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Preparation of NIR Reflective Brown Pigment

T. Thongkanluang[a*], N. Chirakanphaisarn[b], P. Limsuwan[c]

[a]*Ceramics Technology Program, Faculty of Science and Technology, Suratthani Rajabhat University, Suratthani, 84100, Thailand*
[b]*Department of Computer Technology and Engineering, Faculty of Technology and Engineering, Siam Technology College, Bangkok, 10600, Thailand*
[c]*Department of Physics, Faculty of Science, King Mongkut's University of Technology Thonburi, Bangkok, 10140, Thailand*

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Abstract

Complex inorganic brown pigments having a high near infrared solar reflectance have been synthesized. Fe_2O_3 is the host component and mixtures of Sb_2O_3, SiO_2, Al_2O_3, and TiO_2 were used as the guest components. Sb_2O_3, SiO_2, Al_2O_3, and TiO_2 were mixed into 40 different compositions. It was found that a sample, denoted by S31, with a composition of Fe_2O_3, Sb_2O_3, SiO_2, Al_2O_3, and TiO_2 of 70, 10, 12, 2 and 6 wt% respectively, gives a maximum near infrared solar reflectance of 46.7%. Brown pigment colours were measured in CIE L*a*b* colour index. The S31 pigment powder was then prepared as a reflective coating material with different amounts of pigment powder from 4-8 g in 100 g of ceramic glaze. The prepared materials were sprayed on the biscuit clay tiles for reflectance measurements. It was found that 5 g of S31 pigment powder mixed with 100 g of ceramic glaze gives the highest near-infrared reflectance value of 41.3 %. The newly synthesized pigment is a suitable ceramic roof coating for its high reflectance performance and the durability performing once the ceramic roof installed on house.

Keywords: Brown pigment; Ceramic glaze; Near-infrared reflectance

1. Introduction

Inorganic NIR pigments are metal oxides. Two major applications are camouflage and reduced heat build-up. On a building, roof is a major part of heat absorption from solar radiation. The radiation fall upon roof is either reflected into the atmosphere or conductively absorbed into the building. Any solar radiation that is absorbed will heat the roof's surface and some of the heat transfers into the building. The higher the solar radiation absorbed, the more energy consumption, is required to keep the building cool; mostly by air

* Corresponding author. Tel.: +66-7735-5466; fax: +66-7735-5636.
E-mail address: thad2007@gmail.com.

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conditioners which are widely used in big buildings. Air-conditioning energy saving can be achieved by reducing the temperature of the building envelope, which in turn reduces the heat penetrating into the building. Exterior surface temperatures may be reduced by protecting the building envelope from the heat of solar radiation. Several techniques have been proposed for protection from the solar radiation. Among them, the use of cool materials has gained a lot of interests during the past few years. Cool materials refer to pigments having high near infrared (NIR) solar reflectance or low NIR solar absorptance [1] have been widely used as coating on tile roofs and walls.[1-3]

There are currently a number of cool materials commercially available for tile roof coating [4-6]. Inorganic pigments are widely used as cool materials for residential roofs. TiO_2 rutile; a white pigment with a high NIR solar reflectance of about 87.0% [4], is currently regarded as the best pigment for coating materials. However, owners of homes with pitched roofs visible from ground level often prefer non-white roofing products for aesthetic reasons [1,3,7].

Fe_2O_3 brown pigments have been used as coating materials for many kinds of roof to reduce the roof temperature and also simultaneously improving the roof's appearance. Various processes for preparing Fe_2O_3 brown pigments have been developed by researchers [8-9]. Very few studies have reported on complex brown pigments. Also recently, pigments based on rare earth compounds have been extensively studied [10-14]. However, the cost of the rare earth compounds used for synthesizing of pigments is rather high and therefore is not economically viable. In addition, most of the owners of homes with pitched roofs in Thailand prefer brown roof products.

In the present study, new 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 the ability of these pigments used in ceramic glaze for NIR reflective.

2. Experimental

2.1. Pigment preparation

This study is to synthesize new brown pigments of high near infrared reflectance for industrial applications, all materials used are of commercial grade. Fe_2O_3, a brown pigment oxide, 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 40 different compositions (denoted as samples S1 to S40) as shown in Table 1. For each sample preparation, 70 wt% of Fe_2O_3 was mixed with 10 wt% of Sb_2O_3. Then Fe_2O_3 and Sb_2O_3 were mixed with 20 wt% of guest component. The mixed guest components shown in Table 1. All mixed samples were calcined at 1150 °C for 30 min by applying heat at a rate of 4 °C/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 °C for 30 min to release water. Finally, the pigments were sieved to obtain the particle sizes of 0.5-2.0 μm.

2.2. Spectral 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 Fe_2O_3, Sb_2O_3, SiO_2, Al_2O_3 and TiO_2 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. Reflectance of pigment powder in ceramic glaze

It was found, from the spectral reflectance measurement of pigment powders, that the sample S31 gives a maximum reflectance of 46.7 % (see Table 1). Therefore, the sample S31 will be used for further study on reflective coating. In the preparation of reflective coating, the S31 pigment powder was divided into five different groups with the amounts of pigment powder of 4,5,6,7 and 8 g, respectively. Each group was then mixed with 100 g of a commercial ceramic glaze and two samples were prepared for each group. The mixtures were grounded in water by a ball mill for 10 min at a speed of 100 rev/min to obtain very homogeneous slurries with the specific gravity of about 1.40. All prepared samples were sprayed on the biscuit clay tile of dimension 4 cm x 5 cm by a spray gun. The coated clay tile substrates were heated at a rate of 4 °C /min until the maximum temperature of 1100 °C was reached and soaked for 30 min. The samples cool down to room temperature in the air. The coated biscuit clay tile substrates was measured reflectance in the wavelength range of 300 - 2100 nm and calculated in accordance with the procedures in ASTM standard number E891-87.

3. Results and discussion

3.1. Reflectance results of pigment powders

The reflectance spectra of some samples in the wavelength range from 300-2100 nm are shown in Fig. 1 and the NIR solar reflectance values for all samples in the wavelength range of 780-2100 nm are shown in Table 1.

As can be seen in Table 1, the NIR solar reflectance values having a maximum reflectance of 46.7% and a minimum reflectance of 19.6 % were obtained for the samples S31 and S9, respectively. As the results, it can be concluded that the significantly increasing of solar reflectance values for S31 pigment sample prepared in this work is due to the composition of guest components. A conventional pure Fe_2O_3, brown color, from the measurement result yielded a solar reflectance of 26.3 %.

The role of Sb_2O_3, SiO_2, Al_2O_3 and TiO_2 concentrations on the NIR solar reflectance values can be further analyzed. As shown in Table1, the NIR solar reflectance values for pure of Sb_2O_3, SiO_2, Al_2O_3 and TiO_2 are 84.8, 89.4, 98.1 and 96.0, respectively. It is seen that guest component yields high NIR solar reflectance value, when Fe_2O_3 powder mixed with each guest component yields reflectance values are 32.8, 37.2, 36.7 and 33.8% (S37, S38, S39 and S40, respectively). The NIR solar reflectance value of S7 - S10 are low values that composted of SiO_2 2-8 wt%, Al_2O_3 10 wt% and TiO_2 2-8 wt%. High reflectance values of S30, S31, S32 and S33 are 37.1, 46.7, 41.6 and 35.2, respectively. The reflectance values shown the composition composed of SiO_2 8 - 14 wt%, Al_2O_3 2 wt% and TiO_2 4 - 10 wt%. It can be concluded that the suitable amount of added Al_2O_3 is 2 wt% and SiO_2 8 - 14 wt%. Hence sample S31 with a composition of Fe_2O_3, Sb_2O_3, SiO_2, Al_2O_3 and TiO_2 of 70, 10, 12, 2 and 6 wt% is the most suitable pigment powder for ceramic roof coating.

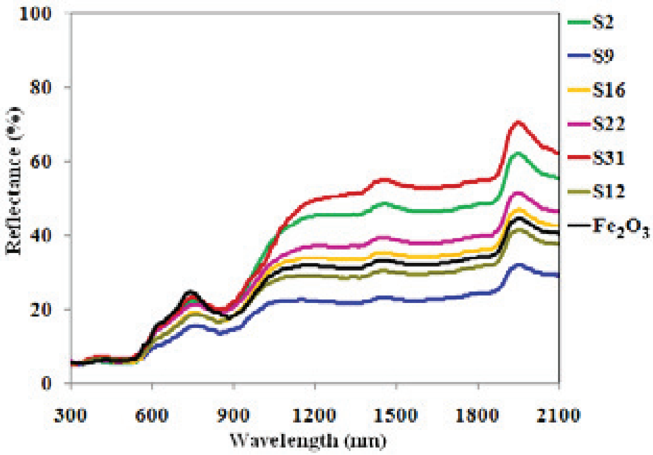

Fig. 1. Some spectra of NIR solar reflectance in wavelength range of 300-2100 nm

Table 1. The NIR solar reflectance values in the wavelength range from 780-2100 nm of pigment powder samples S1-S40. For comparison, pure Fe_2O_3, Sb_2O_3, SiO_2, Al_2O_3 and TiO_2 were also tested

Sample	Composition (wt%)					Solar Reflectance (%)
	Fe_2O_3	Sb_2O_3	SiO_2	Al_2O_3	TiO_2	
S1	70	10	2	16	2	34.0
S2	70	10	4	14	2	34.8
S3	70	10	2	14	4	28.9
S4	70	10	2	12	6	25.0
S5	70	10	4	12	4	28.7
S6	70	10	6	12	2	26.4
S7	70	10	8	10	2	20.2
S8	70	10	6	10	4	22.7
S9	70	10	4	10	6	19.6
S10	70	10	2	10	8	20.9
S11	70	10	2	8	10	28.2
S12	70	10	4	8	8	25.0
S13	70	10	6	8	6	22.3
S14	70	10	8	8	4	29.5
S15	70	10	10	8	2	23.5
S16	70	10	12	6	2	27.7
S17	70	10	10	6	4	24.8
S18	70	10	8	6	6	23.4
S19	70	10	6	6	8	20.5
S20	70	10	4	6	10	21.6
S21	70	10	2	6	12	22.7
S22	70	10	2	4	14	30.5
S23	70	10	4	4	12	25.7
S24	70	10	6	4	10	28.5
S25	70	10	8	4	8	31.2
S26	70	10	10	4	6	33.8
S27	70	10	12	4	4	23.7
S28	70	10	14	4	2	24.3
S29	70	10	16	2	2	30.0
S30	70	10	14	2	4	37.1
S31	70	10	12	2	6	46.7
S32	70	10	10	2	8	41.6
S33	70	10	8	2	10	35.2
S34	70	10	6	2	12	31.6

Table 1. The NIR solar reflectance values in the wavelength range from 780-2100 nm of pigment powder samples S1-S40. For comparison, pure Fe_2O_3, Sb_2O_3, SiO_2, Al_2O_3 and TiO_2 were also tested (Cont.)

Sample	Composition (wt%)					Solar Reflectance (%)
	Fe_2O_3	Sb_2O_3	SiO_2	Al_2O_3	TiO_2	
S35	70	10	4	2	14	33.0
S36	70	10	2	2	16	31.1
S37	70	10	20	-	-	32.8
S38	70	10	-	20	-	37.2
S39	70	10	-	-	20	36.7
S40	70	10	-	-	-	33.8
Fe_2O_3	100	-	-	-	-	26.3
Sb_2O_3	100	-	-	-	-	84.8
SiO_2	100	-	-	-	-	89.4
Al_2O_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 brown pigments are resemble in tones of color.

Table 2. Shows the CIE L*a*b* color index of brown pigment powder and Fe_2O_3 was also tested

Samples	**L***	**a***	**b***	**Samples**	**L***	**a***	**b***
S1	38.75	10.86	8.74	**S20**	35.48	9.59	7.38
S2	38.28	13.31	10.04	**S21**	34.82	9.96	8.84
S3	36.32	12.30	7.96	**S22**	36.95	12.16	12.56
S4	35.66	9.43	6.59	**S23**	36.11	11.22	9.88
S5	36.47	12.63	8.18	**S24**	36.30	12.40	9.33
S6	35.53	10.76	6.78	**S25**	35.81	14.63	10.33
S7	34.58	8.40	5.19	**S26**	36.78	14.67	10.23
S8	35.51	12.30	8.10	**S27**	35.27	11.84	7.61
S9	35.46	8.81	5.78	**S28**	34.53	11.86	7.38
S10	35.93	7.72	6.01	**S29**	34.44	14.22	9.04
S11	37.16	10.73	8.47	**S30**	36.49	14.26	10.03
S12	36.95	9.95	7.47	**S31**	35.57	15.92	11.23
S13	34.92	9.11	6.25	**S32**	36.53	14.76	10.61
S14	35.34	12.95	8.71	**S33**	35.41	14.31	11.16
S15	34.63	10.63	6.59	**S34**	34.39	12.26	9.63
S16	35.29	12.54	7.91	**S35**	37.56	13.24	13.93
S17	34.71	11.78	7.66	**S36**	40.55	13.29	18.27
S18	35.37	10.48	7.00	**Fe_2O_3**	35.75	9.85	3.30
S19	35.46	9.81	6.93				

3.3. Reflectance result of coated biscuit clay tile substrates

The S31 pigment powder of different amounts of 4-8 g were mixed with ceramic glaze and sprayed on biscuit clay tile substrates, as described in Section 2.4, were prepared and the spectral reflectance were measured. Then, the NIR solar reflectance values in the wavelength from 780-2100 nm were determined. The NIR solar reflectance values of the coated biscuit clay tile substrates are shown in Table 3. It is seen that the maximum NIR solar reflectance value was obtained from 5 g of S31 pigment powder mixed with ceramic glaze.

Table 3 The NIR solar reflectance of the S31 pigment powder coated biscuit clay tile substrates

Pigment Amount (g)	Solar Reflectance (%)		
	Sample1	Sample2	Average
4	37.4	37.0	37.2
5	41.0	41.6	41.3
6	41.0	40.8	40.9
7	40.8	40.3	40.6
8	39.6	39.2	39.4

4. Conclusions

Brown pigments having a high NIR solar reflectance for roofing materials have been developed. Pigment powders were prepared from Fe_2O_3 and mixtures of Sb_2O_3, SiO_2, Al_2O_3 and TiO_2. The composition of host component, Fe_2O_3 and Sb_2O_3 was fixed at 70 and 10 by wt%, respectively. Then host component mixed into 39 different compositions of 20 wt% of SiO_2, Al_2O_3 and TiO_2. All prepared samples were calcined at 1150 °C for 30 min. The reflectance spectra show that the sample S31 with a composition of Fe_2O_3, Sb_2O_3, SiO_2, Al_2O_3 and TiO_2 of 70, 10, 12, 2, and 6 wt%, respectively has a maximum NIR solar reflectance value of 46.7%. The S31 pigment powders were then prepared as reflective coating materials with different amounts of pigment powder from 4-8 g in 100 g of ceramic glaze. The prepared materials were sprayed on the biscuit clay tile substrates for reflectance measurements. It was found that 5 g of S31 pigment powder mixture gives the highest near-infrared reflectance value of 41.3 %. It can be concluded that the coating materials containing ceramic glaze and 5 g of S31 pigment powder is suitable for NIR reflective ceramic roof coating.

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