to the eye." In this case, the uneven coin surface allows for the application of layers of different thicknesses of paint and consequently different drying states, and therefore different surface activities are observed. As a consequence of those height variations, paint dries faster in the upper levels (thinner layers) and remains "wet" in the lower ones (thicker regions). On the spray painted surface of the coin we took $N = 100$ successive images and then the TC was calculated as above. As expected, the lower regions with "wet" paint appeared comparatively brighter.

### Space Contrast Method

In this case, for each drying state of the sample, we registered a set of $N$ consecutive spatial speckle images and then the intensity was averaged in each pixel. For this averaged image, the speckle contrast was calculated using equation (3).

Also, the evolution was followed for one hour and the states of the sample were controlled every two minutes at the beginning of the process and then every five minutes in the final part. For each state, $N = 20$ images were registered in less than five seconds.

When applying the spray paint, we observed that the contrast was low because the speckle pattern was blurred.[14] Then it grew monotonically and the maximum contrast was achieved in approximately 30 minutes, corresponding to the state of dryness of paint, as in the previous case. It resembled the case where we had a single polarized speckle pattern.

In order to get a monotonically decreasing curve over time, the following mathematical operation can be performed on the SC:

$$Y = 1 - SC \tag{4}$$

where $Y$ is the graph ordinate. In this way, the typical drying curve pattern is obtained. *Figure* 6 shows the variation of the spray paint on glass substrate according to equation (4). Over time and, for the sake of comparison, the data of the temporal contrast method is also included. We can clearly distinguish the three regions previously described. As can be seen in *Figure* 6 during the initial period, the data points obtained from the space contrast method describe the monotonously drying process behavior (dashed line) better than the temporal contrast method, because the time sampling is more appropriate to this type of fast drying paint.

## CONCLUSIONS

Two methods were developed, based on space and time speckle contrast, and were applied in a fast drying paint process, like that found in a spray paint. The experimental results showed suitable results using both spatial and temporal contrast methods, which encourage more in-depth research. In this case, for the fast drying spray paint, the spatial contrast method seemed to be more useful, as the sampling rate can be higher. The reproducibility of the dynamic speckle method used to characterize time-dependent process was estimated to be better than 5% for each state of sample. This technique is not absolute and detailed studies are in progress to determine quantitative reproducibility values and to assess the influence of sampling area, sample-detector distance, and ambient conditions on final results.

Activity images are useful for processes taking place on uneven surfaces, like those found when painting irregular objects. These methods seem to be complementary to others already developed for the study of paint drying[2,7,15] and might also be tested in other dynamic processes that show surface changes, as in biological tissues like vegetables, seeds, etc. Dynamic speckle techniques to study even faster phenomena are currently under development.

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