EVALUATION OF EFFECT PIGMENT ORIENTATION USING COMBINED SEM, TEM, AND X-RAY CT ANALYSIS AND CONCLUSION FOR INTERPRETING GONIOSPECTRO-PHOTOMETRIC DATA OF EFFECT COATINGS

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ABSTRACT

Plate-like effect pigments (with diameters between 5-40µm and less than 1µm thick) are widely used to create decorative appearance in coatings, prints, plastics and cosmetics. Thereby the color and texture do not only depend strongly on type and concentration of the effect pigment, but also on their orientation distribution in the corresponding medium. In order to predict the appearance of a given effect pigment, its orientation distribution obtained using a specific method of application (spray application, roller blade application, offset printing, brushing, etc.) should be measured. This is usually done by preparing mechanical cross sections and obtaining the 2D particle inclination distribution N(d) by semiautomatic image processing of microscope images.

INTRODUCTION

The appearance of a golden effect pigments applied with spray application (narrow orientation distribution) and powder coating application (broad orientation distribution) is shown in **Fig. 1** and **2**. The differences are obvious and to predict such differences the orientation distribution of the effect pigments should be measured with a high statistical accuracy. Mechanical cross sections allow in insight into the orientation behavior of the effect pigments, but the statistical evaluation of the data is quite troublesome and sometime not accurate enough.

The present poster describes the method of X-ray tomography (SkyScan model 2011 nano-CT) as a noncontact, nondestructive and fast method to examine the 2D and 3D particle orientation in effect coatings. In comparison to X-ray CT a step-by-step ion beam slicing using a FIB V600 CE+ (FEI) was utilised for the same coatings volume. These cross-sections were inspected to find the identical particles already seen in the tomogram and to evaluate them with respect to inclination, thickness and inter-particle distances by SEM (SU70 / Hitachi) and TEM imaging (Tecnai G2 F20 S TWIN / FEI).

EXPERIMENTAL AND DISCUSSION

Although the lateral resolution of all methods is quite different a good correlation could be found for individual particles as well as for big ensembles. In case of adding disorienter particles to the effect pigments this data were used successfully to interpret high resolution macroscopic goniospectrophotometric measurements (Murakami GCMS-4 Gonio-Photospectrometer).

In order to follow the logical way of this research figures on the next page are shown consecutively: **Fig. 3** gives the accustomed view of an ensemble of effect pigments as generated by (BF) light microscopy. The effect pigments are seen as platelets from the top to estimate particle areas and equivalent diameters. **Fig. 4** represents a mechanical cross-section through the effect coating imaged by SEM. Here we get the side view of the particles in order to estimate the thicknesses and more importantly the two-dimensional angular particle inclination δ with respect to the surface normal. N(δ) is close to a Gaussian with a standard deviation of less than 10°. **Fig. 5a** depicts the SEM view of the marked coatings area (volume enclosed between both crosses under the metal protection bar) to be analyzed by X-ray CT. The crosses are identified then in the X-ray CT images as shown in **fig. 5b** (for X-ray CT principles see the scheme in **fig. 6**). The measured and reconstructed ensemble of the effect particles in the volume is given in **fig. 5**. Along the line between both crosses a TEM lamella was prepared using FIB technique. One of the cross-sections is shown in **fig. 8a** as an X-ray CT section, in **fig. 8b** as a SEM – and in **fig. 8c** as a TEM – cross-section image. There is not a full correspondence between the pictures due to the different depth of analyzing, which is much bigger in X-ray CT than in TEM.

Identical volumes are marked by squares and the particles are evaluated with respect to inclination d and thickness. Due to the fact that TEM analysis need thin electron transparent sections of the volume (prepared from both sides of the lamella) a continuous transversal shift of the interface was followed by SEM imaging and correlated with X-ray CT images for a large volume. The Xray CT – SEM - TEM correlation in **fig. 8a, b, c** was carried out only at the final stage, having a thin lamellar residue suitable for TEM. The usage of X-ray CT evaluation of particle inclinations in correspondence to measured goniometric lightness distributions (here light incidence is 45° and the detection angle varies from +75° to -75°, corresponding to an aspecular angle φ from 30° to -120°) is demonstrated in **fig. 9a** – for a series of different effect pigment concentrations, where the lightness curves are shifted to higher values and **fig. 9b** – for a series of effect coatings with rising disorienter concentration, with a “rotation” of the lightness curves: less lightness in direction close to the specular and more lightness in strong aspecular directions. The behavior of log(L*)=f(φ) in both cases fits well with the Xray CT data of particle inclinations as given in the captures. Therefore the gonio-spectrophotometric approach and the X-ray CT data are best suited to study the effect particle orientation distribution in a sufficient large coatings volume and SEM /TEM analysis can be used to investigate detectability of the pigments, resolution and local peculiarities of the methods.

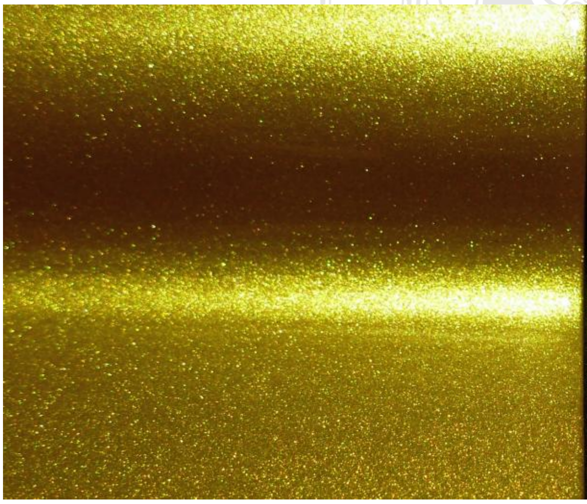

Fig. 1: Photograph of gold effect coatings produced by spray application method on a bended plate yielding strongly oriented effect pigments

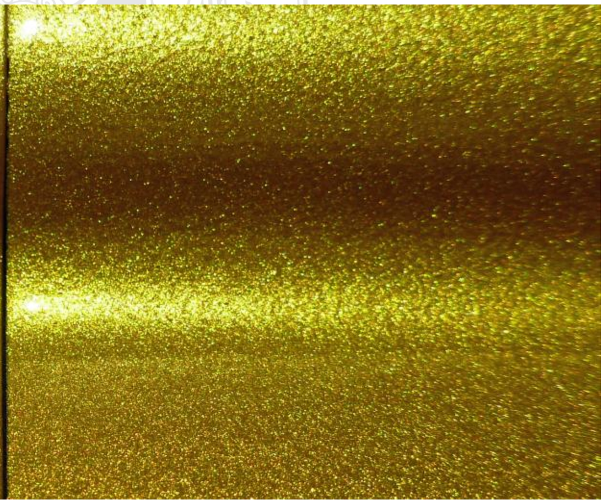

Fig. 2: Photograph of gold effect coatings produced by powder coating method on a bended plate yielding strongly disoriented effect pigments

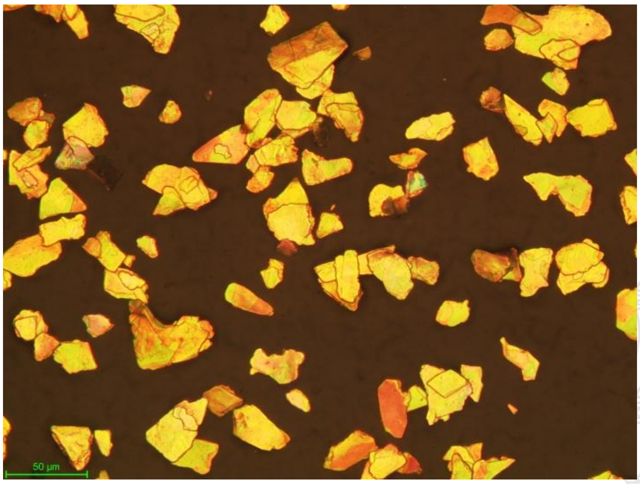

Fig. 3: Light microscope bright field image of the paint film

(1)air, (2)clear coat, (3)base coat (15 µm thick), (4)filler layers and (5)substrate

Fig. 4: Mechanical cross-section of the paint film imaged by SEM

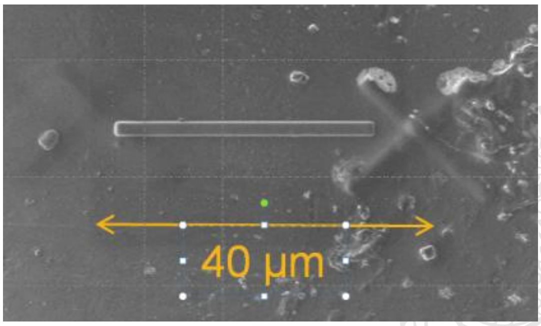

Fig. 5a: Paint surface area marked by crosses in the FIB and imaged by SEM

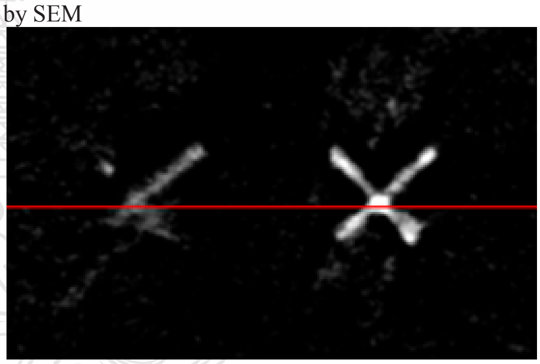

Fig. 5b: X-ray CT identification of the sample by finding the cross-markers

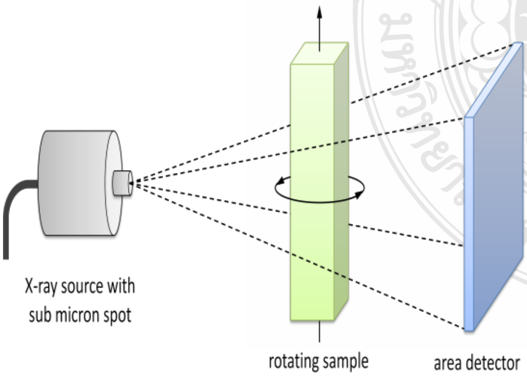

Fig. 6: Scheme of the X-ray CT imaging method

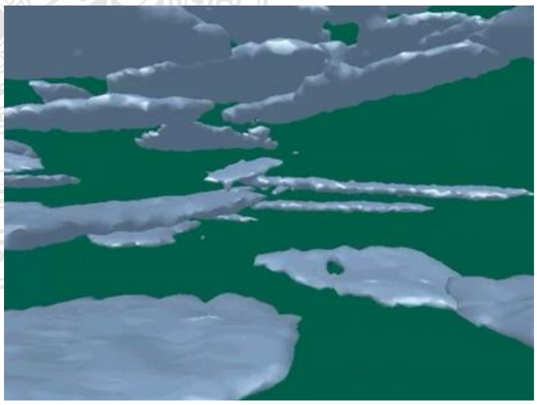

Fig. 7: Still picture of a movie showing the 3D particle ensemble

Poster paper

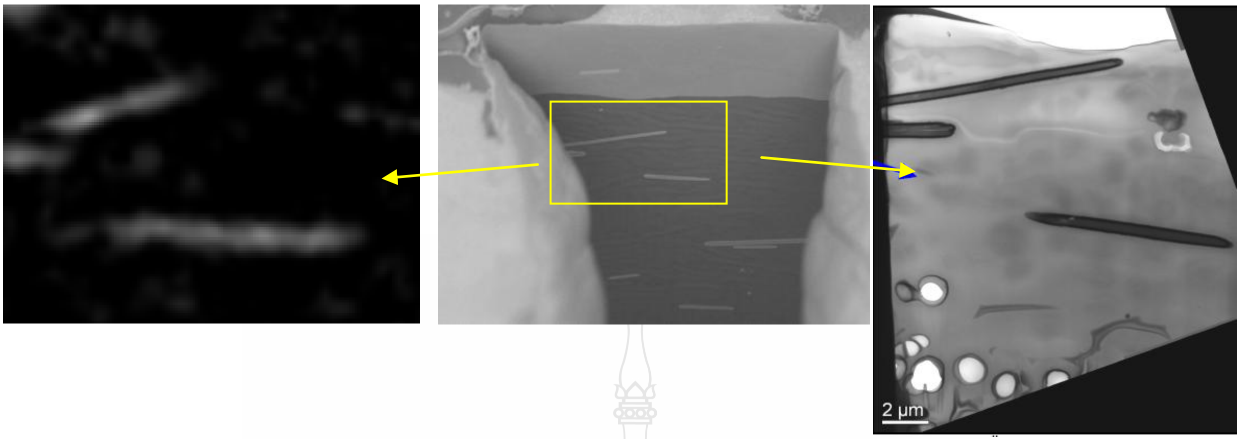

Fig. 8a, b and c: Identical location imaged by X-ray CT cross-section, by SEM of the FIB lamella and by TEM image of FIB lamella.

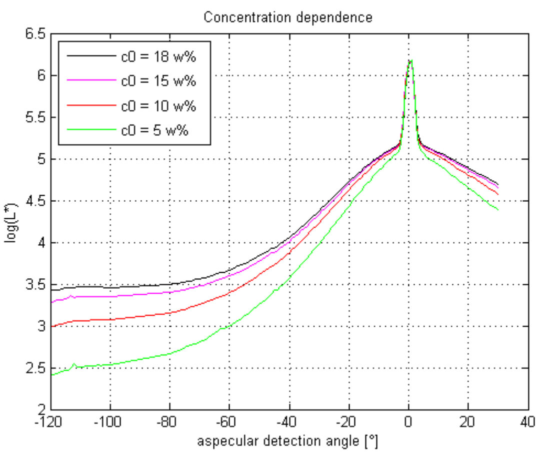

Fig. 9a: Log(L*) =f(φ) for different effect pigment concentration c0: the disorientation measured by X-ray CT is always δ=9.2° ± 0.5°.

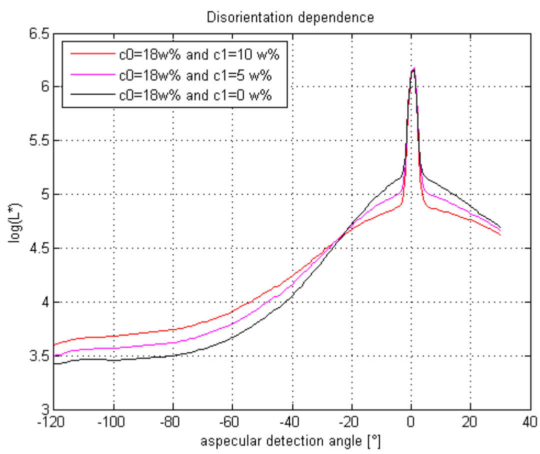

Fig. 9b: Log(L*) =f(φ) for 18w% effect pigment and different concentration of a disorienter c1: the disorientation measured by X-ray CT rises from 9.2°, via 10.5° to 13.1°.

REFERENCES

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