

Review article

A review of ancient Egyptian pigments and cosmetics

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The pigments used by the ancient Egyptians constitute the most diverse pigment palette of the ancient world. This review discusses the pigments and binders which were used, arranged principally in terms of the colours themselves, namely, the white, green, grey, black, brown, blue, red, orange, and yellow pigments. The chemical identity of these pigments is discussed with some details regarding the artefacts or time periods from which the pigment concerned has been identified. Combinations of pigments were used for creating some colours, especially: greens, pinks, blues, yellows, and a variety of pale hues by mixing primary colours with a white addition, such as calcite. The identification of ancient Egyptian pigments is often aggravated by chemical interactions between pigment and binder media, or between the pigment and environmental pollutants, or both. The nature of the binders and varnishes used in ancient Egypt is briefly discussed. The identification of green pigments from ancient Egypt is often difficult, and some of the recent research concerning the topic is reviewed. In addition to including relevant details from older literature, this review provides a synopsis of recent studies which have appeared since the last major review carried out by Lee and Quirke in 2000.

Keywords: Egyptian pigments, Cosmetics, Binder, Varnish, Pigment alteration, Palette, Color, Pigment history

Introduction

In ancient Egypt, the long period of development of civilization before the Roman period resulted in a rich and complex story as far as pigments and cosmetics are concerned. The Egyptian civilization possessed the richest colour palette of any ancient culture over an extraordinary period of time, some 3500 years or more, and as a result there is a great deal of work still to be done concerning the identity of Egyptian pigments, when they were used, in what context, and on which substrates with what binders or varnishes. In a review paper covering this length of time, there are particular problems inherent in the characterization of the pigments, because some of them have altered considerably since they were first employed. These problems are related to (a) inherent vice: some pigments such as arsenic sulphides are not stable to light and may lose colour, (b) interactions with binding media or substrate: this can lead to alteration of colour, alteration of the chemical compound of the original pigment, darkening due to changes in the binding medium itself due to pigment interaction, or

damage to the substrate, the latter seen on copper pigments on papyrus for example, (c) interactions between the pigment and the environment, particularly salt solutions or other environmental pollutants such as fungi, plants, bird droppings, etc. which may change the chemical constitution of the pigment altogether. Some of the Egyptian pigments are themselves synthetic products, such as Egyptian blue or Egyptian green, which are liable to alteration in a variety of environments. There is insufficient space in this review to discuss all of these aspects in detail, but some account of them will be given, with references where further information can be sought.

Cosmetic materials are of interest in their overlap with the field of conventional pigments and their incorporation of novel or unusual substances, so these are briefly reviewed here as well. All dates for dynastic periods quoted here are taken from the review of Hornung *et al.* (2006), which are given for their first occurrence in the text together with the dynasty or period.

Cosmetic materials for body and hair

The first, and to date the only comprehensive account, was given by Lucas in 1930, which has formed the basis of all subsequent studies on the use of eye

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paint, face paint, oils, and solid fats for face and body ointments. The most common cosmetics were malachite, $\text{CuCO}_3 \cdot \text{Cu}(\text{OH})_2$ and galena, PbS, found in graves as raw fragments, stains on palettes and stones on which they were ground for use (Lucas, 1930). Compact ground lumps have been found as dried-out cakes of kohl or as fine powder. Ashton *et al.* (2000) mention cosmetics being found in small linen or leather bags. Malachite was used from the Badarian period (c. 4400 to 4000 BCE) until at least the Nineteenth Dynasty (1292 to 1187 BCE), while galena, PbS, does not appear before late predynastic times (c. 3100 BCE) and continues into the Coptic period (c. third to eighth centuries CE), while Ashton *et al.* (2000) state that by the middle of the Old Kingdom, it was replaced by carbon black and sticks of kohl.

Of 58 samples examined by Lucas, from a variety of periods, 37 proved to be of galena, PbS, two of cerussite, PbCO_3 or hydrocerussite, $\text{Pb}(\text{OH})_2 \cdot \text{PbCO}_3$, one of tenorite, CuO , five of brown ochre, Fe_2O_3 , one of magnetite, Fe_3O_4 , six of pyrolusite, MnO_2 , one of stibnite, Sb_2S_3 , four of malachite, $\text{CuCO}_3 \cdot \text{Cu}(\text{OH})_2$, and one of chrysocola, $\text{Cu}_2\text{H}_2[\text{Si}_2\text{O}_5](\text{OH})_4$. Lucas (1930, p. 43) mentions that ancient records show that eye paint was obtained from the Asiatics in the Twelfth Dynasty (1991 to 1802 BCE), and in the Nineteenth Dynasty from Koptos, while eye cosmetics were received in the Eighteenth Dynasty (1543 BCE to 1292 BCE) from Naharin in western Asia and from Punt.

By the early Twentieth Dynasty (1187 to 1064 BCE), a typical kohl was of soot, made by burning the qurtum plant (*Carthamus tinctorius*), and applying the moistened powder with a small bone or wooden rod. Face paint for colouring the cheeks is suggested by Lucas (1930, p. 44) as the reason for red ochre finds associated with palettes and stains on stones. Organic components of these cosmetics, such as oils or fats were probably used together with unguents, the latter mentioned by both Theophrastus in 315 BCE (1956) and Pliny in 79 CE (1855). The former states that one was made from a variety of ingredients including cinnamon and myrrh, and another from quince. Pliny writes that Egypt was the best producer of unguents, the most esteemed being a Mendesian unguent consisting of oil of ben (*balanus*), resin and myrrh (*Commiphora molmol*), which in later times contained oil of bitter almonds (*metopium*), olive oil (*omphacium*), cardamoms (*Elettaria*), sweet rush (*Aconus calamus*), honey, wine, myrrh, seed of balsamum, galbanum, and turpentine resin (*Terebinthina resina*).

An important component of any cosmetic is the carrier. According to Pliny, Roman perfumers thought that gum or resin added to a cosmetic would

fix the perfume, and Lucas, who thought that the binder was probably gum, found cosmetic sticks had shrunk away from the container, suggesting that the carrier had dried out.

Henna (*Lawsonia inermis*) was possibly used in ancient Egypt, much as in recent times, as a paste to colour the palms, soles of the feet, nails, and the hair. Lucas (1930, p. 46) records details of the hair of the mummy of Honttimihou, Eighteenth Dynasty, as being of bright reddish colour, and Newberry (1893), identified twigs of henna from the Ptolemaic (323 to 30 BCE) cemetery of Hawara.

Cosmetic containers of marble, alabaster, wood, or reed often contain powders in an exceptional state of preservation. Martinetto *et al.* (1999) identified in New Kingdom cosmetic samples, galena, PbS and cerussite, PbCO_3 as well as laurionite, PbOHCl , and phosgenite, $\text{Pb}_2\text{Cl}_2\text{CO}_3$. Some medical recipes from Pliny (1979) and Dioscorides (1968) refer to the use of lead oxide ground and diluted with salted or carbonated water. The authors replicated this in the laboratory and found either laurionite or phosgenite formed. Deeb *et al.* (2004), also refer to the finding of laurionite and phosgenite with galena cosmetics, and mention that cerussite was also mixed with them. A useful review of the Raman spectroscopy (RS) data for boleite, cumengeite, diaboileite, and phosgenite, previously poorly characterized by RS, has been given by Frost *et al.* (2003b).

Examination of New Kingdom materials from the Louvre collections, dated from 1555 to 1080 BCE, showed that one sample of galena contained 13% anglesite, PbSO_4 , which the authors think comes from the lead ore used to make the cosmetic (Martinetto *et al.*, 1999), this seems unlikely to this reviewer and the anglesite may well be another alteration product. The same research has been published again by Martinetto *et al.* (2001), who examined trace elements in these cosmetic powders, concluding they were foreign imports. Walter *et al.* (1999) found mixtures of galena, cerussite, and phosgenite or galena, cerussite, and laurionite. These are interesting discoveries and deserve further research, especially since the colour of the cosmetic substance would either have been black, in the case of galena, or white in the case of anglesite.

Since the Greco-Roman period, at least (332 BCE–395 CE), mixtures of lead oxide and slaked lime were used to dye the hair, and Walter *et al.* (2006) has found interesting evidence for the biomineralization of galena nanocrystals within the hair, producing a very effective black dye.

There is a paucity of new material concerning cosmetics which future research will undoubtedly address.

Binders and varnishes

Recent comprehensive reviews of Egyptian binding media have been given by Serpico and White (2000a, 2000b, 2001), Newman and Serpico (2000), and Newman and Halpine (2001). Plant gums, animal glues, egg white, beeswax, natural resins, and fats and oils have all been used either as binders or varnishes. Newman and Serpico (2000, p. 483) mention that there are few identifications of glue since the work of Lucas, but new discoveries include an Old Kingdom tomb, the ground as well as pigments on a Romano-Egyptian coffin, painted wooden artefacts and possibly a Nubian stone alabastron. Gum has been identified as the binder on Eighteenth Dynasty, Nineteenth Dynasty, Ptolemaic, and Coptic artefacts, mostly assumed to be gum Arabic, but gum tragacanth was found from a Twenty-first Dynasty coffin, and gums occur in the grounds of several cartonnage fragments, while many samples from the Museum of Fine Arts were gum tragacanth as well (Newman & Halpine, 2001). Stulik *et al.* (1993, p. 63) identified gum as binder from the Tomb of Nefertari, as well as coatings of resin and egg white. The only other identification of egg was from a fourth century CE mummy portrait (Ramer, 1979), suggesting its use was Romano-Egyptian or later. Newman and Halpine (2001, p. 25) found many examples where gum Arabic had been used as the binder, on a variety of substrates, including wall paintings, but they also found glucose in several artefacts, suggesting that a plant nectar or juice had been added to the gum. Animal glue was also commonly used, with a few results showing the use of mixed media with both glue and gum binders. Beeswax has been found as a binder in Fayum portraits, which could possibly be Punic wax (Serpico & White, 2000b). Resin has been found as a binder in grounds or gesso (Wright & Wheals, 1987, p. 209), possibly sandarac resin.

Use of bitumen dates back at least to the Eighteenth Dynasty in mummy varnishes and other objects. Serpico and White (2001) examined yellow and black varnishes from New Kingdom contexts, and showed them to be made from pistacia resin, sometimes called mastic. The black varnish was found to be especially complex, often made from heated (blackened) pistacia resin, used perhaps from the time of Hatshepsut (c.1508 to 1458 BCE) (Serpico & White, 2001), and imports of the resin to Egypt have been found in shipwrecks. Serpico and White (2001) found a whole range of varnishes made from: yellow pistacia resin over a bituminous coating, a mixture of pistacia resin, either yellow or blackened and bitumen, yellow varnish over blackened pistacia resin, a mixture of heated pistacia resin, a *Pinaceae*

pitch and a lipid, or a mixture of unheated pistacia and *Pinaceae* resins, lipid, beeswax, and unknown components.

Bonizzoni *et al.*, (2011) identified beeswax as a coating on a Twenty-sixth Dynasty coffin, with a few indications for gum Arabic, but do not say what the paint binder was. They also found evidence for pistacia resins, such as mastic. Clearly, our knowledge concerning binders and any preferences related to different substrates will continue to develop.

Pigment studies

Egyptian pigments provoked the curiosity of several nineteenth-century scientists, when much research began (Kakoulli, 2009). The Germans (Max Planck Institute) (El Goresy *et al.*, 1986; Schiegl *et al.*, 1989, 1992, Blom-Böer, 1994; Schiegl, 1994; El Goresy & Schiegl, 1996; Riederer, 1997; Schiegl & El Goresy, 2006), the British (British Museum) (Green, 1995, 2001; Middleton, 1999; Lee & Quirke, 2000; Middleton & Humphrey, 2001; Daniels & Leach, 2004; Daniels *et al.*, 2004; Daniels, 2007), the French (Musées de Louvre) (Colinart, 2001, Pagès-Camagna & Colinart, 2003, Pagès-Camagna, 2005, 2010; Pagès-Camagna *et al.*, 2006), and the Japanese (Uda *et al.*, 1993, 2000a, 2000b; Nagashima *et al.*, 1996; Uda, 2004) have carried out major reviews of Egyptian pigments used on artefacts in their collections, which has substantially added to our current knowledge, and work is still in progress. There is a continual stream of papers in the scientific literature since the advent of the twenty-first century which will continue to add to our state of knowledge concerning pigments, when they were used, and their degradation, as well as expanding the methodologies employed for their identification (Uda, 2004; Wagner *et al.*, 2007; Verri, 2009; Zoppi *et al.*, 2010; Bonizzoni *et al.*, 2011).

Black pigments

Charcoal or carbon black

The most common black pigment in Egypt is carbon black. One sample examined by Lucas (1962, p. 340) contained burnt plant material, showing that the black was a charcoal from burnt twigs or other small plants. Other samples were of such fine particle size that Lucas (1962) concluded they were made from soot. Ambers (2004, p. 771) did not find any phosphorus in black samples from the Old Kingdom and concludes that they were probably made from lamp black or soot. Bonizzoni *et al.*, (2011) utilizing many techniques, had trouble in identifying a black pigment. Polarized light microscopy could have differentiated here between a charcoal black or a fine soot, but there is a tendency for the technique not to be employed by scientists outside of the conservation

profession. The black lettering on texts and coffins is usually carbon black in a gum Arabic binder.

Pyrolusite

Pyrolusite, MnO_2 , was found at the Lower Egyptian Predynastic site of Maadi (c. 3000 BCE) possibly used as a pigment or eye paint, and an analysis by Spurrell (1895) identified pyrolusite from a Twelfth Dynasty tomb at Beni Hasan. Pyrolusite has also been found on pottery from the Eighteenth Dynasty (Lee & Quirke, 2000), although in the absence of further examples, there is always doubt about the extent of its use compared with carbon black, and it appears to be rare.

Black organic coatings

Black coatings were made using bitumen or heated pistacia resin for shabtis, figures, and coffins during the New Kingdom (1600 to 1100 BCE) (Serpico & White, 2001), which has been discussed in the section on binders already, and in two Eighteenth Dynasty coffins this had been mixed with bitumen to enhance the colour.

Brown pigments

Browns are not very common but were made from umbers or burnt ochres (Lucas, 1962), with samples dating from the Fourth Dynasty (c. 2613 to 2494 BCE) through to the Late period. Spurrell (1895) identified a brown as being made from a red ochre painted over black, while Lee and Quirke (2000, p. 111) mention that brown was found from Eleventh–Twelfth Dynasty tombs by varnishing over a deep yellow ochre. The project at the Max Planck Institute also found ochre used for brown from the Fifth Dynasty (2494 to 2345 BCE) to the Roman period. Lee and Quirke (2000) found a brown on an Eighteenth Dynasty papyrus to be a mixture of haematite, orpiment, and carbon black, while a red–brown on a Nineteenth Dynasty papyrus was identified as realgar and haematite (Evans *et al.*, 1980).

Blue pigments

Egyptian blue

The predominant pigment was Egyptian blue, used from about the Fourth Dynasty (c. 2639 to 2504 BCE) through to the Roman period (332 BCE to 395 CE) (Riederer, 1997, p. 23). To make the pigment, scrap copper or bronze was mixed with crushed quartz sand, lime, and a soda flux and heated to about 850–950°C (Tite *et al.*, 1987), producing a mixture of cuprorivaite, $\text{CaCuSi}_4\text{O}_{10}$, silica phases such as quartz, tridymite, or cristobalite, a copper alkali-bearing silicate glass, and occasionally incorporating a copper oxide phase, such as cuprite, Cu_2O , or tenorite, CuO , and some unreacted bronze fragments (Schiegl *et al.*, 1989).

Egyptian blue as a pigment preparation usually comprises small crystals of cuprorivaite and a glassy phase, a copper-containing wollastonite, $(\text{Ca,Cu})_3(\text{Si}_3\text{O}_9)$, which tends to be sensitive to various decay processes (Tite *et al.*, 1987; Riederer, 1997). The crystal structure of Egyptian blue was elucidated by Ford and Hitchman in 1979, and was further refined from a single crystal of Roman origin by Mirti *et al.* (1995), and shown to be a four-coordinate, planar ligand structure.

Egyptian blue was the most successful synthetic blue pigment in history, lasting for 3000 years, and was employed on wood, papyri, stone, cartonnage, stucco, plaster, and metallic objects (Tite *et al.*, 1987). Its use even lasted until the eleventh century CE, discovered from an altarpiece in the Church of Sant Pere, Terrassa, Catalonia (Lluveras *et al.*, 2010) which is inexplicable outside of the re-use of an ancient Roman pigment sample, perhaps derived from excavated pigment or balls of blue frit, as there is no evidence for production at this late date. Yet more strange occurrences of Egyptian blue have recently come to light, such as a *Last Judgement* by Nicolaus and Joannes, in the Vatican Museum dated to the twelfth century (Pozza *et al.*, 2000) and in a painting by Giovanni Battista Benvenuto (ca. 1480–1525) from 1524 (Bredal-Jørgensen *et al.*, 2011).

Trace elements such as lead and tin are often found in Egyptian blue. For example, Hussein (2012) mentions that significant amounts of lead were found in Egyptian blue from the Ptolemaic temple of Hathor, Thebes, located in the necropolis of Deir el Medina, derived from the re-use of leaded bronze scrap. The identification of Egyptian blue is fairly straightforward and most microsamples can be characterized by polarized light microscopy, as Egyptian blue is easily distinguished from other common pigments such as azurite, indigo, smalt, or ultramarine. A recent and valuable non-sampling strategy to characterize and visualize the pigment has proved useful, employing photo-induced luminescence (Verri, 2009). When irradiated with light, the pigment emits infrared (IR) radiation and luminescence can be recorded on a camera which is sensitive to IR radiation. Even single Egyptian blue particles can be imaged using this technique as they appear as ‘glowing white’ in the IR image.

Degradation of Egyptian blue

Egyptian blue is liable to undergo different types of degradation. One of these effects transforms the blue colour of the original to a dark green or green–blue, because of alteration to a mixture consisting predominantly of the basic copper chlorides with some malachite $\text{CuCO}_3\text{Cu(OH)}_2$. The majority of the basic

copper chlorides are isomers of trihydroxychloride $\text{Cu}_2(\text{OH})_3\text{Cl}$ (Scott, 2002).

The identity of the degradation products resulting from the weathering of Egyptian blue pigments was investigated by Schiegl *et al.* (1989, 1992), who began their studies with the basic copper chloride greens (the copper trihydroxychlorides) found in tombs of the Old Kingdom and the Middle Kingdom, such as those of Nefer (Fifth Dynasty, c. 2494 to 2345 BCE) at Saqqara, and Sefrekhu (Sixth Dynasty, 2345 to 2181 BCE) at El Hawawish. El Goresy *et al.*, (1986) originally thought these were synthetically prepared green pigments, but this was an erroneous conclusion. They were not the original pigments at all, but degradation products. The original pigments must have been Egyptian blue, azurite, or malachite.

According to Schiegl *et al.* (1992, p. 847), this degradation can result in Egyptian blue becoming a more intense blue as the greenish glass phase is lost, although as the basic copper chlorides build up, the consequence is that a pigment layer originally blue becomes altered to green. Egyptian blue may also darken, which could be due to a number of causes. One possibility is alteration to tenorite or cuprite. One of the earliest reports of the blackening phenomenon in Egyptian tombs which could be related to tenorite formation was by Lucas (1934), who studied the colour change of trefoil marks on a painting from the tomb of Tutankhamun. Decomposition of Egyptian blue to a combination of cuprite mixed with one of the copper trihydroxychlorides would also give a visual appearance of brown or greenish-brown. There is another explanation however, Daniels *et al.* (2004) investigated the brownish-green or black discoloured paint found on several Egyptian coffins in the British Museum collections. They concluded that surface dirt accumulation or the ageing of gum Arabic to a brown, discoloured product may also be a cause of colour change, resulting in a dark, greenish appearance to the Egyptian blue in some cases.

Discoloured varnishes or coatings may also be a contributory factor in the alteration of blue to green. The common use of pistacia resin as a yellow varnish, especially in its heated form, if used over an Egyptian blue, creates a green-coloured visual effect.

Optical blue

In the First Intermediate period tomb-chapels (c. 2181 to 2055 BCE, approximately the Seventh–Eleventh Dynasty), the Max Planck project found blue–grey pigments of calcite and carbon black, and of an iron–titanium compound mixed with calcium carbonate (Blom-Böer, 1994, p. 73). The blue–grey colour, effectively a way to make a blue tint without any blue

pigment *per se*, was discovered by most cultures of the ancient world and is referred to as an ‘optical blue’ (Howard, 2003).

Cobalt blue

A cobalt-based blue, $\text{CoO} \cdot \text{Al}_2\text{O}_3$, used for post-fired painting on pottery (Riederer, 1974), made an appearance during the Amarna period around 1370 BCE, but lacks any further identification. Abe *et al.* (2012) state that the inception of cobalt blue as a colourant can be dated to the Eighteenth Dynasty (1479 to 1424 BCE), produced from cobalt alum found in the Western Desert. Much of this was used in glass making but pigment derived from this alum was produced according to Arnold and Bourriau (1993) until the Twentieth Dynasty, with some variations (Abe *et al.*, 2009, 2012). This disappeared at the end of the New Kingdom, and did not re-appear until the Late period, but these late examples (mid seventh century BCE) were different in composition and not derived from cobalt alum (Shortland *et al.*, 2006).

Azurite

Azurite, $\text{Cu}(\text{OH})_2 \cdot 2\text{CuCO}_3$, was rarely employed as a blue pigment. However, it was identified by Spurrell in 1895, on a shell used as a palette in a Fourth Dynasty context (ca. 2613 to 2494 BCE) at Meidum, and in a number of Eighteenth Dynasty wall paintings. Research at the Metropolitan Museum of Art by Heywood (2001b) found examples of both azurite and lapis lazuli, dated on stylistic grounds to the early Eighteenth Dynasty, specifically between the reign of Ahmose I and the co-regency of Hatshepsut and Thutmose III (1550 to 1458 BCE). Azurite has long been assumed to have been used as a pigment in ancient Egypt, but because the identification by Spurrell in 1895 was so long ago, doubt has been expressed concerning its veracity. A lump of azurite was identified among a group of raw pigment samples from Tel-el-Amarna that were excavated by Flinders Petrie in 1892, currently housed in the Manchester Museum of Egyptology (David *et al.*, 2001). The weight of evidence now shows that azurite was used as a pigment from at least the Fourth to Eighteenth Dynasty. Azurite is liable to undergo slow deterioration with loss of carbon dioxide and conversion to malachite, and in some circumstances it can transform to a black tenorite, CuO . Further examples of its use may be found in future studies, although given its comparative rarity, Egyptian blue was always the most commonly used blue.

Ultramarine

Traces of poorly adhered natural ultramarine pigment were found in the incised lines of Queen Hatshepsut's broad collar (Heywood, 2009). Natural ultramarine

is made by grinding lapis lazuli, a stone composed primarily of lazurite, $(\text{Na,Ca})_8[(\text{S,Cl,SO}_4,\text{OH})_2(\text{Al}_6\text{Si}_6\text{O}_{24})]$, calcite, CaCO_3 , and pyrite, FeS_2 . Lapis lazuli was used in Egypt as early as the Predynastic period for beads, inlay, amulets, cylinder seals, small vessels, and scarabs. Although Egyptian texts indicate that *lapis* was purchased from trading stations in western Asia, and from Meroe and Punt, to the south of Egypt, all of the lapis in ancient Egypt is thought to have originated from the quarries in Afghanistan. The use of natural ultramarine from the Eighteenth Dynasty is the earliest recorded use of the pigment in the world.

White pigments

Calcite, gypsum, and anhydrite

The most available white pigments were calcite, CaCO_3 , gypsum, $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, and anhydrite, CaSO_4 (Eastaugh *et al.*, 2004). All three are very common, either as the grounds of paintings to be made on cartonnage or wood, or as pigments used for the final paintings themselves. Natural calcium sulphates are widely available in Egypt, and those examined from the British Museum by Ambers (2004, p. 771) were a mixture of gypsum and anhydrite. If gypsum is heated to around 90°C , it converts to hemihydrate, $2\text{CaSO}_4 \cdot \text{H}_2\text{O}$, while heating to 200°C produces anhydrite, CaSO_4 . In some grounds, these dehydrated phases are found, showing that the gypsum has been heated. Some natural deposits are also a mixture of anhydrite and gypsum. Calcite and gypsum were used extensively, often with gum Arabic or animal glue as binder. Blom-Böer (1994) recorded both calcite and gypsum from the Fifth Dynasty to the Roman period. Moussa *et al.* (2009) paid special attention to the composition of the grounds used on the wall paintings of Al Qurna, some of which are Coptic, and include the Egyptian temples of Madinet Habu and Kha'Em Het. Some of the plasters used had been pigmented a yellow colour. For example, plasters from the tomb of Kha'Em Het consisted of anhydrite, calcite, goethite, orpiment, and quartz, while in the tomb of Amen Em Habit, the plaster consisted of anhydrite, calcite, quartz, jarosite, plagioclase, and clay minerals. These detailed results reveal that grounds were deliberately prepared from pigment mixtures in order to create the desired base colour for the painted scenes. Chu *et al.* (2008) have been able to differentiate between calcites which have formed geologically, archaeological plasters, modern plasters, and those experimentally prepared using IR spectroscopy. Some of these are hard to distinguish, but anthropogenic calcites have different spectra from geogenic-derived calcites, a subject of some interest for ancient Egyptian plaster studies as calcite is often found.

Quartz

Quartz, SiO_2 , was found in ground layers mixed with calcite or gypsum, in several cartonnage fragments, dating from third century BCE to first century CE, in the Petrie Museum (Scott *et al.*, 2009). This strengthens the ground layer, as it is not necessarily present in the paint layers of calcite itself. Its discovery here predates the use of ground glass or quartz in Renaissance paintings (Spring, 2012).

Tridymite

Dodd *et al.* (2009) found tridymite, $\beta\text{-SiO}_2$, a high-temperature modification of quartz, to have been used over calcite on a shabti figurine. This is the only known occurrence to date of the use of tridymite, one possible source being the heated quartz that was used for the manufacture of Egyptian blue. Scott *et al.* (2009) found moganite, yet another quartz polymorph, used in a cartonnage of the third–first centuries BCE. Moganite is a constituent of many siliceous rocks, and may be an entirely adventitious component.

Huntite

Analyses of whites from Theban tombs of the Twelfth Dynasty (1991 to 1802 BCE) and from the Eighteenth to Twentieth Dynasty (1543 to 1064 BCE) (Riederer, 1974) revealed huntite, $\text{Mg}_3\text{Ca}(\text{CO}_3)_4$, a mineral unknown to modern science until 1953, which provides a brighter white than calcite. Subsequently, huntite has been found on objects dating from the Old Kingdom (Ambers, 2004, p. 771), for example, on a Fourth Dynasty tomb (c. 2600 BCE), probably for artistic effect rather than elite purposes in this case. Huntite use continues through to the Roman period, although its use peaked during the Middle Kingdom (Heywood, 2001a), and then declined during the Late and Ptolemaic periods. Uda *et al.* (1993) found huntite on wall paintings of the Eighteenth Dynasty. The discovery of 1 kg of pure huntite in an Italian shipwreck dated to the second–third century CE is proof of long-distance trade in the mineral (Barbieri *et al.*, 1974). El Goresy *et al.* (1986) originally suggested that huntite was reserved for royal use, however, more recent studies have begun to show that the pigment was more widely used and is thought to be the predominant pigment used during the New Kingdom (Heywood, 2001a). Huntite has also been used as a pigment underlayer covered with red ochre (McCarthy, 2001) in one New Kingdom example, and under red and yellow ochres from the tomb of Nebamun, also from the New Kingdom (Ambers, 2004). Huntite is the major white pigment (Chiari, 2014) used in the tomb of King Tutankhamen (reigned 1332 to 1323 BCE), and has

been found on a Ptolemaic polychrome sculpture of Osiris (Casadio *et al.*, 2005).

Clearly, this is not the last word on the interesting story of the prevalence of the use of huntite, which requires further research.

Lead white

In the Graeco-Roman period, lead white begins to be used for the first time, and subsequently becomes common. Scott *et al.* (2004) found that it was used on a cartonnage broad collar, dated to between 512 and 351 BCE. Edwards *et al.* (2004) examined several pigments from the Seventeenth Dynasty to the Graeco-Roman period and found Ptolemaic coffins, where hydrocerussite had been used, especially in pigment mixtures; they also found whites to be a mixture of calcite and hydrocerussite, or calcite and anhydrite, both from Twenty-first Dynasty contexts. Aragonite was also found and probably derives from sea shells or white coral and has not been reported before.

Alteration products of white pigments

Lead white may blacken due to interaction with environmental pollutants or adjacent pigments containing sulphur. Biological interactions create the commonly seen calcium oxalates, whewellite, and weddellite. Bonizzoni *et al.* (2011) showed that these had formed from alteration of the ground layer, whose composition could not be identified due to lack of a sample. The common degradation products of lead white are plattnerite, PbO_2 , or galena, PbS .

Pigment mixtures for grey

This colour is usually obtained by a mixture of gypsum or calcite with ground charcoal. Blom-Böer (1994, p. 74) found that this was used from the Fifth to the Twentieth Dynasty, and Scott (2014a) found it on Saite period coffins in the San Diego Museum of Man. Ambers (2004, p. 771) identified one grey on an Old Kingdom wall painting as a mixture of gypsum and carbon, and on another, as a mixture of whites, black, and yellows. The exact combination varies: in one, gypsum, anhydrite, and huntite were used in combination with goethite and carbon, while in another, anhydrite had been mixed with natrojarosite and carbon. These may represent different workshop practices or what kind of colour was available to produce the desired grey tone.

On one of several Ptolemaic and Roman period papyrus fragments from the Fayum and elsewhere, a grey pigment was identified as kaolinite mixed with quartz (Blom-Böer, 1994). This combination is not unknown, but is highly unusual as a grey pigment and there is a paucity of other examples. Coal could be present as a possible contributing component in

some of these cases, as on X-ray diffraction (XRD) it is mostly hidden by the major peak for quartz.

Orange Pigments

Realgar and pararealgar

Lucas (1962, p. 346) identified orange as red painted over yellow, or as a mixture of red and yellow. Lucas did not specify whether red ochre, Fe_2O_3 , and yellow ochre, $FeOOH$, were meant, but this cannot be assumed as work at the British Museum found orpiment, As_2S_3 , or pararealgar, AsS , with red ochre in one Ramesside papyrus (Daniels & Leach, 2004). Vandenbeeke *et al.* (2001, 2009) found realgar in two Ptolemaic masks from El-Hiba, while David *et al.*, (2001) identified a pigment from Tel-el-Amarna, dated to about 1340 BCE as realgar and pararealgar. Daniels and Leach (2004) found one pigment from a Thirtieth Dynasty papyrus (380 to 343 BCE) to be pararealgar and another to be realgar. In at least one case, red–orange realgar-containing ink had now turned to yellow pararealgar. The chemical degradation of realgar pigments is a complex affair. According to Macchia *et al.* (2013), realgar is a natural α - As_4S_4/β - As_4S_4 and synthetic χ - As_4S_4 or $As_4S_4[II]$ mineral used as both pigment and cosmetic in ancient Egypt. The beta form is actually a high-temperature phase which has not been reported from art objects. χ - As_4S_4 is seen as an intermediate phase between alpha realgar and pararealgar, with the latter being a stable polymorph at room temperature (Daniels & Leach, 2004).

There are possible confusions between realgar, As_4S_4 , and orpiment, As_2S_3 , because of the transformations which realgar can undergo. It used to be thought that realgar underwent photogradation to orpiment and arsenolite, As_2O_3 , the latter often producing twinkling light effects and altering the colour of the orange pigment to an off-white. This conclusion was regarded as reasonable as the colour of the pigment often changed overall from orange to yellow, because of alteration to the intermediate form χ - As_4S_4 before reaching total photodegradation to arsenolite. However, Douglass and Shing (1992) showed that the realgar was transforming to pararealgar, which can be regarded as an intermediate product on the eventual route to arsenolite (Wallert, 1984). Sulphur is then released from As_4S_5 , a mineral called uzonite, which was described by Bindi *et al.* in 2003, and which reverts to the stoichiometry of AsS as the excess sulphur is released.

Could pararealgar have been deliberately prepared as a pigment by the ancient Egyptians? It is not clear that was ever used, especially since Daniels and Leach (2004) noted that the double bands of orange and yellow, employed on many copies of the Book of the Dead, mostly comprised realgar and orpiment.

Pagès-Camagna *et al.* (2010, p. 667) examined a number of wall paintings and identified arsenolite and calcium arsenate $\text{Ca}_2\text{As}_2\text{O}_7$. Arsenolite mixed with other white compounds was found in the underlayer below yellow or red tints in the tomb of Nefertari (Stulik *et al.*, 1993), while realgar, orpiment, and arsenolite were identified from the tomb of Amenophis III, which according to Pagès-Camagna *et al.* (2010, p. 668) could be deliberate admixtures.

Yellow pigments

Gold

Gold leaf was extensively employed in all periods. Bonizzoni *et al.* (2011) found gilding on a Twenty-sixth Dynasty coffin, and suggested the leaf had been laid over a white preparation layer, covered with Egyptian blue and finally orpiment, a sophisticated approach to preparation of the underlayer for gilding. The gold used was quite pure with 0.47% silver content. Gilding was used in some illustrated funerary papyri, both from the New Kingdom and the Late period (Lee & Quirke, 2000, p. 116), and on a Book of the Dead of Tjenena, of the New Kingdom. Oddy (1993) reports gilding over a calcite ground used as preparatory layer on a statuette of Isis, from the Late Dynastic period of about 550 BCE and the head of a statue of Hathor from the Late New Kingdom of about 1000 BC.

The binders used to adhere the gold have not yet been investigated. Although historically egg was often employed, there is no evidence that the ancient Egyptians made much use of it in gilding on stone, wood, or other substrates. In addition, as shown above, a preparatory gesso layer was sometimes used to which gold leaf or foil could then be applied.

Goethite

The most common yellow is goethite, yellow ochre, $\alpha\text{-FeOOH}$, identified by the Max Planck project from the Fifth Dynasty to the Roman period (Blom-Böer, 1994). Goethite is quite stable in all Egyptian binding media, such as gum Arabic, and no alteration products have been determined from its examination during numerous studies. There are many identifications, for example, Ambers (2004, p. 771) found goethite on two Old Kingdom limestone sculptures. According to Colinart (2001), Egyptian yellow ochre may contain trace amounts of jarosite or natrojarosite.

Orpiment

Orpiment, As_2S_3 , was also used extensively, and identified in a pure state by the Max Planck project on Eighteenth and Nineteenth Dynasty sarcophagi of kings (Blom-Böer, 1994) and on the walls of the tomb of Thutmose IV (c. 1400 BCE). Edwards *et al.* (2004) mention that it has been found from Twelfth

Dynasty contexts. Mixtures of orpiment and yellow ochre (Blom-Böer, 1994) were used in the New Kingdom (1570 to 1069 BCE), while Scott *et al.* (2009) showed that goethite and orpiment continued in use through the Graeco-Roman period. Uda *et al.* (1993) found goethite mixed with degraded orpiment on Eighteenth Dynasty wall paintings and from the Malqata Palace (Uda *et al.*, 2000a). In some cases, different shades of colour were obtained by mixing yellow with Egyptian blue, and in one example, an upper layer of Egyptian blue was painted over a layer of orpiment. The source for the orpiment is not known, but the pigment was found in the cargo of the Ulu Burun shipwreck off the southern Anatolian coast dating to the late fourteenth century BCE (Moorey, 1994), in which realgar was also found. A fragment of a limestone grinding-bowl reported by Weatherhead and Buckley (1989) and Weatherhead (1995) from an early industrial area to the south of central Amarna was found to contain coarsely ground orpiment. Orpiment must remain coarsely ground when used as a pigment, otherwise the full visual effect of the colour is diminished. This is advantageous for polarized light microscopy of orpiment pigments, as the strongly birefringent particles can easily be identified. These often appear partially degraded to a dark alteration product, either of the pigment itself or of contiguous pigments or media degraded by the sulphur released from the decomposing orpiment. Keune and Boon (2011) maintain that the final degradation product, arsenic oxide, As_2O_3 , can be transported across the binding media and structural layers due to water-induced deterioration of paintings, resulting in arsenic being detected in many zones where the pigment had never been applied, this could impact the identification of arsenolite as a deliberate additional pigment. An unusual pattern of flaking paint involving orpiment on a Ptolemaic mummy cartonnage has been reported by Genbrugge (2011).

Organic yellows

Lucas (1962, p. 351) mentions the detection of a vegetable yellow, which could not be identified. Scott *et al.* (2009) also had difficulties with yellow and three examples from first century CE cartonnage fragments were probably of an organic yellow which could not be identified further.

Jarosites

Noll (1981) identified jarosite, $\text{KFe}_3(\text{OH})_6(\text{SO}_4)_2$, from Eleventh Dynasty pottery from el-Tarif. Additional examples were found from tomb walls from the Fifth to Twelfth Dynasties, on several early Middle Kingdom coffins (2000 to 1700 BCE), as well as on stone artefacts from the Fourth and Fifth

Dynasty to the Ptolemaic period (305 to 30 BCE) (Middleton, 1999; Colinart, 2001; Middleton & Humphrey, 2001), although natrojarosite, $\text{NaFe}_3(\text{OH})_6(\text{SO}_4)_2$, seems to be the commonly identified variety. It was originally suggested that jarosite was (yet another) deterioration product rather than a pigment (Schiegl *et al.*, 1992), but this has now been discounted and the jarosites are now recognized as another class of pigments used by the ancient Egyptians. Ambers (2004, p. 771) found natrojarosite on three different limestone sculptures from the Fifth to Sixth Dynasty.

Painting equipment from Karnak included jarosite and there is a possible source of the mineral in the western desert at Aswan (Pagès-Camagna *et al.*, 2010). The use of jarosite and yellow ochre in different areas of a painted stela of Nefertibet, c. 2590 to 2565 BCE, where jarosite was used for the yellow dress and yellow ochre for the skin of Nefertibet, testifies to the sophistication of its use in Egyptian art at an early date (Colinart, 2001).

Massicot

Massicot, PbO , was identified by Laurie (1913) in a pigment palette dating to 400 BCE, but there have been no further reports of its use.

Red pigments

Haematite

Haematite, Fe_2O_3 , or red ochre, is the most commonly used red in all periods. The Max Planck project found that it was used from the Fifth Dynasty to the Roman period. Ambers (2004) found that it was used on Old Kingdom limestone sculptures from about 2300 BCE, Uda *et al.* (1993) found that it was on wall paintings of the Eighteenth Dynasty, and probably in varieties of both relatively pure haematite and red ochre. Uda *et al.* (2000a) also found red ochre on the walls of an Eighteenth Dynasty palace and compared the particles found with those obtained from heating yellow ochre. They found distinctly different particle morphologies, showing that the red ochre used was naturally occurring, not derived from heated goethite (Uda *et al.*, 2000a, p. 760).

Realgar

Realgar, As_2S_3 , has been reported from Eighteenth Dynasty tombs at Thebes, the contents of a receptacle in the tomb of Kheruef, and from a tomb wall of Thutmose IV. Degraded realgar, which tends to alter to pararealgar, as noted above, has been found at Amarna (Weatherhead, 1995, p. 395). Lee and Quirke (2000) found mostly red ochre, but on one Nineteenth Dynasty papyrus, pararealgar with orpiment and a trace of haematite. Realgar became more

common from the New Kingdom onwards (Middleton & Humphrey, 2001).

Cinnabar

Cinnabar provides an intense red but there is little evidence for its use until the Late or Roman period (c. 30 BCE–500 CE). However, Quirke (1993) states that it has been identified as a pigment on a papyrus of the Late period, from 747 to 332 BC, and its use has been documented since the middle of the Twelfth Dynasty (Fitzhugh, 1997). Bonizzoni *et al.* (2011) examined a sarcophagus of the Twenty-sixth Dynasty, about 664 to 525 BCE, made for Shepsesptah, a priest of Sechem, where cinnabar was used to paint a decorative flower element, but examples of its identification are still rare.

Minium

Minium (red lead oxide), Pb_3O_4 , and to a certain extent, cinnabar, HgS , have mostly been associated with the Ptolemaic and Roman periods (Green, 2001; Middleton & Humphrey, 2001; Scott *et al.*, 2004). However, the use of red lead has been documented in Egypt as early as the Second Intermediate period, incorporated into medicinal balms for stomach ailments (Bryan, 1930, p. 54). Its use as a pigment was scarce prior to the Roman period, when it was used to paint linen cloths for funerary shrouds (Flinders Petrie, 1911; Wisseman, 2003).

Investigation of a group of mummies from the early Roman period, 31 BCE–first century CE (Walton & Trentleman, 2009), known as the ‘red shroud mummies’ showed all examples to be painted entirely in red lead, with minor phase inclusions of lead–tin yellow, Pb_2SnO_4 , normally produced by heating mixtures of lead and tin oxides to between 650 and 800°C (Kühn, 1993b). Evidence shows that silver production from Rio Tinto, Spain, produced litharge from cupellation of jarosites. Heated later to below 500°C to make the red lead, litharge was the source of this red lead pigment, which was then exported to Egypt (Walton & Trentleman *et al.*, 2009). Lead–tin oxide, or lead–tin yellow, as the pigment is commonly called, was not known to have been used as a pigment before the fourteenth century CE (Kühn, 1993b), and its occurrence in this Egyptian context is probably adventitious.

Pink pigments

Organic pinks

An unusually bright pink on a Third Intermediate period papyrus was found to be an organic pink over gypsum (Lee & Quirke, 2000, p. 113), probably madder. Scott *et al.* (2009) found high levels of glucose in a Graeco-Roman period pink, which also

suggested the use of madder root (*Rubia tinctorum* or *Rubia peregrina*). Other mentions are rare.

Pigment mixtures for pink

Pink is usually obtained with a mixture of pigments such as red ochre and calcite, or ochre and gypsum. For example, Spurrell (1895) found pinks from the Old and New Kingdoms to be a mixture of red ochre and gypsum. For the Hellenistic and Roman periods, Russell (1893–1894) identified madder over gypsum, Uda *et al.* (1993) found huntite mixed with red ochre, and from Nineteenth Dynasty contexts, Uda *et al.* (2000a) found a mixture of huntite and haematite. Calza *et al.* (2007a, 2007b, 2008) found pink to be made from realgar, red ochre, and gypsum or chalk, on a Roman-Egyptian coffin of the first century BCE, but only X-ray fluorescence (XRF) was used so the results here are dubious. Lee and Quirke (2000) found vermilion as a pink on a funerary papyrus from the Ptolemaic period, and during this period, the use of pink becomes common.

Green pigments

Green pigments have created considerable problems in terms of identification and characterization, and while significant progress in understanding the nature of these problems has been made over the past two decades, many papers have been published where the identity of the green pigments is still a cause for concern, and some recent papers expand the possible techniques of identification (Frost *et al.*, 2003a).

Egyptian green

For a long time, Egyptian green was considerably less well-known than its blue counterpart. This is partly because many of the seminal publications on Egyptian green were in German (Noll & Hangst, 1975; Noll, 1979; 1980, 1981; Jaksch, 1985; Schiegl *et al.*, 1989; Schiegl, 1994), and because of variations in colour which extended the blue range into turquoise or blue–green, confusing the issue of a separate but distinct fabrication technology. Pagès-Camagna and Colinart (2003), Pagès-Camagna (2005), and Pagès-Camagna *et al.* (2006) give directions for the synthesis of Egyptian blue and green. The principal phase responsible for the green colour is a silica-rich, copper-containing glass, according to Pagès-Camagna and Colinart (2003) and Pagès-Camagna *et al.*, (2006). The published work from 2006 drew a critical response from Schiegl and El Goresy (2006), in which the authors criticize Pagès-Camagna and Colinart for misrepresenting their work as stating that green frit is a misfired Egyptian blue. They also maintain that they had previously published the successive use of scrap metals derived from arsenical copper, tin bronze, and lead-bearing bronze in the

production of green frit and Egyptian blue. As a result, the introduction of tin bronze could be dated to the reign of Thutmosis III in the Eighteenth Dynasty and leaded bronzes to the reign of Sethos II or Tauseret-Setnakht in the Nineteenth, entirely on the basis of their use in pigment preparation. In the authors' reply to these criticisms (Pagès-Camagna & Colinart, 2006), the researchers state that they disagree with the German work which suggested that the green pigment owes its colour to a copper-bearing wollastonite. Further work on the fabrication technology has been published by Hatton *et al.* (2008).

Pagès-Camagna and Colinart (2006) found evidence for the use of Egyptian green on wood, stone, and pottery from as early as the First Intermediate period, around 2200 BCE until the Third Intermediate period, around 1069 BCE. But interestingly, this particular green pigment has not been found outside of Egyptian territory, unlike Egyptian blue, which was widely dispersed and used by many cultures and which even made its way up to the Arctic circle in the far north of Norway. There it was used as a pigment on a shield found in the graves of the Bo people from around 250 AD (Rosenquist, 1959). Recent identifications include the work of Moussa *et al.* (2009), who found Egyptian green from the temple of Madinet Habu, a mortuary temple of Ramesses III, about 1150 BCE.

Malachite

There was a problem with the recognition that malachite was quite a common pigment in ancient Egypt, and even as late as 2000, Ashton *et al.* write that '...malachite was very occasionally used as a green pigment...' but this aberration has been corrected since then. Pagès-Camagna (2010) has found it on several stone and wooden artefacts from the Old Kingdom onwards, Scott (2014a) has found malachite on several Twenty-sixth Dynasty coffin cases in the collections of the Museum of Man, San Diego, and Lee and Quirke (2000, p. 110, 112) identified it from a First Intermediate period or early Middle Kingdom wooden coffin of Khuit in the collections of the British Museum. Pigment samples originally excavated by Sir Flinders Petrie in the 1890s, from an Eighteenth Dynasty, New Kingdom context at the time of King Akhenaten (c. 1340 BCE), were examined using Raman spectroscopy and malachite was identified (David *et al.*, 2001).

No instances of the use of malachite were found by Blom-Böer (1994, pp. 74–5) on stone artefacts from the Fifth Dynasty through to the Roman period. However, she reported a copper-containing vitreous pigment, presumably Egyptian green, in use from the Fifth Dynasty to the Twelfth, whose colour ranged from blue to bright green. Analyses showed the

presence of basic copper chloride (presumably atacamite, although the isomer is not defined) and malachite (Blom-Böer, 1994, p. 65). Lee and Quirke (2000, p. 112) found a degraded malachite green on one Thirtieth Dynasty or early Ptolemaic funerary papyrus in the British Museum.

Degradation problems

Malachite may be present as an original pigment or may have formed as a result of the degradation of Egyptian blue or Egyptian green, or azurite. But malachite itself is liable to undergo alteration, which obscures the extent to which it was probably used. This is because the highly saturated nature of salt solutions in the Egyptian environment tends to decompose malachite and other copper-containing pigments, and alter it to one of the copper trihydroxychlorides, which are also green in colour.

Copper-containing pigments are well known as a source of the catalyzed degradation of cellulose-based materials, papyrus, or protein-based materials, such as parchment (Banik *et al.*, 1982; Banik, 1989). Middleton (1999) and Middleton and Humphrey (2001) mention green and blue pigments on stone fragments from the Middle Kingdom site of El-Bersheh which appeared to be mixtures of atacamite and malachite.

Malachite has been reported to be the result of the decay of blue glass producing atacamite and malachite (Schiegl *et al.*, 1992). A small blue area on the throne on the first sheet of the British Museum's *Papyrus of Penmaat*, from the tenth century BCE, now appears a patchy dark green. Basic copper chlorides have been identified in several degraded green pigments on other Egyptian art in the collections of the British Museum (Green, 2001, p. 45). Atacamite is the most common of the copper trihydroxychlorides, $\text{Cu}(\text{OH})_3\text{CuCl}_2$, but there are at least four isomers, all of which have been identified from Egyptian pigment and bronze corrosion contexts (Scott, 2002, 2014a, 2014b; Martens *et al.*, 2003). In arid zones or in contexts in which crystalline compounds can dry out preferentially and rather rapidly, which prevents one form of the isomer recrystallizing as another (Church, 1865), botallackite can form, and this has indeed been identified from Egyptian contexts (Frondel, 1950).

From a Theban Nineteenth Dynasty (c. 1295 to 1186 BCE), papyrus *Book of the Dead*, Olsson *et al.* (2001) found that a copper map of the distribution of the green pigment area obtained by proton-induced X-ray emission coincided with a large hole in the papyrus, where copper and arsenic were the major constituents. The presence of copper was attributed to neutral verdigris, since this pigment is known to degrade papyrus, parchment, and paper, but this

identification is unsecure and lacks confirmation. Green (1995) examined several papyri in the British Museum. In one case, this green was found to comprise Egyptian blue with atacamite, and in an eleventh century BCE papyrus (EA 10472.4), a mixture of Egyptian blue with a compound similar to malachite. When found on papyrus, Green (1995, p. 88) states that atacamite appears to cause the papyrus to be particularly fragile and brittle; additional evidence that the inference for the presence of verdigris is unsubstantiated.

Chrysocolla

The extent to which the naturally occurring copper green pigment, chrysocolla, $(\text{Cu},\text{Al})_2\text{H}_2(\text{Si}_2\text{O}_5)(\text{OH})_{4-n}(\text{H}_2\text{O})_n$, was used in ancient Egypt remains obscure. The mineral itself was well known, and was even described by Theophrastus in 315 BCE as being used for gold solder (Theophrastus, 1956). Of three green pigment samples from the Eighteenth Dynasty Workmen's Village at Amarna, two were identified as chrysocolla, while the third was a green frit (Weatherhead & Buckley, 1989). The occurrence of these examples of chrysocolla from pigment samples, rather than from objects themselves, tends to suggest that the use of chrysocolla could be more common than previously thought. Spurrell (1895) found the pigment on Twelfth Dynasty tomb walls at El-Bersheh.

Rouchon *et al.* (1990) claim to have found chrysocolla in green pigment samples from Karnak, but this may simply be the residue of an Egyptian green glassy frit rather than chrysocolla. Riederer (1974) found it on the walls of Theban temples, although it was found 'frequently' with frit, so the extent to which the scientific reports have been able to differentiate between a green frit and chrysocolla remains a matter of debate.

Chrysocolla does not usually create a very strongly coloured green and determination of its presence can be troublesome, as it tends to have varying optical characteristics, such as colour, birefringence, degree of crystallinity, and even difficult XRD patterns. These characteristics could result in confusion with Egyptian green, and earlier identifications would have to be confirmed by further research.

Green earth

Green earth (terre verte) usually consists of a mixture of celadonite, $\text{K}(\text{Mg},\text{Fe},\text{Al})_2(\text{Si},\text{Al})_4\text{O}_{10}(\text{OH})_2$, and glauconite, $(\text{K},\text{Ca},\text{Na})_{0.84}(\text{Al}_{0.47}\text{Fe}_{0.66}\text{Mg}_{0.40})(\text{Si},\text{Al})_4\text{O}_{10}(\text{OH})_2$. Until recently, there were only two reports of the use of green earth on Egyptian material: one from a Romano-Egyptian shrine (Berry, 1999), and the other from a Romano-Egyptian mummy cloth (Hillyer, 1984), which always seemed anomalous,

given the fact that deposits of green earth supply what would almost certainly have been a cheaper green pigment than malachite.

Scott *et al.* (2003) identified as green earth the turquoise-green surround of a sphinx on a Graeco-Roman cartonnage fragment, which was radiocarbon dated to between 395 and 184 BCE. Scott *et al.* (2009) also found green earth on cartonnage fragments in the Petrie Museum, University College London, where most of the cartonnage dates from the Twenty-second Dynasty through to the Graeco-Roman period. The earliest occurrence is on a cartonnage fragment from the Third Intermediate period, around the ninth century BCE, showing attested use well before the Graeco-Roman period, when it becomes much more common (Scott, 2010). Pagès-Camagna *et al.*, (2010) report several examples from the Louvre collections. Calza *et al.* (2007a, 2007b) probably found green earth on a Roman-Egyptian coffin of first century BCE. Clearly, the extent to which terre verte was used in Egyptian art was previously underestimated and requires further research.

Copper organometallics

Considerable problems have been created with the identification of copper organometallics (Kühn, 1993a; Lay *et al.*, 2008) which mostly derive from chemical interactions between pigment and binding media. Bimson (1987) found a green on a model axe which she thought might be an organometallic compound. (Scott *et al.*, 2003, 2004; Daniels, 2007; Scott, 2010) identified copper proteinate, copper carbohydrate, and copper wax paints, adding considerably to the complexity of characterizing the Egyptian green palette.

Copper-proteinate pigment

A copper-proteinate green was identified by Scott *et al.* (2003) from a cartonnage fragment of Graeco-Roman period. In some areas, this dark green had undergone fading to brown. This discoloration is a phenomenon well documented for copper resinates and proteinate pigments, usually prepared from verdigris dissolved in an organic medium (Scott *et al.*, 2002). Copper proteinate paints may be applied in either broad brush strokes, or with a textile swab (Woudhuysen-Keller, 1995; Woudhuysen-Keller & Woudhuysen, 1998), this could explain the unusual presence of numerous small unwoven brown fibres within the olive green paint, which is not observed in any other areas of the painted surface, explained further in Scott *et al.* (2003).

Copper-carbohydrate pigment

Another example of a copper-organometallic paint was found in the collections of the University of Southern California on an unprovenanced cartonnage

broad collar, a *wesekh* fragment, stylistically dated to between the Twenty-sixth Dynasty and the late Ptolemaic period. It was radiocarbon dated to between 512 and 351 BCE, in agreement with the stylistic analysis (Scott *et al.*, 2004). The presence of copper in the individual pigment grains was confirmed by environmental scanning electron microscopy and microscanning XRF, while gas chromatography–mass spectrometry work showed the principal binder to be gum Arabic.

Copper-wax pigments

Daniels (2007) noticed an unusual, waxy, translucent, emerald green paint on four artefacts in the collections of the British Museum, for example, a green faced coffin of Hineb, from the Twenty-sixth Dynasty. Fourier transform infrared spectroscopy and XRF analyses showed that the greens contained copper and beeswax. Beeswax heated in a copper vessel at 110°C was found to create a good green colour (Daniels, 2007). Wrapson (2010) also found evidence for a copper-wax green pigment on a red-shroud mummy in the collections of the Fitzwilliam Museum, while Liang and Scott (2014) found two examples of copper green-waxy pigments from the collections of the Metropolitan of Art small stone shabti fragments from the Nineteenth Dynasty (1292 to 1182 BCE). Such waxy pigments could be made by stewing-up verdigris with beeswax, but Liang and Scott (2014) found that malachite itself could react with beeswax during heating. This might suggest that the deep waxy-green pigments have formed as a result of the chemical interactions between pigment and binder over the long periods of time since they were fabricated by the ancient Egyptians, or they could be deliberately produced. Further research would have to be undertaken into this interesting topic.

Pigment mixtures for green

The green on a Nineteenth Dynasty papyrus at the University of Pennsylvania Museum of Archaeology and Anthropology was shown by Evans *et al.* (1980) to be made from a mixture of Egyptian blue and orpiment. Mixtures of Egyptian blue with orpiment were also identified by Green (1995) at the British Museum on a late Ptolemaic papyrus dated to the first century BCE. The degradation of orpiment and the release of sulphurous emissions may have degraded the contiguous Egyptian blue regions, producing a dark brown or dark greenish colour. Uda (2004) examined pigments in the tomb of Amenhotep III, from the Eighteenth Dynasty, using portable XRD and XRF equipments. A green on a female robe was found to consist of an iron-containing yellow and Egyptian blue.

Cartonnage fragments examined by Scott *et al.* (2009) from the Twenty-second Dynasty to the Ptolemaic period show the use of blue and yellow pigment mixtures to make green: Egyptian blue and yellow ochre, Egyptian blue and green earth, Egyptian blue and goethite, as well as Egyptian blue with an organic yellow which could not be identified. Walton and Trentleman (2009) identified another variant of pigment mixtures in the Romano-Egyptian period to create green: indigo and orpiment.

Lucas (1962) also found that a green-coloured plaster from a stick dated to the Eighteenth Dynasty, owed its colour to a mixture of blue frit and a yellow colourant that was not identified, but which was not yellow ochre.

Pigment mixtures and their importance.

Some examples of pigment mixtures have been given already in this review, and the extent to which these have been used has probably been underestimated. For example, Edwards *et al.* (2004) examined several pigments from the Seventeenth Dynasty to the Graeco-Roman period. They found a Ptolemaic coffin with a mixture of cinnabar with hydrocerussite and calcite to produce a light red, also red ochre mixed with calcite or gypsum, to produce pale red, while carbon was mixed with red ochre to make a dark red. A bright pink was made with minium mixed with calcite, and pinks were found also to be made from haematite and gypsum, while in a Twenty-first Dynasty coffin, anhydrite, gypsum, haematite, and calcite were identified. Another red was a mixture of calcite, haematite, and pararealgar, the latter probably from alteration of realgar. Yellows were admixtures of calcite, hydrocerussite, and goethite. Different shades of yellow were made from orpiment and goethite. Orpiment was also found with calcite and carbon from the Twenty-fifth Dynasty. The discovery of alternating stripes of yellow pararealgar and red haematite suggests that the use of pararealgar was deliberate. Edwards *et al.* (2004) found yellow and blue used to make green, Egyptian blue and orpiment used often, while a blue-green was found to be a mixture of calcite, Egyptian blue, carbon, and possibly an organic resin. Egyptian blue could be used to create a variety of different tonalities by addition to other pigments (Edreira *et al.*, 2003), or by different sizes of pigment particle, if ground to less than 15 μm Egyptian blue can be quite pale in colour, or deeper if kept to greater than 30 μm (Pagès-Camagna *et al.*, 2010). The samples examined by Edreira *et al.* (2003) were late, from the second century CE *Casa del Mitreo* in Spain. There Egyptian blue was found to have been mixed with yellow goethite to make green, while brown was made from a mix of haematite, calcite,

and Egyptian blue. A grey was created by adding small amounts of red ochre to Egyptian blue and white made from calcite with small additions of Egyptian blue to increase the luminosity of the pigment.

Conclusions

The rich and complex pigment palette of ancient Egyptian art represents a challenge to categorize based on substrate or function. As Bryan (2010) and Baines (1985) would remind us, the concept of colour differentiation employed here, as regards the pigments, is not how ancient Egyptian artisans would necessarily have defined them or thought about them. Nonetheless, a very rough chronology of pigment use is presented in Table 1, which not only reveals the large number of chemical compounds involved, but the difficulties of determining the time periods when they were employed.

Analytical difficulties are created by the inherent vice of some pigments which undergo photodegradation, such as realgar and orpiment, and by the numerous interactions between the pigments themselves, the burial environment, and the binding media. All of the copper-containing pigments are especially liable to undergo alteration. In the case of environmental factors, saline solutions or groundwater interact with the original pigments, such as malachite, Egyptian green, and Egyptian blue to create entirely new compounds which are green or green-blue in colour, such as atacamite or paratacamite. The knowledge of these pigment alteration problems has now permeated widely through the literature, resulting in their discussion or realization in an increasing number of published works.

With binding media derived from waxes, animal glue, and gum Arabic, organometallic copper compounds, which are often called copper soaps (Robinet & Corbeil, 2003), can be created which disguise the original identity of the pigment itself and are responsible for many problems in the identification of the green pigments.

Unlike Egyptian blue, whose use was widespread and continuous from the Fourth Dynasty to the Roman period and beyond, Egyptian green must have been kept specifically for Egyptian use as it has not been found outside of Egypt and it only seems to have been in common use from the First Intermediate period, around 2200 BCE until the Third Intermediate period, around 1069 BCE. Exactly when the pigment was first used or why it ceased production is a matter for further research, especially since it is closely allied with Egyptian blue which had such a long run of success, and which one might have thought was made as a frit in the same production process as that used for Egyptian blue.

Table 1 Chronology of painting materials use

Material	Approximate early date	Approximate end date	Notes
Anhydrite	2494 BCE	Roman period	Less common white
Azurite	2613 BCE	1334 BCE	Needs confirmation
Beeswax	3000 BCE	Roman period	As binder, commonly employed
Bitumen	1543 BCE	Not known	As varnish
Calcite	2494 BCE	Roman period	Very common
Carbon black	3500 BCE	Roman period	Probably even earlier
Chrysocolla	1991 BCE	1292 BCE	Needs confirmation
Cinnabar	747 BCE	Roman period	Rare in early periods
Cobalt blues	1479 to 1077 BCE	700 BCE to 700 CE	Gap in usage: different in type
Egg	1500 BCE	Roman period	As binder, use rare in early periods
Egyptian blue	2639 BCE	395 CE	Use continues beyond end date
Egyptian green	2200 BCE	1069 BCE	Only used in Egypt
Galena	3100 BCE	Roman period	As cosmetic
Glue	3000 BCE	Roman period	As binder
Goethite	4000 BCE	Roman period	
Gold leaf	4000 BCE	Roman period	
Green earth	900 BCE	Roman period	Needs further examples
Gums	2000 BCE	Roman period	As binder, probably earlier too
Gypsum	2494 BCE	Roman period	Very common
Haematite	4000 BCE	Roman period	
Huntite	2600 BCE	200 CE	Less common in later periods
Jarosites	2494 BCE	30 BCE	Less common after Egyptian period
Lead white	512 BCE	Roman period	Usually in later periods
Madder	1500 BCE	Roman period	Needs further work
Malachite	4000 BCE	Roman period	Formerly regarded as rare
Malachite	4400 BCE	1187 BCE	As cosmetic
Massicot	400 BCE	Not known	Needs confirmation
Minium	30 BCE	Roman period	Rare in early periods
Optical blues	2181 BCE	Not known	
Orpiment	1991 BCE	Roman period	
Pyrolusite	3000 BCE	1292 BCE	Use rare
Realgar	1543 BCE	Not known	
Realgar	2494 BCE	343 BCE	Must continue later
Soot	1187 BCE	Roman period	As cosmetic, probably earlier too
Tridymite	300 BCE	100 CE	Needs further examples
Ultramarine	1550 BCE	Not known	Use is rare as Egyptian pigment
Yellow and black varnish	1508 BCE	Not known	As varnish

The strangely sparse reports of the use of malachite and that of terre verte as pigments in ancient Egypt has only recently been corrected by research carried out in the last decade or so, and the extent of use of other minerals, such as chrysocolla are not easily resolved.

The identification of organic binders and varnishes has made considerable progress, but there are still areas where there is a lack of information, such as the extent of use of organic yellow pigments and of madder.

Further research is needed on a number of issues related to substrates. For example, whether wood, stone, papyri, wall paintings, metals, or baskets were painted in different pigments, what pigments were

available at particular times, and, depending on the status of the individual concerned, on context, on the event or God depicted, whether on elite burials or ordinary coffins, and on periods of use of the pigments themselves, which in some cases are restricted to later Greco-Roman or Ptolemaic periods. New evidence for the importation of some pigments has been found, in the case of orpiment, realgar, huntite, lapis lazuli, red lead, and jarosite. Some of these, such as huntite and jarosite, were also locally available, so the historical context of their trade requires further research. The availability or introduction of new pigments with the advent of the Romano-Egyptian period is well known, and the palette of this period is clearly different than that of other epochs.

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