Accepted Manuscript

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PII:	S0026-265X(17)30569-6
DOI:	https://doi.org/10.1016/j.microc.2018.01.004
Reference:	MICROC 2996
To appear in:	Microchemical Journal
Received date:	19 June 2017
Revised date:	31 December 2017
Accepted date:	5 January 2018

Please cite this article as: Marc Vermeulen, Koen Janssens, Jana Sanyova, Vanoushe Rahemi, Chris McGlinchey, Karolien De Wael, Assessing the stability of arsenic sulfide pigments and influence of the binding media on their degradation by means of spectroscopic and electrochemical techniques. The address for the corresponding author was captured as affiliation for all authors. Please check if appropriate. Microc(2017), https://doi.org/10.1016/j.microc.2018.01.004

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Assessing the stability of arsenic sulfide pigments and influence of the binding media on their degradation by means of spectroscopic and electrochemical techniques

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Abstract

In this paper, we used the semiconducting and lightfastness properties of synthetic and mineral arsenic sulfide pigments to study their stability by means of electrochemical and microfadometric techniques. A combination of these techniques shows that in the early stage of the degradation process, amorphous arsenic sulfides are more stable than both crystalline forms, while upon longer exposure time, amorphous pigments will fade more than both mineral pigments, making it less suitable. While the stability study was carried out on unbound pigments, the influence of the organic binder on the relative degradation of the arsenic sulfide pigments was investigated through a multianalytical approach on pigment/binder mock-up paint samples. For this purpose, the formation of arsenic trioxide was assessed by micro Fourier transform infrared (μ -FTIR) spectroscopy while the influence of the binder on the formation of sulfates was studied by means of synchrotron radiation X-ray near edge structure (μ -XANES). Both techniques elucidate a higher stability of all pigments in gum arabic while the use of egg yolk as binder leads to the most degradation, most likely due to its sulfur-rich composition. In the context of the degradation of arsenic sulfide pigments, other binders such as animal glue, egg white or linseed oil show an intermediate impact.

Keywords: Arsenic sulfide pigment; Stability; Pigment degradation; µ-FTIR; XANES; Electrochemistry

1. Introduction

Arsenic sulfide pigments such as natural orpiment (As_2S_3 ; golden yellow), natural realgar (As_4S_4 , As_2S_2 or AsS; orange-red) or their synthetic amorphous forms ($g-As_xS_{100-x}$; color varying from bright yellow to red depending on the relative amount of sulfur and arsenic in its composition) have been identified as artists' pigments in works of art from antiquity until the early 20th century [1–6]. Even though only identified in works of art in the past decades, artificial arsenic sulfides have been known for centuries and their existence has been described as early as the 14th century by Cennino Cennini in his *Libro dell'arte* as well as in other painting manuscripts [7,8].

In art objects, natural and artificial arsenic sulfide pigments have been found mixed in various binding media, ranging from polysaccharide-based gum arabic to protein-based or oil-containing binders [2,4,9,10]. One of their main characteristics, extensively described in manuscripts and

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