

# ChemistryOpen

Supporting Information

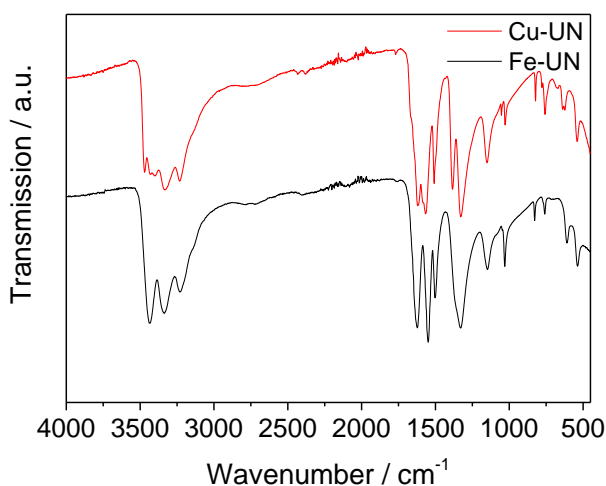
## **Environmentally Benign Solution-Based Procedure for the Fabrication of Metal Oxide Coatings on Metallic Pigments**

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### 1. Infrared spectra of the precursor compounds

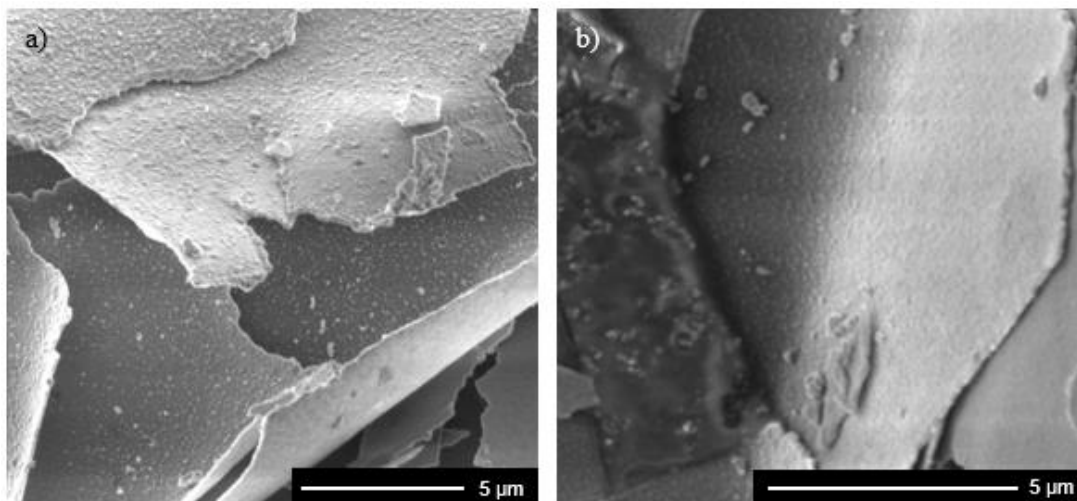


S 1. Infrared spectra of **1** (black) and **2** (red).

### 2. Influence of water addition on the coatings obtained from thermal decomposition of

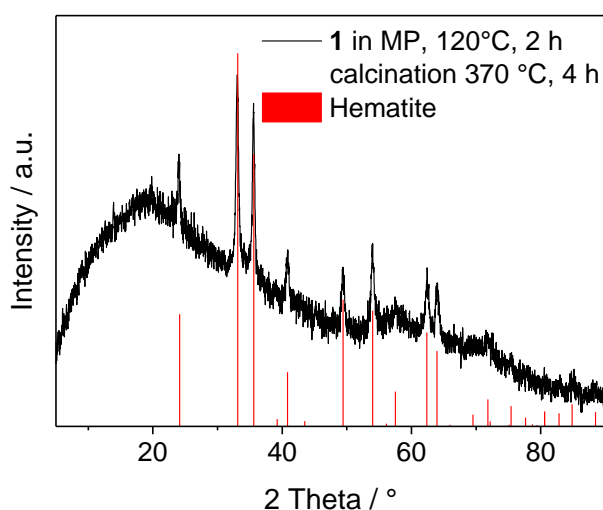
**1**

The addition of water to the reaction mixture used for the decomposition of **1** has a major effect on the organic residues. However, the coating process in terms of the topography of the coated aluminum pigments is not significantly affected. For pigments coated without addition of water, a gloss of 52.5 and a flop index of 23.6 were measured. For a sample obtained with addition of 1 % water, gloss and flop index are determined to be 51.6 and 22.4, respectively. As these values are highly depending on the pigment's surface, it can be concluded that the influence of water addition on the topography of the obtained product is negligible. SEM images further prove this assumption, as no significant difference can be observed (S2).



S 2: SEM images of coated pigments obtained from decomposition of **1** with (a) and without (b) addition of 1 % H<sub>2</sub>O.

### 3. XRD analysis of the calcined precipitate obtained from **1**



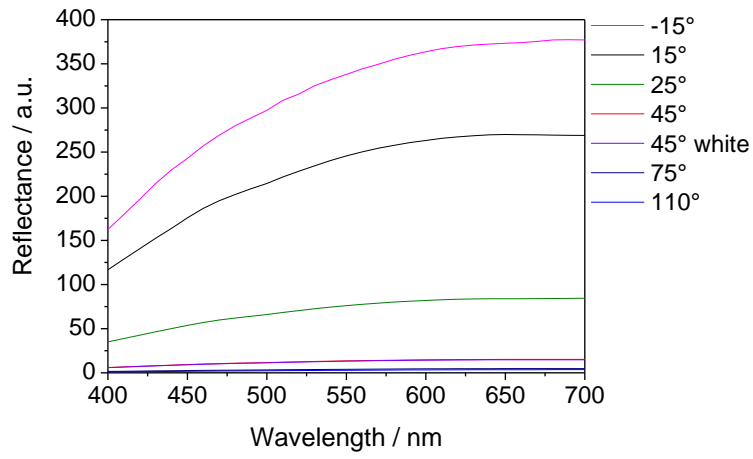
S 3. XRD analysis of the precipitate obtained from **1** in Methoxypropanol (MP), 120 °C, 2 h and subsequent calcination at 370 °C, 4 h (black) compared with hematite (red) (JCPDS 33-0664).

### 4. SLS analysis

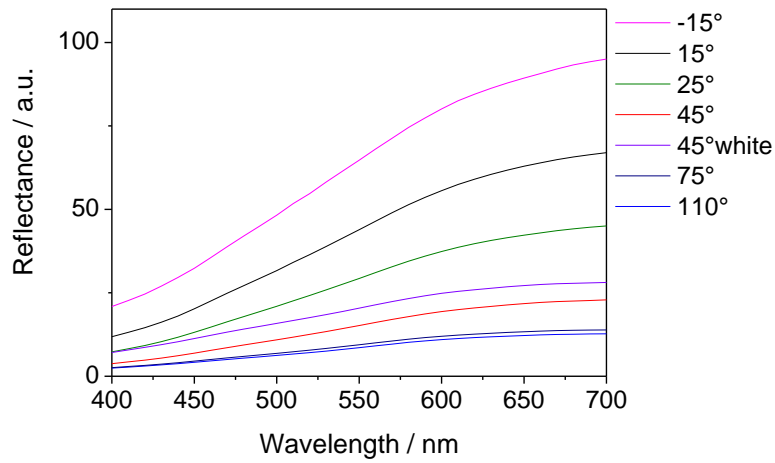
Sample treatment	d10 [μm]	d50 [μm]	d90 [μm]	Span
uncoated	10.6	22.3	35.3	1.11
1-opt; coated (1) but not dried	10.4	21.4	34.8	1.14
coated (1) and dried	12.2	25.7	42.0	1.16
1-nopt-calc; coated (1), dried, and calcined at ambient atmosphere, 400 °C, 4 h	12.5	29.0	56.1	1.51
1-opt-DPE coated (1), not dried, treated by refluxing in phenyl ether for 5 minutes	10.5	22.3	38.1	1.24
2-opt-DPE; coated (2), not dried, treated by refluxing in phenyl ether for 5 minutes	11.2	23.6	38.7	1.17

S 4. Particle size distributions of aluminum pigments uncoated, coated with the precipitate obtained from the thermal decomposition of **1**, after calcining these pigments in ambient atmosphere at 400 °C, after treatment in phenyl ether and after coating by thermal decomposition of **2** with subsequent annealing in phenyl ether for 5 minutes.

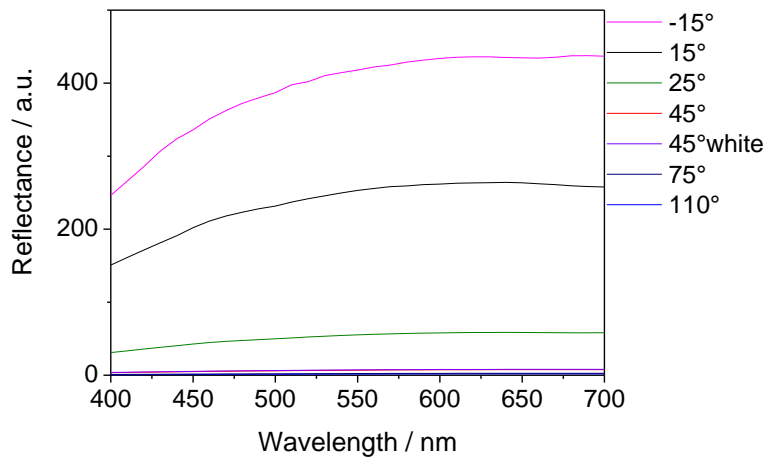
### 5. Optical spectra of the investigated pigment films



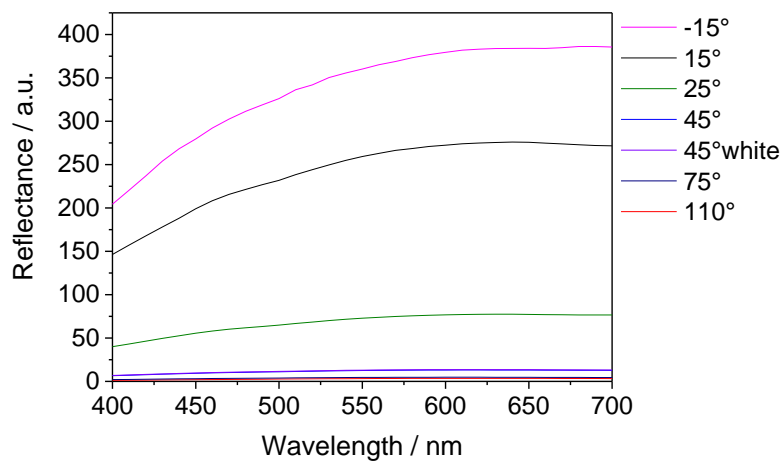
**S 5.** Angle dependent optical spectra of the pigments coated by thermal decomposition of **1** without optimization on a black background and 45° additionally on a white background.



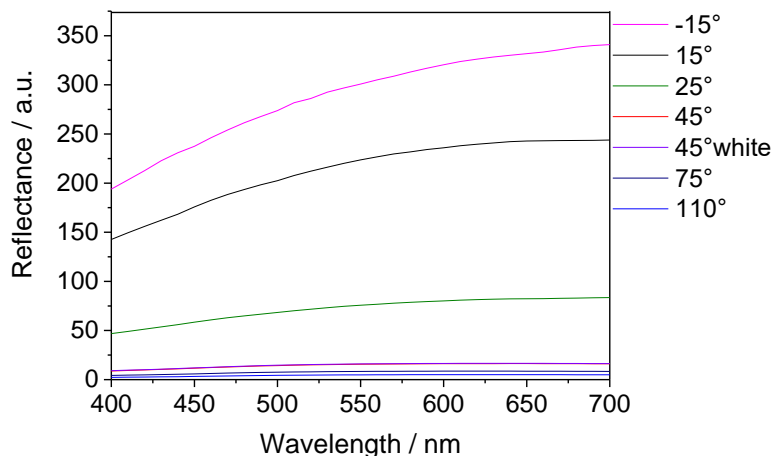
**S 6.** Angle dependent optical spectra of the pigments coated by thermal decomposition of **1** without optimization after calcination in ambient atmosphere, 4 h 400 °C on a black background and 45° additionally on a white background.



**S 7.** Angle dependent optical spectra of the optimized pigments coated by thermal decomposition of **1** on a black background and 45° additionally on a white background.



**S 8.** Angle dependent optical spectra of the optimized pigments coated by thermal decomposition of **1** after heat treatment in phenyl ether, 5 min 295 °C on a black background and 45° additionally on a white background.



**S 9.** Angle dependent optical spectra of the optimized pigments coated by thermal decomposition of **2** after heat treatment in phenyl ether, 5 min 295 °C on a black background and 45° additionally on a white background.

## 6. Colorimetric values of the investigated pigment films

Sample	Gloss	h15°	S15°	L*15	L*110	flop index	dE45°
Paliocrom® Brilliant Gold L 2050, 7 %	42.4	48.0	1.041	126.4	9.7	31.1	7.7
Alegrace® Aurous A 21/71-1 White Gold, 7 %	68,7	55,8	0,236	148,6	10,7	32,9	0,5
1-nopt - no solvent exchange – 3 %	51.6	67.2	0.269	143	20.9	22.4	0.8
1-nopt-calc - no solvent exchange - calcination at ambient atmosphere, 400 °C, 4 h - 3 %	6.4	54.3	0.694	72.6	35.9	5.4	7.1
1-opt - optimized procedure – 3 %	101	70.9	0.245	146	11.3	32.3	0.2
1-opt-DPE - optimized procedure - treatment in phenyl ether, 259 °C, 5 min - 3 %	46.9	65.7	0.333	138.7	19.1	21.5	0.3
2-opt-DPE - optimized procedure, treatment in phenyl ether, 259 °C, 5 min - 3 %	48	65.1	0.24	135.8	25.9	18.2	0.2

**S 10.** Results of the colorimetric measurements of the obtained pigments at different stages of the process compared with Paliocrom® Brilliant Gold L 2050 and Alegrace® Aurous A 21/71-1 White Gold. Bold numbers indicate the complex used for the precipitation; the percentage values represent the particle concentration in the varnish. Shown are the results for the gloss, hue angle (h), color saturation from 15° (S15°), lightness from 15° (L\*15) and from 110° (L\*110), flop index and the color distance between black and white background from 45° (dE45°).