

Yellowish-Brown Pigment with High Near Infrared Reflective

Thadsanee Thongkanluang^{1,a}, Jantharat Wutisatwongkul^{2,b}
 and Prayoon Surin^{3,c}

¹Program in Materials Technology and Manufacturing, Faculty of Science and Technology,
 Suratthani Rajabhat University, Suratthani, 84100, Thailand

²Faculty of Science and Technology, Suan Dusit Rajabhat University, Bangkok, 10300, Thailand

³Department of Industrials Engineering, Pathumwan Institute of Technology, Bangkok, 10330,
 Thailand

^athad2007@gmail.com, ^bjantharat85@gmail.com, ^cprayoon99@gmail.com

Keywords: Yellowish-Brown Pigment; Near Infrared Reflective

Abstract. Yellowish-brown inorganic pigments having a high near infrared solar reflectance have been synthesized. In this research, Fe_2O_3 was used as the host component, whereas, the mixtures of Sb_2O_3 , SiO_2 , Al_2O_3 , and TiO_2 were used as the guest components. The guest components were investigated over a range of 36 different compositions. The results showed that the pigment, denoted by YB32, with a composition of Fe_2O_3 , Sb_2O_3 , SiO_2 , Al_2O_3 , and TiO_2 of 65, 15, 10, 2 and 8 wt.% respectively, generated a maximum near infrared solar reflectance of 40.8% while the YB3 pigment was found to have a minimum reflectance of 29.3%. The CIE $L^*a^*b^*$ colour index was used to measure the yellowish-brown pigment colours. The YB32 and YB3 pigment powders were also characterized by powder X-ray diffraction technique. It was found that the YB3 powder developed a new phase, FeSb_2O_3 , which is mainly responsible for the decrease in the near infrared solar reflectance.

1. Introduction

NIR Reflective pigments are complex inorganic metal oxide. Inorganic metal oxide produce color pigment that use for camouflage coatings and cool color. Cr_2O_3 complex color pigments with near infrared reflective have been developed [1–4]. Army use chromium oxide (Cr_2O_3) green pigments for prevent detection of objects [5]. For cool color, TiO_2 white pigment has been used as a pigment with high near infrared solar reflectance [1]. The past few years, customers prefer to non – white cool pigment [6–8]. Reddish Brown near infrared reflective pigments have been developed for coating over a TiO_2 that coat on asbestos cement sheet. The researchers found that the NIR solar reflectance value is 49% in a thickness of $145\mu\text{m}$ [9]. In the present study, yellowish brown pigments based on a Fe_2O_3 - Sb_2O_3 - SiO_2 - Al_2O_3 - TiO_2 composition have been synthesized and studied on characteristic to be use as a reflective pigment.

2. Experimental

2.1 Pigment preparation

The synthesise new yellowish brown pigments with near infrared reflective. Materials used are of commercial grade. Fe_2O_3 , a brown pigment, was used as a host component and mixtures of Sb_2O_3 , SiO_2 , Al_2O_3 and TiO_2 were used as guest components. Sb_2O_3 , SiO_2 , Al_2O_3 and TiO_2 were mixed into 36 different compositions (denoted as samples YB1 to YB36) as shown in Table 1. For each sample preparation, 65 wt% of Fe_2O_3 was mixed with 15 wt% of Sb_2O_3 . The mixed guest components shown in Table 1. All mixed samples were calcined at $1150\text{ }^\circ\text{C}$ for 30 min by applying heat at a rate of $4\text{ }^\circ\text{C}/\text{min}$ and cooled down naturally to ambient temperature. The samples were milled in an agate ball mill for 7 min at a speed of 250 rev/min and then baked at $110\text{ }^\circ\text{C}$ for 30 min.

2.2 Reflectance measurements of pigment powders

The pigment powders were compressed in a mold to form of thin disks with a diameter of 2.7 cm and a thickness of 4 mm. The spectral reflectance of the samples was measured using a UV-Vis-NIR spectrophotometer (Shimadzu 3100) in the wavelength range from 300-2100 nm. The spectral reflectance data were used to calculate the solar reflectance of each sample. The solar reflectance value in the NIR for the wavelength range from 780-2100 nm was calculated in accordance with the procedures in ASTM standard number E891-87. The NIR solar reflectance values for all samples, pure Fe₂O₃, Sb₂O₃, SiO₂, Al₂O₃ and TiO₂ are shown in Table 1.

2.3 Colour measurements of pigment powders

The pigment powders were compressed in a mold to form of thin disks with a diameter of 2.7 cm and a thickness of 4 mm. Colour of the samples was measured using a UV-Vis-NIR spectrophotometer (Shimadzu 3100) in the wavelength range from 380-780 nm. The spectral reflectance data were used to calculate the colour of each sample. The program of Colour Analysis, version 3.02 were used to calculate value of CIE L* a* b* index as shown in Table 2.

2.4 Crystal structure characterization

The crystal structure of the pigment powder samples YB3 and YB32 was characterized by a X-ray diffractometer (Bruker, D8 Advance) with CuK_α radiation, 40 kV, 20 mA at 0.02 degree per step with a step time of 2 s and a scan time of 1.23 h. The XRD patterns were recorded in the 2θ range of 20°-70°.

3. Results and discussion

3.1 Reflectance results of pigment powders

Table 1 shows the NIR solar reflectance values for all samples in the wavelength range of 780-2100 nm. As can be seen, the NIR solar reflectance values having a maximum reflectance of 40.8% and a minimum reflectance of 29.3 % were obtained for the samples YB32 and YB3, respectively. As the results, the significantly increasing of solar reflectance values for YB32 pigment is due to the composition of guest components while conventional pure Fe₂O₃ yielded a solar reflectance of 26.3 %.

Table 1. The NIR solar reflectance values in the wavelength range from 780-2100 nm of pigment powder samples YB1-YB36. Pure Fe₂O₃, Sb₂O₃, SiO₂, Al₂O₃ and TiO₂ were also tested.

Sample	Composition (wt%)					Solar Reflectance (%)
	Fe ₂ O ₃	Sb ₂ O ₃	SiO ₂	Al ₂ O ₃	TiO ₂	
YB1	65	15	2	16	2	31.7
YB2	65	15	4	14	2	30.3
YB3	65	15	2	14	4	29.3
YB4	65	15	2	12	6	30.8
YB5	65	15	4	12	4	30.7
YB6	65	15	6	12	2	29.9
YB7	65	15	8	10	2	30.0
YB8	65	15	6	10	4	29.7
YB9	65	15	4	10	6	30.8
YB10	65	15	2	10	8	30.3
YB11	65	15	2	8	10	29.9
YB12	65	15	4	8	8	29.4
YB13	65	15	6	8	6	32.2
YB14	65	15	8	8	4	32.5
YB15	65	15	10	8	2	29.7
YB16	65	15	12	6	2	31.7
YB17	65	15	10	6	4	32.6
YB18	65	15	8	6	6	33.7
YB19	65	15	6	6	8	29.9

Sample	Composition (wt%)					Solar Reflectance (%)
	Fe ₂ O ₃	Sb ₂ O ₃	SiO ₂	Al ₂ O ₃	TiO ₂	
YB20	65	15	4	6	10	29.8
YB21	65	15	2	6	12	31.6
YB22	65	15	2	4	14	33.2
YB23	65	15	4	4	12	32.4
YB24	65	15	6	4	10	34.2
YB25	65	15	8	4	8	32.1
YB26	65	15	10	4	6	34.1
YB27	65	15	12	4	4	32.8
YB28	65	15	14	4	2	33.1
YB29	65	15	16	2	2	37.2
YB30	65	15	14	2	4	36.3
YB31	65	15	12	2	6	35.0
YB32	65	15	10	2	8	40.8
YB33	65	15	8	2	10	36.1
YB34	65	15	6	2	12	35.7
YB35	65	15	4	2	14	35.6
YB36	65	15	2	2	16	34.4
Fe ₂ O ₃	100	-	-	-	-	26.3
Sb ₂ O ₃	-	100	-	-	-	84.8
SiO ₂	-	-	100	-	-	89.4
Al ₂ O ₃	-	-	-	100	-	98.1
TiO ₂	-	-	-	-	100	96.0

3.2 Colour results of pigment powders

Table 2 shows the CIE L*a*b* color index. It was found that yellowish brown pigments are resemble in tones of color.

Table 2. Shows the CIE L*a*b* color index of yellowish brown pigment powder and Fe₂O₃.

Samples	L*	a*	b*	Samples	L*	a*	b*	Samples	L*	a*	b*
YB1	44.75	6.02	12.54	YB14	43.66	6.97	15.32	YB27	42.50	7.51	16.53
YB2	42.09	5.79	12.08	YB15	41.73	6.64	14.33	YB28	41.65	7.61	15.94
YB3	42.75	5.79	11.89	YB16	41.15	7.11	15.05	YB29	42.40	8.60	17.27
YB4	43.54	6.62	13.70	YB17	41.51	7.11	15.62	YB30	41.90	8.78	17.38
YB5	42.43	6.09	13.40	YB18	41.58	7.10	15.60	YB31	43.24	8.92	17.98
YB6	40.31	5.53	11.09	YB19	42.23	6.96	15.56	YB32	43.38	8.78	17.97
YB7	41.29	6.25	12.30	YB20	40.63	6.54	15.26	YB33	43.07	8.61	18.05
YB8	40.15	6.32	13.68	YB21	43.51	7.65	16.90	YB34	43.32	8.88	17.03
YB9	41.22	6.50	13.96	YB22	44.56	8.35	17.24	YB35	43.49	8.87	16.76
YB10	43.10	6.15	13.63	YB23	44.08	7.72	16.67	YB36	44.64	13.29	17.69
YB11	42.87	6.30	14.08	YB24	42.97	7.60	16.65	Fe ₂ O ₃	35.75	9.85	3.30
YB12	43.56	6.47	14.65	YB25	42.35	8.02	17.25				
YB13	43.64	6.52	15.11	YB26	42.39	7.57	17.60				

3.3 Crystal structure of pigment powder

Fig. 1 shows the XRD patterns of sample YB32 that have the peaks of Fe₂O₃, Sb₂O₃, SiO₂ and TiO₂ been observed. YB32 XRD patterns revealed that Fe₂O₃ and guest compounds were not melt to form solid solution. For Fig. 2 shows the XRD patterns of samples YB3. The sample YB3 shows the peaks of Fe₂O₃, Sb₂O₃, Al₂O₃ and FeSb₂O₃ are clearly observed. However, no peaks of TiO₂ and SiO₂ appeared due to the lesser amounts exist in the composition of 4 and 2 wt%, respectively. FeSb₂O₃ is a new phase of solid solution that occurs during calcination. It can be conclude that new phase may be decrease in the near infrared solar reflectance.

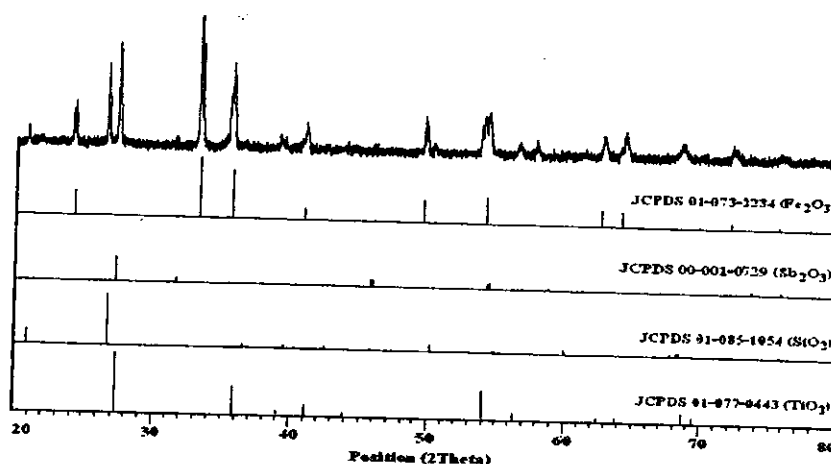


Fig. 1. XRD patterns of YB32 compare with JCPDS of Fe₂O₃, Sb₂O₃, SiO₂ and TiO₂.

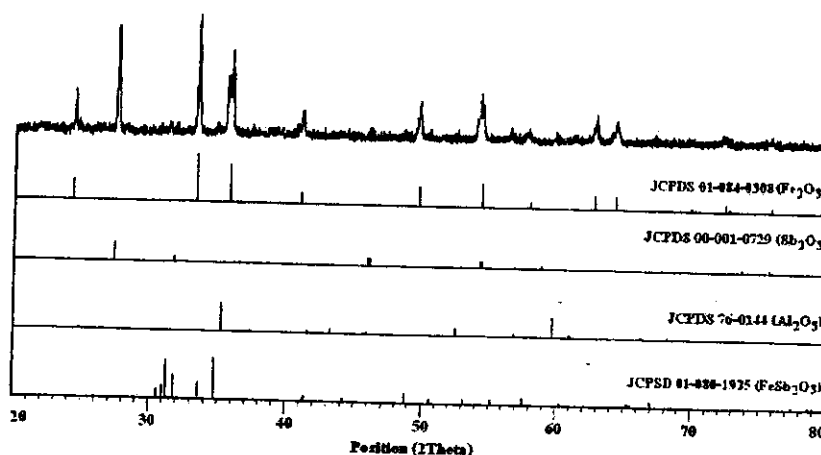


Fig. 2. XRD patterns of YB3 compare with JCPDS of Fe₂O₃, Sb₂O₃, Al₂O₃ and FeSb₂O₃.

4. Conclusions

Yellowish brown pigments having NIR solar reflectance have been developed. Pigment powders were prepared from Fe₂O₃ and mixtures of Sb₂O₃, SiO₂, Al₂O₃ and TiO₂. Then measure the reflectance value. The reflectance value show that the sample YB32 with a composition of Fe₂O₃, Sb₂O₃, SiO₂, Al₂O₃ and TiO₂ of 65, 15, 10, 2, and 8 wt%, respectively. YB32 pigment has a maximum NIR solar reflectance value of 40.8%. The study of XRD pattern shows new phase of FeSb₂O₃. It can be conclude that new phase may be decrease in the near infrared solar reflectance value.

Acknowledgements

This work was supported by Suratthani Rajabhat University.

References

- [1] R. Levinson, P. Berdahl and H. Akbari, Solar Spectral Optical Properties of Pigments-Part II: Survey of Common Colorants, *Sol. Energy Mat. & Sol. Cells.* 89 (2005) 351-389.
- [2] E. Ozel and S. Turan, Production and Characterization of Iron-Chromium Pigments and Their Interactions with Transparent Glazes, *J. Euro. Ceram Soc.* 23 (2003) 2097-2104.
- [3] G. Costa, V.P. Della, M.J. Ribeiro, A.P.N. Oliveira, G. Monrós and J.A. Labrincha, Synthesis of Black Ceramic Pigments from Secondary Raw Materials, *Dyes and Pigments.* 77 (2008) 137-144.

- [4] P. Li, H.B. Xu, Y. Zhang, Z.H. Li, S.L. Zheng and Y.L. Bai, The Effects of Al and Ba on the Colour Performance of Chromic Oxide Green Pigment, *Dyes and Pigments*. 80 (2009) 287-291.
- [5] R.P. Blonski, T.R. Sliwinski and R.A. Pipoly, U.S. Patent 6,174,360 B1. (2001)
- [6] A. Synnefa, M. Santamouris and K. Apostolakis, On the Development, Optical Properties and Thermal Performance of Cool Colored Coatings for the Urban Environment, *Sol. Energy*. 81 (2007) 488-497.
- [7] H. Takebayashi and M. Moriyama, Surface Heat Budget on Green Roof and High Reflection Roof for Mitigation of Urban Heat Island, *Building and Environment*. 42 (2007) 2971-2979.
- [8] R. Levinson, H. Akbari and J.C. Reilly, Cooler Tile-Roofed Buildings with Near-Infrared-Reflective Non-White Coatings, *Building and Environment*. 42 (2007) 2591-2605.
- [9] V.S.Vishnu and M.L.Reddy, Near-infrared reflecting inorganic pigments based on molybdenum and praseodymium doped yttrium cerate: Synthesis, characterization and optical properties, *Sol. Energy Mat & Sol.Cells*. 95 (2011) 2685-2692.