Naturally Occurring Quinones and Flavonoid Dyes for Wool: Insect Feeding Deterrents

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ABSTRACT: This study investigated the ability of natural dyes to impede the attack of black carpet beetles on wool. The dyes evaluated were naturally occurring quinines (cochineal, madder, and walnut) and flavonoids (chestnut, fustic, indigo, and logwood). All of the dyes, except indigo, were applied by using five mordanting agents (aluminum, chrome, copper, iron, and tin). Mordanting agents were used to bind natural dyes on wool. The insect resistance of the controls and dyed specimens was evaluated, following the fabric weight loss procedures in AATCC Test Method 24. All of the dyes, except indigo, increased the insect resistance of the wool fabric to attack by black carpet beetles. The wool specimens dyed with cochineal and madder, naturally occurring anthraquinone dyes, had the lowest fabric weight loss among eight natural dyes investigated. The flavonoid dyes were not effective in enhancing insect resistance. Alum, copper, and iron have no significant effect on enhancing or reducing insect resistance of any of the eight natural dyes used in this study. However, tin and chrome, when used as a mordant for cochineal, reduced the insect resistance dramatically. Surprisingly, tin improves the insect resistance of wool fabrics dyed with fustic dyes. The anthraquionones including cochineal, madder, and walnut were found to be quite effective in protecting wool fabric against black carpet beetles. Thus, the naturally occurring mordant dyes provide an alternative to insecticides in protecting wool textiles from insect attack. © 2005 Wiley Periodicals, Inc. J Appl Polym Sci 98: 322–328, 2005

Key words: natural dyes; mordanting agents; black carpet beetle; insect resistance; quinones; flavonoids; wool fabrics

INTRODUCTION

Wool and specialty hair fibers are natural fibers with outstanding esthetic and performance properties. Wool comes from different breeds of sheep, whereas specialty hair fibers are obtained from animals in the goat and camel family, and other animals, such as rabbit, fox, and mink. Wool and specialty hair fibers have almost similar chemical properties, but they differ in morphology, performance, and physical characteristics, such as fiber length, crimp, scale structure, strength, and color. Wool and specialty hair fibers have many distinguished properties, such as heat insulation, flame retardancy, softness, and resilience. These inherent qualities influence a number of end uses, such as apparel, home furnishings, and decorative and industrial items.[1] However, wool has a serious problem in that it is susceptible to insect damage from clothes moth and carpet beetle larvae.[2]

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Insect damage on wool-containing products, including carpets, garments, upholstered furniture, blankets, and priceless heirlooms, has increased annual financial loss in United States.[3] Even a small amount of wool eaten from a suit or a rug can ruin its functional and/or esthetic properties.[4] Continuous damage can result in significant economic losses, especially in warehouses and showrooms.

Wool and specialty hair fibers are composed of the insoluble protein, keratin, which contains 18–19 specific amino acids and have highly crosslinking disulfide bonds. Some insect larvae are able to eat wool[2] to obtain sulfur, which is an essential factor in insect growth.[5]

Clothes moths, a family of Tineidae, and carpet beetles, a family of Dermestidae, are the major insects that cause serious destruction of protein-containing textiles. The most common species of clothes moths are the webbing clothes moth, *Tineola bisselliella* Hum., the case-making clothes moth, *Tinea pellionella* L., and the carpet moth, *Trichophage tapetzell*a L. They are distinguished by appearance. The six types of beetles are the black carpet beetle, *Attagenus megatoma* F., the varied carpet beetle, *Anthrenus verbasci* L., the common carpet beetle, *Anthrenus scrophulariae* L., the cabinet beetle, *Trogoderma* spp., the hide beetle, *Dermestes* spp., and the odd beetle, *Thylodrias contractus*.[3,6] Generally, carpet beetle larvae have small, elliptical, brownish bodies covered with long hairs. Several conditions, including food, temperature, and moisture, influence the growth of the insects, especially larvae.

There are two major approaches to controlling clothes moths and carpet beetles, as follows: (1) no-chemical methods and (2) chemical methods. No-chemical methods use extremely high temperature or electromagnetic radiation frequencies to kill insects. Chemical methods include the use of several natural and synthetic insecticides, biological control, chemical modification of wool, laundering with strong soap solutions, and dry-cleaning.[3] Chemical methods are more popular, common, and easy to apply.

Depending on chemicals and target insects, chemical methods function by specific modes of action. For example, insect repellents function by preventing or discouraging insects from landing in the vicinity of treated fabric, whereas insecticides are chemicals that kill insects, usually after a small amount of the treated fiber has been ingested.[6]

To minimize the toxicity and side effects of synthetic pesticides, many scientists have researched naturally occurring products.[7] A limited number of natural products, including polygodial and 9-deoxymuzidadial extracted from the leaves of the New Zealand native tree,[8] sweet flag,[9,10] pyrethrins,[9] and cedar wood,[9] have been evaluated on protein fibers and/or fabrics against fabric pests. Another important category of natural products that have potential use is natural dyes, such as chestnut, cochineal, fustic, indigo, madder, logwood, walnut, which comes from plant's leaves, roots, and flowers, and insects. Natural dyes can be dyed on wool to provide brilliant colors, and they are related quinones- and flavonoids-containing products. Naturally occurring quinines and flavonoids are known to have insecticidal or insect-repellent properties.[11]

When applying many natural dyes on wool, metal mordanting agents are needed to bind the dye molecule to wool. Mordanting agents, such as chrome, copper, iron, and tin, can also be toxic depending on the concentration used. Today, aluminum is often the preferred mordant agent because of its low toxicity. The combinations of natural dyes and mordanting agents create brilliant colors. Indigo, a vat dye, does not require the use of mordanting agents to provide adequate durability. Natural mordant and vat dyes, such as indigo, also may enhance the resistance of wool textiles to insect attack.

Natural dyes would provide an alternative to synthetic insecticides, which are more detrimental to the health and safety of humans and the environment. By properly selecting the dyestuffs for wool textiles that have inherent resistance to insects, the need for insecticidal chemicals could be greatly reduced. It also would reduce the cost of manufacturing because conventional insect repellency treatments would not be needed. Therefore, the objective of this study was to evaluate eight natural dyes and the combination of dyes and mordanting agents for insect repellency and/or insecticidal effect against black carpet beetle.

TABLE I
Names, Chemical Structures of Mordanting Agents

Mordanting Agents		Chemical Structures
Aluminum	Potassium aluminum sulfate	$KAl(SO_4)_2$, $12H_2O$
Chrome	Potassium dichromate	$K_2Cr_2O_7$
Copper	Copper sulfate, blue vitriol	$CuSO_4$
Iron	Ferrous sulfate	$FeSO_4$
Tin	Stannous chloride	$SnCl_2$

EXPERIMENTAL

This study focused on evaluating the effectiveness of eight natural dyes in protecting wool fabric from black carpet beetles. Seven of the natural dyes were applied in conjunction with five mordanting agents, whereas the indigo was applied by a conventional reduction/oxidation method.

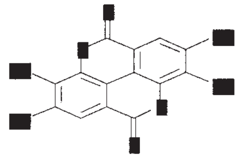
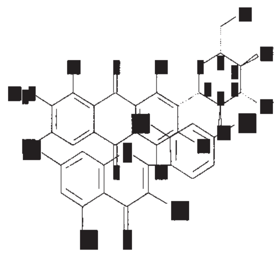
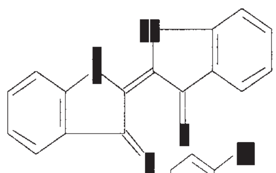
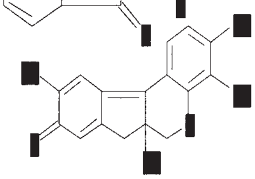
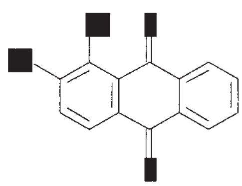
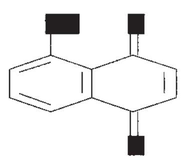
Materials

The fabric used for the experiments was a medium-weight (g/m^2) wool flannel, style 527, obtained from Testfabrics, Inc (Pittston, PA). The fabric was prescoured prior to mordanting and dyeing processes to remove surface impurities that would influence treatment and/or insect testing. The scouring solution contained 0.1% (w/w) AATCC soap and 10% (w/w) sodium carbonate. All chemicals were purchased from Fisher Scientific, Pittsburgh, PA. The fabric was scoured in the solution by using a 40 : 1 liquor-to-goods ratio at 52 ± 2°C for 5 min. Two warm rinses were followed by 3 min of neutralization in a water bath with 1% acetic acid at 35 ± 2°C. After scouring, the wool was air dried in ambient conditions, cut into the requisite specimen size, and randomly assigned to the treatment subgroups.

Mordanting

The five mordanting agents selected for evaluation were aluminum (aluminum sulfate), chrome (potassium dichromate), copper (copper sulfate), iron (iron oxide), and tin (stannous chloride). All of these mordanting agents have been used historically as mordanting agents in the textile industry and by handweavers. A premordanting procedure was used in which the specimens were mordanted prior to dyeing. The chemical structures of mordanting agents are listed in Table I.

TABLE II
Names, Chemical Structures, Active Ingredients, Colors, and Sources[11] of Natural Dyes Evaluated

Natural dyes	Active ingredient	Chemical structure	Color imparted	Source
Chestnut, *Costanea sativa*	Ellagic acid (isoflavones)		Butter yellow	Bark and heartwood
Cochineal, *Dactylopus coccus*	Carminic acid (anthraquinones)		Fuchsia to purple	Dried bodies of the female beetles, *Coccus cacti* L.
Fustic, *Chlorophora tinctoria*	Morin, a clavonol, and maclurin		Golden yellow	Heartwood of mulberry
Indigo, *Indigofera suffruticosa, tinctoria*	Indigotin (indigoid)		Light to navy blue	Leaves of various species of indigofera
Logwood, *Haematoxylon campechianum*	Hematein and haematoxylin		Gray/black and purple	Heartwood of logwood plants
Madder, *Rubia tinctoria*	Alizarin and purpurin (anthraquinones)		Orange to brick red	Ground root of the plant, *Runis tinctorum*
Walnut, *Juglans nigra*	Juglone (naphthoquinone)		Fawn to chocolate brown	Outer covering of the shells of the walnut

Each mordant solution contained 4% owf (on the weight of fabric), except aluminum, which is nontoxic, so a 25% owf concentration was used. The fabrics were treated in the solution by using a 40 : 1 liquor-to-goods ratio at 93 ± 2°C for 1 h, followed by a 2-min warm wash, until the water ran clear.

Application of natural dyes

Eight natural dyes were selected for evaluation, based on their historical/commercial importance and potential insecticidal properties. The dyestuffs included three naturally occurring quinines (cochineal, madder, and black walnut) and six flavonoids (chestnut, fustic, indigo, and logwood). The chemical structures and the active ingredient of natural dyes are listed in Table II.

Application of natural dyes

All of the natural dyes were applied to premordanted wool as described below, except indigo, which was applied by a conventional reduction/dyeing/oxidation procedure.

Indigo

The indigo dye stock solution was prepared by mixing 56 g of indigo in warm water to make a paste. The solution was opaque and blue. To dissolve and reduce the indigo, 2 tablespoons of alkali, sodium hydroxide, and thiourea dioxide were added to the stock solution. After 15 min, the stock solution changed from a dark blue to a translucent green-yellow. It was important to observe the color change of solution because it represented the state of reduced or oxidized solution.

The indigo dye bath was prepared by diluting the stock solution and adjusting the pH 10.5 ± 0.5 and temperature to 125 ± 5°C. The specimens were dyed for 5 min and then removed and oxidized by exposing to room temperature.

Application of natural dyes

Dye baths of the seven natural mordant dyes were prepared by dissolving the required amount of dye (chestnut, 10% owf; cochineal, 12% owf; fustic, 3.3% owf; logwood purple and gray, 19% owf; madder, 80% owf; and walnut, 10% owf) in water to give a 40 : 1 liquor-to-goods ratio. The pH of the baths was adjusted to 5.5 with diluted sodium hydroxide. The premordanted wool fabrics (33 g) were prewetted in distilled water and then immersed in the dye baths at 45°C. The temperature of the dye baths was raised to 93 ± 2°C over 30 min and dyeing continued for 45 min at this temperature. Then the temperature was allowed to cool slightly to 55°C. Finally, the samples were removed and rinsed thoroughly in warm, distilled water.

Evaluation of the insect resistance

Black carpet beetle culture

The black carpet beetles were reared on finely ground, sterilized, laboratory chow (5001 Rodent Diet, PMI Nutrition International, Inc., Brentwood, MO) with the moisture adjusted to 13% (w/w). Periodically, the jars, containing the black carpet beetle larvae, were examined for pupae. The carpet beetle larvae were 3–4 months old, had an average weight of 6–7 mg, and were retained on a No. 16 screen. All of the specimens were incubated for 14 days in a Hot Pack Environmental Chamber maintained at 27 ± 1°C and 55 ± 2% relative humidity (RH). The original culture of the carpet beetles was obtained from S.C. Johnson and Son Corp. (Racine, WI).

Biological properties analysis

Several methods were used to evaluate the insect resistance of wool. The most common methods for measuring insect damage are (1) visual examination, (2) determining the quantity of excrement matter deposited by the larvae, (3) specimen weight loss for assessing damage,[12] and (4) the percent weight gained/lost by certain insect larvae, which is used as a measure of the insect resistance property of wool fibers.[13] Visual examination of feeding is a simple and direct way. The excrement weight method is only used for carpet beetles because of the difficulties encountered in collecting the excrement of the clothes moths. The most general method is to determine the actual loss in weight of wool sample. The weight loss was based on the difference of the weight of the specimen before and after testing. The percent weight gain and loss data was obtained by the oven-dry weight of the specimen, both before and after treatment.

Testing for insect resistance consisted of two categories: biological test method and chemical test methods. Biological test methods are more commonly used. International Wool Textile Organization (IWTO), American Association of Textile Chemists and Colorists (AATCC) Test Method 24, International Organization for Standardization (ISO) ISO 3998: 1977, Australian Standard AS 2001.6.1.1980, and Swiss Standard SNV 195901 have their own methods. IWTO is reading weight loss in comparison with untreated controls and controls treated with dinitronaphthol. AATCC test method is based on the excrement weight or fabric weight loss for beetles and fabric weight loss only for moths. ISO 3998-1977 and AS 2001 are concerned with visual assessment of cropping and holes and fabric weight loss. SNV 195901 takes only weight loss in comparison with untreated controls.[13] In addition to fabric weight loss, the actual weight gain/loss of the insect larvae can also be measured on the basis of oven dried body weight. This test method is reliable and reproducible, especially if the substrate is not in the fabric form.

Insect feeding deterrents

The insect feeding of the untreated and treated wool specimens by black carpet beetle, *A. megatoma*, larvae

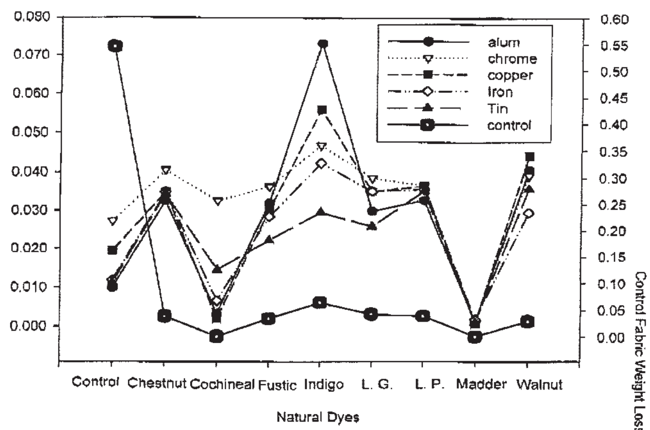


Figure 1. Weight loss of control fabric and treated fabrics. (L.G. is Logwood Gray, and L.P. is Logwood Purple.)

TABLE III
ANOVA (two-way) for Main Effects of Mordanting Agents and Natural Dyes and Interaction

Source	DF	Type 1 SS	Mean square	*F* value	Pr > F
MA	5	0.08327283	0.01665457	24.87	<.0001
ND	8	0.12204995	0.01525624	22.79	<.0001
REP	2	0.00081185	0.0000592	0.61	0.5473
MA*ND	40	0.62305871	0.01557647	23.26	<.0001

MA is Mordanting Agent; ND is natural dye; and REP is replication.

was evaluated following the procedures in AATCC Test Method 24-1999, Insect Resistance of Textiles (AATCC, 1999). Specimens, measuring 3.5 × 3.5 cm, of the untreated and treated wool fabric were placed in clear plastic insect boxes. Next, 10 larvae were placed on the face of the preweighed specimens; the boxes were securely closed and placed in the Hot pack environmental chamber for 14 days at 27 ± 1°C (80 ± 2°F) and 55 ± 5% RH with light excluded. After the 14-day test period, the insects were removed carefully with forceps. The samples were tapped gently to dislodge the excrement.

The test specimens and humidity check specimens were weighed after testing on a microbalance to the nearest 0.0001 g (1 mg). The specimen weight loss in milligrams was adjusted for humidity changes based on the weight changes in the humidity check specimens that accompanied the test specimens throughout the testing. The number of live and dead insects removed from the specimens was counted and recorded. Each test cage initially had 10 insects:

$$L = [(AC)/B] - D \quad (1)$$

where$L$ is the adjusted loss in weight in milligrams due to insect feeding; A is the average weight of the four test specimens before testing; B is the average weight of the four humidity check specimens before testing; C is the average weight of the four humidity check specimens after testing; and D is the average weight of the four test specimens after testing.

RESULTS AND DISCUSSION

This study examined the potential use of one natural vat dye (indigo) and seven natural mordant dyes, applied in combination with five mordanting agents, to impede the feeding of black carpet beetle on wool flannel. Presented in Figure 1 is the fabric weight loss data for the wool specimens subjected to carpet beetle larvae for the 14-day test under controlled conditions. The mean weight loss values for the three experimental replications ranged from 0.6647 mg for the untreated controls to 0.0010 mg for the specimens dyed with madder.

The weight loss of wool fabric analyzed using a two-way ANOVA statistical test to examine the influence of the main effects of mordanting agents and natural dyes and their interactions on insect resistance is presented in Table III. Statistical significance was recorded for mordanting agents ($P < 0.0001$) and natural dyes ($P < 0.0001$) in terms of the amount of weight loss attributed to insect feeding. Significant interaction also was noted among the mordanting agents and natural dyes ($F_{obs} = 23.26$ and $P < 0.0001$). Conversely, there were no significant differences among three replications ($F_{obs} = 0.61$ and $P = 0.5473$).

Effects of natural dyes

The anthroquinone dyes, such as cochineal and madder, had the least amount of damage, indicating that the dye was effective in reducing insect attack[14] (Table IV). The wool specimens dyed with madder also exhibited no holes or surface damage. However, such an insect resistance was not observed for wool fabrics dyed with walnut, even though walnut is also an anthroquinone product[14,15] (Table IV).

There was significant fabric weight loss among the specimens dyed with chestnut, fustic, logwood, and indigo (Table IV), indicating that the flavonoid prod-

TABLE IV
Fabric Weight Loss (%) for Quinone[a] and Flavonoid[b] Dyes

	Chestnut (mg)	Cochineal (mg)	Fustic (mg)	Indigo (mg)	Logwood gray (mg)	Logwood purple (mg)	Madder (mg)	Walnut (mg)
Mean (SD)	0.0383 (0.0034)	0.0012 (0.0012)	0.0333 (0.0008)	0.0633 (0.0034)	0.0423 (0.0036)	0.0388 (0.0034)	0.0011 (0.0010)	0.0286 (0.0039)
% loss[c]	5.76%	0.18%	5.01%	9.53%	6.37%	5.84%	0.17%	4.31%

[a] Quinones include cochineal, madder, and walnut.
[b] Flavonoids include chestnut, fustic, indigo, and logwood.
[c] The % fabric weight loss is based on the average weight of the conditioned fabric specimen (5 cm^2), which is 0.6647 mg (SD: 0.0221).

ucts did not offer wool products any protection from insect attack.[11,16] In fact, the black carpet beetles ate a considerable amount of the indigo-dyed fabrics.

Effect of mordanting agents

Alum, copper, and iron were good mordants for all the natural dyes tested in improving color and they had no significant effects in enhancing or reducing insect resistance of the eight dyes studied. There were significant differences between cochineal and madder with chrome- and tin-treated fabrics. Both tin and chrome as mordants for cochineal dye increased the fabric weight loss due to insect attack, thereby reducing the insect resistance provided by cochineal. Chestnut, logwood gray, logwood purple, fustic, madder, and walnut had a similar amount of weight loss because of insect feeding, and there was no significant difference among mordanting agents. The black carpet beetles consumed a significant amount of indigo-dyed fabric during the experiment.

The effect of mordanting agents on insect resistance also is shown in Figure 1. Among the mordants, the undyed specimen weight loss was the lowest due to insect feeding for alum (0.0098 mg). Tin and iron showed slightly higher weight losses, and chrome and copper showed serious weight loss and fabric surface damage, indicating that they were less effective in reducing insect attack.

When using aluminum as a mordanting agent, the wool specimens dyed with chestnut, fustic, logwood gray, logwood purple, cochineal, madder, and walnut had similar weight losses (Table V). Chrome as a mordant with cochineal dramatically increased the fabric weight loss compared to other dyes (Table V). Copper and iron resulted in similar fabric weight losses on the naturally dyed wool, thereby having no influence on enhancing or impeding the insect attack. Like chrome, tin also had a significant increase on the amount of insect damage in the specimens dyed with cochineal (fabric percent weight loss increased from 0.18 to 2.17%). However, tin improved the insect resistance offered by fustic dye (Table V).

CONCLUSIONS

The insect resistance of eight naturally occurring quinones and flavonoids dyes applied to wool with five mordanting agents was studied in detail by using laboratory-reared black carpet beetles. The anthraquionone dyes, such as cochineal and madder, were found to be very effective in protecting the wool fabric against black carpet beetles. Madder resulted in the lowest weight loss and the best effect of insect deterrence against black carpet beetles. Among the flavonoids, chestnut, fustic, logwood, and indigo dye when used on wool will not provide insect resistance.

TABLE V
Fabric Weight Loss (%) for Five Mordants Used with Each Dye Type

Mordant	Chestnut[2]	Cochineal[1]	Fustic[2]	Indigo[2]	Logwood[2] gray	Logwood[2] purple	Madder[1]	Walnut[1]
Alum	0.0325[a] (0.0040)[b] 4.89%[c]	0.0032 (0.0016) 0.48%	0.0297 (0.0044) 4.46%	0.0732 (0.0116) 11.01%	0.0298 (0.0010) 4.49%	0.0328 (0.0050) 4.94%	0.0010 (0.0010) 0.15%	0.0406 (0.0012) 6.10%
Chrome	0.0406 (0.0046) 6.10%	0.0325 (0.0037) 4.89%	0.0362 (0.0077) 5.45%	0.0468 (0.0046) 7.04%	0.0385 (0.034) 5.79%	0.0367 (0.0047) 5.53%	0.0012 (0.0009) 0.19%	0.0391 (0.0040) 5.88%
Copper	0.0348 (0.0059) 5.24%	0.0018 (0.0015) 0.27%	0.0301 (0.0027) 4.53%	0.0560 (0.0114) 8.42%	0.0350 (0.0030) 5.27%	0.0365 (0.0032) 5.49%	0.0005 (0.0003) 0.08%	0.0443 (0.0068) 6.66%
Iron	0.0348 (0.0061) 5.24%	0.0064 (0.0031) 0.96%	0.0283 (0.0030) 4.26%	0.0423 (0.0043) 6.37%	0.0350 (0.0019) 5.27%	0.0356 (0.0099) 5.31%	0.0015 (0.0015) 0.23%	0.0294 (0.0021) 4.42%
Tin	0.0343 (0.0086) 5.17%	0.0144 (0.0047) 2.17%	0.0221 (0.0029) 3.33%	0.0294 (0.0024) 4.42%	0.0256 (0.0016) 3.86%	0.0351 (0.0078) 5.28%	0.0017 (0.0011) 0.26%	0.0356 (0.0022) 5.36%

[a] Mean of weight loss (mg).
[b] Standard deviation.
[c] % Weight loss based on the conditioned fabric specimen (5 cm^2), which is 0.6647 mg (SD: 0.0221).

Among mordanting agents, alum, copper, and iron can be used with the eight dyes without increasing insect susceptibility of the dyed wool products. Using chrome or tin as a mordanting agent for dyeing wool with cochineal is not recommended as it increases insect susceptibility.

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