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Review Article

Methods and Materials of Making Ancient and Modern Papyrus Sheets: A review.

Arzak Mohamed^{a, b,*}^a School of Natural Sciences, Faculty of Science and Engineering, Macquarie University, Sydney, NSW, 2109, Australia.^b Department of Restoration, Faculty of Archaeology, Fayoum University, El Fayoum, 63514, Egypt.

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ABSTRACT

Papyrus sheets can contain valuable medical, political, historical, and scientific information on ancient civilizations. However, their methods of manufacture remain a subject of research. This review gathers, compiles, organizes, and evaluates the available theoretical and experimental studies, and includes observations of papyrus manufacture in a modern Egyptian village, Qaramos, that is famous for making papyrus. The review also summarises past and present papyrus making techniques, including the use of strips or peels for sheet manufacture, different adhesives to bond papyrus to create a cohesive single sheet, and additives including natron (a naturally occurring mixture of sodium salts), milk, clay, cedar oil, starch, egg white, bee glue, and lotus seeds used to prepare the surface for writing. The results showed that most authors concur that ancient papyri were made using strips, but to understand the pattern and timing of use of adhesives and surface coatings better, further analyses are required. Moreover, making modern papyrus sheets requires adding chemical substances including sodium hydroxide (NaOH) and chlorine to soften the papyrus strips and lighten sheet colour.

1. Introduction

Papyrus was the first flexible writing support [1]. The oldest blank papyrus sheet, dating to the First Dynasty around 2900 BC, was found in the tomb of Hemaka at Saqqara, while the earliest inscribed papyri date to the Fourth Dynasty at 2500 BC [2,3]. Papyrus was used until the second half of the eleventh century AD when paper took its place [4]. There is no evidence that papyrus plants were used in Egypt during the Middle Ages [5], but they were cultivated in the twentieth century from roots from the Giza Zoological Garden in Cairo, and from Sudan [6]. Being portable and storable made papyrus sheets an ideal writing support in ancient times [7].

Identifying the methods and materials of making papyrus helps in understanding the deterioration mechanisms of ancient papyrus sheets and selecting appropriate preservation treatments, which might protect huge numbers of papyrus sheets around the world. Understanding the methods and materials of papyrus manufacture can also allow better understanding and interpretation of chemical analyses of archaeological papyri, which in turn allows a range of questions regarding the development of writing technology, papyri provenance, and authenticity to be addressed.

There is a lack of the literature describes in detail how papyrus sheets were made, and there is no evidence to imply that the method of papyrus manufacture has changed over time. What is known comes largely from a description by Pliny the Elder in his work Natural History [8], modern analyses of ancient papyri and modern manufacture experiments [9]. Ancient Egyptians had an economic driver to withhold information on the method of manufacture, as they sought to monopolize the production and trade of this important writing material [10]. There is a gap in information about making papyrus sheets, which highlights the need for more research focusing on the technology of making ancient papyrus.

This review addresses four questions; i.e., 1. How were ancient papyrus sheet layers prepared? 2. Which methods and materials were used to bond papyrus layers? 3. How were papyrus surfaces prepared for writing? 4. What methods and materials are used to make modern commercial papyrus?

2. Composition of papyrus plants

Papyrus supports were made from papyrus plants (*Cyperus papyrus*) [2], which grew in swamps of the Nile River delta in Egypt, with stems up to 4 m high [11]. Papyrus plants are comprised of roots, rhizomes, a triangular stem and flowers; their taxonomy, description and distribution are well

*Corresponding author.

E-mail address: Arzak.mohamed@hdr.mq.edu.au (A. Mohamed); Tel.: 00610468740416DOI: [10.21608/ifjsis.2023.208886.1017](https://doi.org/10.21608/ifjsis.2023.208886.1017)

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documented [6,12]. The chemical compositions of lignin in pith and rinds of papyrus plants were characterized [13]; white pith from the stem contains 54-60 % cellulose ($C_6H_{10}O_5$)_n and 36-40 % lignin ($C_{81}H_{92}O_{28}$) [14], however lignin forms only ~5 % of modern, commercial papyrus sheets and 16 % of papyrus pith [15]. Papyrus stems contain hydrocarbons, fatty acids, 2-hydroxyfatty acids, fatty alcohols, phytol and phytol esters, alkylamides, mono- and diglycerides, steroids (sterols, ketones, hydrocarbons, esters and glycosides), tocopherols and tocopheryl esters, alkyl ferulates, ω -carboxyalkyl ferulates (and their monoglycerides), and acylglycerol glycosides [16].

3. Making papyrus in ancient Egypt

Papyrus sheets can be made using either strips or peeling methods, as described below:

3.1. Strips

To create a papyrus sheet using strips, papyrus stems were cut to a manageable size, the outer green rind was removed, and the stem pith was cut into thin strips. The strips were then laid in orthogonal layers, without gaps between the strips (Fig. 1). The layers were then pressed to form a coherent sheet [1,2,17]. Sheets of papyri were also created in modern times using strips, and the resultant sheets were similar to the majority of ancient sheets [10,18].

3.2. Peeling

An alternative interpretation of Pliny's description is that peeling can be used to make papyrus sheets, and to test this idea, a sheet of papyrus was manufactured using stalks of the papyrus plant from the Hortus Botanicus Haren, the botanical garden of the University of Groningen [19]. A needle was used to peel the papyrus stem continuously down to its core to obtain a single sheet (Fig. 1). Orthogonal sheets were then pressed together to create a cohesive papyrus support [19,20]. Fresh plants, and modern and ancient papyri were examined using optical and electron microscopy to determine whether or not peeling was also used to make papyri in antiquity.

Peeling was found to have been used to make some ancient papyri, and differences between papyri made using strips or peels can be characterized through surface morphology. Wallert [20] claimed that papyrus surfaces made by peeling are ~50 μ m rougher than papyrus made from strips. Because of the lack of the information about making papyrus, tools involved in the manufacture were not determined. Pliny mentioned that a needle was used to cut the papyrus plan pith to obtain the peels, however the size and shape of this needle had not been previously discussed [20]. A piece of ivory or smooth shell was used to polish the ancient papyrus surface [6].

The quality of sheets created from peels or strips depends on the harvest season and which part of the papyrus plant was used to make the sheet. The mechanical properties of papyrus sheets manufactured from plants that were harvested monthly over a year were measured [21]. Plants harvested in the summer months of June, July and August gave the strongest sheets of papyrus, whereas sheets manufactured in the winter months January and February contained more lignin than samples taken from papyrus in the following months [21], and this additional lignin weakened the sheets. Which part of the stalk was used is also crucial for sheet quality. Sheets made with the lower part of the plant are more transparent and adhere together better than sheets made from the upper part of the plant [21,22].

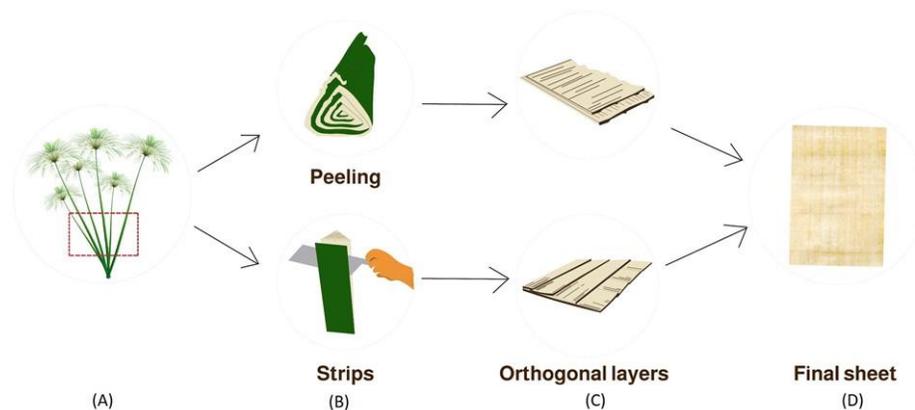


Fig. 1. A schematic showing two methods of making papyrus. (A) Remove and discard the upper and lower parts of the plant to obtain the triangular stem; (B) Peel the stem (upper) or cut it into strips (lower); (C) Layer the peels or strips into orthogonal layers; (D) Press and dry to form a final sheet.

4. Theories of sheet adhesion

Four theories might explain how papyrus layers bond together during manufacture to produce flexible cohesive sheets, which can be discussed as follow.

4.1. Nile River water

Pliny (Natural History XIII 77) [8] mentioned that muddy Nile River water has “qualities of glue”. The role of unfiltered river water and soaking time in the gluing of papyrus strips to make papyrus sheets was assessed via investigation of strip surface morphologies and measurement of breaking

strength [23]. Nile River water created adhesive forces between papyrus strips because of microorganisms, probably mostly fungi, which degraded strip surfaces, creating more adhesive regions. Strips soaked for four days in Nile River water had a breaking strength of 211 N/mm², much greater than the 69 N/mm² strength of strips that were immersed for 10 minutes only [23]. It is also possible that precipitation of dissolved salts, and electrostatic forces between clay particles, also contribute to this adhesion.

4.2. Natural gum in papyrus cell sap

Pressing or beating papyrus slices during manufacture releases naturally-occurring starch which can be sufficient to adhere the papyrus strips and layers together without additional adhesive [24]. Chemical analyses of aqueous extracts of the pith of papyrus revealed a water-soluble polymer of galactose (C₆H₁₂O₆), arabinose (C₅H₁₀O₅), and a trace of rhamnose (C₆H₁₂O₅), which are thought to be responsible for gluing the strips together, and these substances are found in the cell liquid liberated by crushing the plants during manufacture [25]. To substantiate this theory, papyrus sheets were produced successfully by soaking dried papyrus strips in water for three days without adhesive [26]. Investigation of the surface appearances of ancient samples using optical and electron microscopy supported the suggestion that natural gum in papyrus cell sap helps to bind papyrus sheets [27].

4.3. Starch adhesive

Examination of the papyrus of Ebers, held in the library of the University of Leipzig, and dated to the 16th century BC, revealed starch adhesive between the papyrus layers [28]. Analyses of various ancient Egyptian papyri reported that papyri created before 350 BC contained starch between their layers, suggesting that the use of starch as an adhesive during papyrus manufacture was common at that time [29]. Although starch adhesive can create strong bonds between layers [14] its use was not universal; examination of stained ancient papyrus samples from the Sorbonne Institute of Papyrology found no starch [22], and the age of these papyri was not reported.

4.4. Physical adhesion of the strips

Another theory claimed that physical bonds between the strips were sufficient and there was no need for other forms of adhesion [5,18,30]. Consistent with this theory, modern papyrus sheets made in this way were similar in colour to ancient sheets [30]. Papyrus plants consist of ovoid parenchyma cells surrounding hollow air spaces and vascular bundles, which when cut have a cupped shape [18]. During sheet manufacture the two strips or peels of papyrus are pressed together, and the parenchyma tissue on the surface of one strip is forced to merge into the hollow air spaces on the surface of the other strip forming a dovetail bond. Upon drying, these interlocking tissues shrink, forming a tight bond, assuring permanent adherence of the strips [6,18].

5. Additives

Flieder et al. [22] explained the brown discoloration of papyrus pith during the manufacturing process as a result of forming quinones on the last step of the oxidation of phenols in the plant pith, which support the opinion that papyrus sheets were treated with different materials to prepare the surface for writing, preventing over-absorption of ink, protecting the papyrus from deterioration, and also preventing discoloration during the manufacturing process [17,31,32].

Measurement of organic compounds in 30 papyrus samples of different ages (Pharaonic, Ptolemaic, Roman, Byzantine, and Arabic) held at The Papyrus Museum, Siracusa, Italy suggested that protein binders (milk, chicken egg white, animal glue), beeswax, cedar oil, mastic resin and binders of vegetable origin (gum Arabic, flour glue) were used to help prepare papyrus sheet surfaces for writing [31,32]. The main substances used for the treatment of papyrus sheets are discussed below, in alphabetical order.

5.1. Bee glue

Bee glue is a resinous material that worker bees collect from various plants [33] including poplars, palms and pines. Bees use the glue to seal cracks and holes in bee hives, as well as to protect the hives from microbial infection [34]. The chemical composition and colour of the bee glue differs depending on the plant species it was created from [35]. Biological characteristics of the bee glue depend upon its chemical composition, plant sources, geographical zone and seasons [34]; and more than 241 compounds have been recorded in bee glue including flavonoids, terpenes, phenolics and their esters, sugars, hydrocarbons and other mineral elements [36]. Bee glue has antibacterial, antifungal, and antioxidant effects [37], so ancient Egyptians also used it as a preservative with which to embalm mummies [38].

5.2. Cedar oil

Papyrus sheets were often covered with cedar oil [17], which was extracted from Cedar trees (*Cedrus spp.*) and contains volatile oil and compounds (α -pinene, myrcene, limonene, terpinolene and α -terpinene) which protect papyri from bacteria, fungi and attack from some insects [39].

5.3. Egg white

Egg white, also called albumen [40] or glair [41], represents about 59 % by weight of the egg composition and consists of water, proteins, carbohydrates, some minerals and vitamins [42]. Protein content includes ovalbumin 54 %, ovomucoid 11 %, ovotransferrin (12–13 %), lysozyme (3.4–3.5 %), ovomucin (1.5–3.5 %) [40] and other compounds. Although egg white is stringy and cannot be spread easily by a brush [43], it has been used as a varnish to protect paintings [44].

5.4. Clay

Sediment from Nile River contains hydrated aluminium silicates, organic matter, iron oxides and quartz (SiO₂) [45]. Clay suspended in water can

be added during papyrus sheet manufacture to make papyrus fibres denser and less absorbent [46], in addition to the adhesive role noted previously. Quantitative evidence of clay addition was provided by elemental analyses of ancient papyrus and fresh papyrus [47]; whereby ancient samples had 4.1 % aluminium (Al) and 7.2 % silicon (Si), whereas fresh papyrus had 0.009 % Al and 0.54 % Si, respectively.

5.5. Lotus seeds

An extract from Lotus (*Nelumbo nucifera*) seeds has been described [48,49], and although its purpose is not certain, it probably acted as either an adhesive or sealant. Lotus seeds were mixed with water and put on papyrus strips during manufacture and left until dry. A small piece of wood was then used to polish the ancient papyrus surface [48,49]. Lotus seeds have been used as both food and medicine for at least 7000 years, due to their soluble components, which include flavonoids, glycosides, phenolic compounds and alkaloids [50]. It is unclear which of these components acted upon the papyri, and this relationship requires further research.

5.6. Milk

Milk is one of the binding substances used in treatment of papyrus fibre in order to prevent spread of the ink on the surface during writing [51]. Mammalian milk, produced by goats, sheep, camels, cows and humans amongst other animals, is a significant resource of proteins, sugars, fats, vitamins, minerals and essential amino acids [51]. While the animal producing the milk used in ancient times was not recorded, cow's milk contains 3-4 % protein, consisting of the six major proteins α -lactalbumin, β -lactoglobulin, α s1-casein, α s2-casein, β -casein and κ -casein as well as minor proteins including bovine serum albumin, γ -casein, immunoglobulins and lactoferrin [51]. Casein, the principal protein in cow's milk [52], is insoluble in acidic to neutral water but dissolves readily in alkaline conditions, such as is found in milk [53].

5.7. Natron

Natron is a naturally occurring white, crystalline, hygroscopic material composed of sodium bicarbonate (NaHCO_3), sodium carbonate (Na_2CO_3), sodium sulfate (Na_2SO_4), sodium chloride (NaCl) and other more complex salts [54] with varying degrees of hydration [55,56], and in some samples from Egypt's Wadi Natrun, aluminium sulfate $\text{Al}_2(\text{SO}_4)_3$ has also been found [57]. In Egypt, natron is found in Wadi Natrun, which is an evaporative depression on the western edge of Egypt's Nile River delta [54]. Natron is a desiccant which is believed to prevent degradation by fungi and bacteria during mummification [56]. Natron might also be used in a similar way in papyrus manufacture. A modern attempt to manufacture papyrus in the ancient style used different concentrations of aqueous solutions of carbonate and bicarbonate of soda (Na_2CO_3 , NaHCO_3); however, the resultant papyri were brown stained even before fully dry. Treating papyrus strips with a dilute (2.5 %) aqueous solution of aluminium sulfate yielded a "satisfactory result" regarding papyrus discolouration [58]. Ancient Egyptians may also have treated papyrus strips with aluminium sulfate to obtain high quality paper, and the larger amount of aluminium in ancient papyrus compared with fresh plants is consistent with this theory [58].

5.8. Starch

Starch can be found in the leaves, seeds, fruits, stems, and roots of most plants [59]. Starch is composed of two glucose polymers, amylose (a linear chain of α -1,4 linked glucose residues with relatively few α -1,6 linked branches), and amylopectin (a branched molecule of shorter α -1,4 linked glucose molecules and more frequent α -1,6 branches) [59-61]. Starch is insoluble in cold water, and by heating a mixture of starch and water, a viscous solution is formed, which becomes a gel on cooling [62]. Starch paste was used to prepare the papyrus surface for writing by covering the pores in the fibres and prevent over-absorption and lateral spreading of the ink. Wheat starch in particular was used to increase the ability of papyrus surface to hold ink without absorption [59].

6. Forming a long roll

The size of papyrus sheets varied during the different chronological periods; during the Old Kingdom (c. 2700-2200 BC) sheets were 21-24 cm in height, whereas during the Middle Kingdom (c. 2040-1782 BC) sheets were 38 to 42 cm, and they ranged from 16 to 20 cm in the New Kingdom (1550-1070 BC) [2]. The Ptolemaic (332-30 BC) and Roman Period (30 BC-284 AD) have two heights of papyrus sheets; Augusta was 25.4 cm and Emporetica was 11.4 cm [63].

Papyrus sheets were stuck together to form a long roll, typically twenty sheets long [64]. Starch was used as an adhesive to join the sheets on the same roll [22,63]. Transmitted light was used to investigate the "Book of the Dead of the Goldworker" in Brooklyn Museum and it was observed that the join between two sheets on the roll had a dark vertical line, revealing that an adhesive was used to join the sheets to form a long roll [65]. Krutzsch [66] discussed the joining forms between papyrus sheets found in Egyptian Museum, Berlin.

Table 1. Additives involved in papyrus manufacture..

Materials	Chemical contents	Purpose	References
Bee glue	More than 241 compounds including flavonoids, terpenes, phenolics and their esters, sugars, hydrocarbons and minerals.	Acts as a preservative and to prepare the surface for writing.	[36,38,51]
Cedar oil	Volatile oil and essential ingredients (α -terpinene, myrcene, limonene, terpinolene).	Protection of the papyrus from bacteria, fungi and some insect attack.	[39]
Egg white	Water, proteins, carbohydrates, some minerals and vitamins.	A surface coating to prepare the surface for writing.	[17,42]
Clay	Hydrated aluminium silicate, organic matter, iron compounds and quartz.	Make the papyrus fibres denser and less absorbent.	[45,46]
Lotus seeds	Flavonoids, glycosides, phenolic compounds and alkaloids	Seal the pores in papyrus or as adhesive for papyrus layers.	[50]
Milk	Proteins, sugars, fats, vitamins, minerals and essential amino acids.	Prevents spread of the ink on the surface during writing.	[51]
Natron	Sodium bicarbonate, sodium carbonate, sodium sulfate and sodium chloride.	Prevents attack by fungi and bacteria.	[56]
Starch	Glucose polymers, amylose and amylopectin.	Fills papyrus pores, preventing over-absorption of ink, and increases the ability of the papyrus surface to hold ink. Helps to create a strong bond between layers.	[14,59–61]

7. Technology of making modern papyrus sheets

Bausch et al. [67] summarised the modern attempts to produce papyrus sheets. Commercially produced papyrus sheets are made in Qaramos, Egypt at the present day and the method of manufacture of papyrus in Qaramos, observed by the author (Fig. 2), and can be summarised as follows:

- Trimming harvested stems into desirable lengths using a wooden, electrically powered cutter.
- Removing the green outer rind and slicing the fresh papyrus stems into strips with a nylon thread.
- Soaking papyrus strips in a sodium hydroxide (NaOH) solution (1 cup in 20 L water) for 8-12 h to soften the strips.
- Soaking the strips in a solution of water and chlorine for 1.0 to 1.5 h to lighten sheet colour.
- Using the strips method to make papyrus sheets between linen.
- Squeezing the sheets in a press to remove excess water.
- Drying the strips between two pieces of cardboard, with three changes of cardboard until the sheet is dry.

The pH values of commercial papyrus sheets from the same village visited by the author ranged from 7.9 to 9.8, which can be attributed to the alkaline treatment step during papyrus sheet manufacture, while the pH of ancient papyrus in general ranged from 5.5 to 6.0 [67].



Fig. 2. Commercial papyrus manufacture at Qaramos, Egypt. (a) Papyrus plants ready for cultivation; (b) Cutting the stems using an electrical cutter; (c) Slicing the papyrus stem into strips with a nylon thread; (d) Soaking papyrus strips in a dilute sodium hydroxide solution; (e) Layering the strips into orthogonal layers; (f) Placing papyrus sheets between linen and cardboard; (g) Pressing papyrus sheets using a manual press; (h) A final papyrus sheet.

8. Discussion

The ways that papyrus sheets were manufactured in antiquity are little recorded, which forms a challenge for researchers, historians, analysts and conservators. Further non-destructive analyses and investigations will help to reconstruct methods of manufacture in greater detail. This shortage of historical sources documenting papyrus manufacture increases the reliance on instrumental analytical methods to unlock the ancient secrets of making papyrus, particularly regarding the use of adhesives and additives. Understanding the ways in which papyri were made will help researchers choose the most appropriate analytical methods and guide the best ways for conservators to manage their collections. This review addressed four questions.

8.1. How were ancient papyrus sheet layers prepared? Papyrus plant stems were the raw material for the manufacture of papyrus sheets. Papyrus sheets are typically manufactured using strips, and while the use of peels is also possible, the extent of that manufacturing technique in ancient times requires further investigation. Microscopic examination of ancient papyrus samples could be useful to distinguish between the two methods through the difference in the surface morphology of the tested samples. Papyrus sheet made by peeling method could be uneven because the short movement of the needle used in separating the peels from papyrus plant pith.

8.2. Which methods and materials were used to bond papyrus layers? The use of adhesives such as starch to bond the layers during creation of a cohesive papyrus sheet has been documented by some researchers, but the use of starch was not universal. Nile River water had a vital role in making papyrus as it contained microorganisms, and possibly also salts and clays, that can cause strong adhesion between the papyrus layers. Investigating if adhesives were used to join the two layers of strips or peels is a difficult process as it requires split the papyrus sheets into two layers and examine the traces of any adhesive inside. Although it is a destructive method, it can be applied to a few of ancient plain fragments.

8.3. How were papyrus surfaces prepared for writing? Some additives including bee glue, clay, egg white, milk and starch were used to prepare papyrus surfaces for writing in order to prevent over-absorption of the ink, and to increase the ability of papyrus surfaces to hold ink. Clay minerals introduced from Nile River water may also have made the papyrus fibres denser and less absorbent. Other materials such as cedar oil and natron were used to protect papyrus sheets from bacteria, fungi and insect attack. Again though, the use of these preparations on papyrus surfaces are little documented.

8.4. What materials and methods are used to make modern commercial papyrus? Modern Egyptians create papyrus sheets for tourism and sale using papyrus strips treated with chemicals including sodium hydroxide and chlorine, as untreated papyrus strips form dark paper that is unsuitable for writing. Papyrus sheets with different colour grades, ranging from dark brown to cream coloured, have been created, reflecting the concentration of chemicals used. Workers in Qaramos make papyri by strips and do not have information about the use of peeling. The idea of utilising additives in ancient times during making papyrus sheet, is verified by the observation of the production of papyrus sheets in contemporary times, particularly to prevent staining strips or peels during the process and reduce the darkening of sheets with time.

9. Conclusions

In this review, the methods and materials involved in papyrus sheet manufacture, which was an important industry in ancient Egypt, were summarised. The methods of making papyrus sheets, the adhesives, and additives used for preparing the surface for writing were discussed. A detailed description of making modern papyrus sheets in Egypt was presented. Further research using non-destructive techniques for analysing ancient papyrus sheets, is needed to characterize papyri from different ages and regions to identify differences in the methods of manufacture, with differences in structure and chemical composition.

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Declaration of Competing Interest

The author declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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