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## Influence of aluminum source on the color tone of cobalt blue pigment

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### ABSTRACT

Cobalt blue is an inorganic pigment with an intense blue color, and consists mainly of cobalt aluminate ( $\text{CoAl}_2\text{O}_4$ ) spinel. Many synthesis methods for cobalt blue have been reported, but there are no reports on the color tone of cobalt blue synthesized from different starting materials. Thus, the relationship between starting material and the color tone of the resulting product has not yet been clarified. We experimentally investigate the influence of different starting materials on the color tone of cobalt blue, synthesized by solid-state reaction. Cobalt oxide ( $\text{Co}_3\text{O}_4$ ) was used as the cobalt source, and three types of aluminum source,  $\text{Al}(\text{OH})_3$ ,  $\gamma\text{-Al}_2\text{O}_3$ , and  $\alpha\text{-Al}_2\text{O}_3$ , were used. The color tone of cobalt blue was evaluated by the lightness  $L^*$  and Chroma  $C^*$  values, based on CIE  $L^*a^*b^*$  color coordinates. The properties of the products were characterized by spectrophotometry, X-ray diffraction, scanning electron microscopy (SEM), particle size distribution analysis, and thermal gravimetric-differential thermal analyses. SEM images and particle size distribution analysis revealed that the lightness  $L^*$  value depends on the mass median diameter of the cobalt blue. The Chroma  $C^*$  value is affected by the crystallinity of the aluminum source. These results suggest that the lightness  $L^*$  and Chroma  $C^*$  values of cobalt blue can be controlled by the particle size and crystallinity of the starting materials.

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### 1. Introduction

Pigments are insoluble powders used for coloring, and can be classified as organic and inorganic pigments. Inorganic pigments generally have a low Chroma, low tinting strength, and high hiding power, compared to organic pigments. Inorganic pigments exhibit good weather and chemical resistance, and are generally low cost. Cobalt blue is an inorganic pigment with an intense blue color, and consists mainly of cobalt aluminate ( $\text{CoAl}_2\text{O}_4$ ) spinel. Cobalt blue is chemically and thermally stable, and is widely used to color materials like ceramics, plastics, and fibers [1–6].

Cobalt blue has been synthesized by many methods, including the sol-gel method [2], hydrothermal method [3], and low-temperature combustion method [4,5]. Solid-state reaction is the most common method used to industrially synthesize cobalt blue, because of its simple production flow and high cost-performance. Srisawad et al. [6] synthesized cobalt blue by solid-state reaction, using cobalt chloride hexahydrate and two types of gibbsite with different particle sizes. They reported that decreasing the starting particle size resulted in a lower

crystallization temperature of  $\text{CoAl}_2\text{O}_4$  formation. They also carried out syntheses using various cobalt salts and gibbsite [7], and reported the catalytic activities of the products. Melo et al. [8] synthesized cobalt blue using cobalt oxalate and alumina, with various calcination temperatures. They compared the color tones of the cobalt blue, when it was used as a glaze. While cobalt blue has been synthesized from various materials, there are no reports on the color tone of cobalt blue synthesized from different starting materials. Thus, the relationship between starting material and color tone has not yet been clarified. In the current study, we systematically investigated the influence of different starting materials on the color tone of cobalt blue, synthesized by solid-state reaction.

Because there are no harmful gas generation when sintering, aluminum hydroxide ( $\text{Al}(\text{OH})_3$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) can be used as aluminum sources for synthesizing cobalt blue. Bolt et al. [9] investigated the rate of metal aluminate formation from metal oxide and alumina. They found that  $\gamma\text{-Al}_2\text{O}_3$  reacted with cobalt oxide at a much higher rate than  $\alpha\text{-Al}_2\text{O}_3$ . In addition, heating aluminum hydroxide ( $\text{Al}(\text{OH})_3$ ) at low and high temperature reportedly yields spinel ( $\gamma\text{-Al}_2\text{O}_3$ ) and corundum ( $\alpha\text{-Al}_2\text{O}_3$ ), respectively [10]. These reports suggest that vivid cobalt blue pigment should be synthesized from  $\gamma\text{-Al}_2\text{O}_3$  which has a high reaction rate, or from  $\text{Al}(\text{OH})_3$  which forms  $\gamma\text{-Al}_2\text{O}_3$  during heating. Thus, we selected three sources,  $\text{Al}(\text{OH})_3$ ,  $\gamma\text{-Al}_2\text{O}_3$ , and  $\alpha\text{-Al}_2\text{O}_3$ , and experimentally investigated the influence of aluminum source on the color tone of the resulting cobalt blue.

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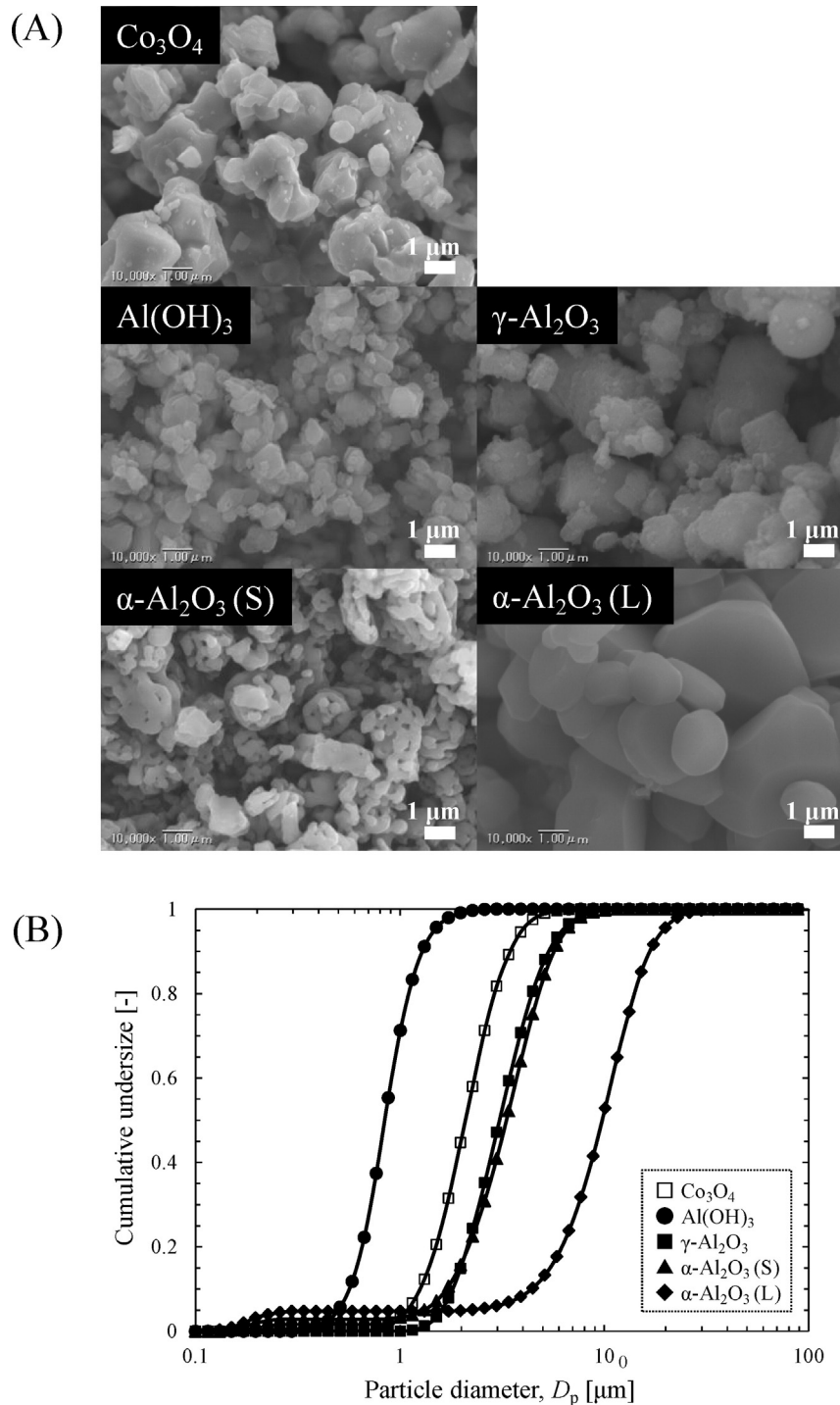


Fig. 1. (A) SEM images showing morphologies, and (B) cumulative size distributions showing particle sizes, of starting materials.

Table 1

Mass median diameters ( $D_{p50}$ ) and geometric standard deviations ( $\sigma_g$ ) of starting materials.

Starting material	$D_{p50}$ [ $\mu\text{m}$ ]	$\sigma_g$ [-]
$\text{Co}_3\text{O}_4$	2.10	1.49
$\text{Al}(\text{OH})_3$	0.84	1.38
$\gamma\text{-Al}_2\text{O}_3$	3.08	1.55
$\alpha\text{-Al}_2\text{O}_3$ (S)	3.32	1.53
$\alpha\text{-Al}_2\text{O}_3$ (L)	9.78	1.53

## 2. Experimental

### 2.1. Materials

Cobalt oxide ( $\text{Co}_3\text{O}_4$ , High Purity Chemicals, Osaka, Japan) was used as the cobalt source. Aluminum hydroxide ( $\text{Al}(\text{OH})_3$ , Wako),  $\gamma$ -alumina ( $\gamma\text{-Al}_2\text{O}_3$ , High Purity Chemicals), and  $\alpha$ -alumina ( $\alpha\text{-Al}_2\text{O}_3$  (S), High Purity Chemicals,  $\alpha\text{-Al}_2\text{O}_3$  (L), Wako, Osaka, Japan) were used as the aluminum source. These reagents were used as received without further purification.

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