

Graft Copolymerization of Acrylamide on Cotton Cellulose in a Limited Aqueous System Following Pretreatment Technique

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SYNOPSIS

Graft copolymerization of acrylamide on cotton (dialdehyde cellulose, DAC) fibers and fabrics was studied in a limited aqueous system using $K_2S_2O_8$ as the initiator. Grafting parameters under different sets of conditions were determined and the mechanism of graft copolymerization discussed. Optimum conditions for grafting were established and the effect of polyacrylamide grafting on tenacity, modulus, breaking elongation, and stiffness of the cotton (DAC) fabrics and on their dyeability and moisture regain properties were also studied; 9–10% grafting of polyacrylamide on (DAC) fabric at pH 7–10 imparts an improved balance in its mechanical and other properties. © 1995 John Wiley & Sons, Inc.

INTRODUCTION

Reports on graft copolymerization of acrylic monomer such as acrylonitrile, methyl acrylate, and methyl methacrylate are widely available in the literature.^{1–6} Grafting in such cases renders cellulose fibers and fabrics somewhat less hygroscopic or hydrophilic in view of the hydrophobic nature of the relevant grafted-on acrylic polymers. Acrylamide monomer giving a hydrophilic polymer has drawn limited attention for studies in graft copolymerization on cellulose. Likewise, vinyl grafting on oxycellulose has also been studied^{5–9} scantily. It is in this background that studies of graft copolymerization of acrylamide on cotton cellulose fibers and fabrics of a controlled, low degree of oxidation were undertaken and relevant results are reported in this paper.

EXPERIMENTAL

Materials

Cotton Cellulose

Mill cleaned S-4 quality cotton fiber and a plain weave cotton (poplin) fabric (having an average of

315 ends and picks per dm and an average warp and weft count of 47.23 tex) were used. The fabric weighed about 108 g m².

Monomer and Other Chemicals

AR-grade acrylamide (AAm) (monomer), potassium persulfate (initiator), and sodium metaperiodate (reagent for oxidation of cotton cellulose) were obtained from E. Merck.

Methods

Scouring of Cotton Cellulose

A 10 g portion of the selected cotton sample was briskly boiled for 3 min in 500 mL dilute NaOH solution (0.08%) containing 0.1 g of a nonionic surfactant. The sample was then washed successively with hot water, cold water, dilute HCl solution (1%), and finally with cold water until it was acid free. The scoured cellulose thus obtained was dried at room temperature in a current of dust-free air.

Limited Oxidation of Cotton Cellulose

Selective oxidation of scoured cotton cellulose to dialdehyde cellulose (DAC) was carried out using an aqueous solution of $NaIO_4$ (0.05 mol L⁻¹) according to a known procedure.⁵ Limited oxidation of the

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secondary alcoholic groups (—CHOH) in the C-2 and C-3 positions of the anhydro β -glucose units of cellulose to aldehyde (—CHO) groups was readily and selectively accomplished under the given conditions. The degree of oxidation accomplished was estimated and expressed in terms of copper number following a standard procedure.¹⁰ Oxycellulose of low degree of oxidation (copper number 4.0) thus obtained will be referred to as dialdehyde cellulose (DAC). Cellulose fiber or fabric (copper number 0.24 ± 0.01) was subjected to mild oxidation so as to make it more reactive and amenable to grafting.⁵⁻⁹ The degree of oxidation was limited to a low level (giving an increase in copper number from nearly 0.25 to 4.0) because a higher degree of oxidation may tend to weaken¹¹ the base fiber and fabric.

Graft Copolymerization

A known weight (0.2–2.5 g) of DAC prepared was soaked in an aqueous solution of $\text{K}_2\text{S}_2\text{O}_8$ initiator of known concentration (0.008–0.04 mol L^{-1}) for 5 min using a fiber/liquor ratio of 1 : 10 (w/w). The DAC substrate was then squeezed hard to remove excess solution as far as possible. The uptake of $\text{K}_2\text{S}_2\text{O}_8$ was calculated from the increase in weight after squeezing, assuming no preferential absorption. The squeezed sample was then taken in a stoppered test tube or conical flask, flushed with purified N_2 for 5 min, tightly stoppered, and exposed to light from a pair of 40 W fluorescent tube lamps for 30 min to obtain pretreated DAC. To this a requisite volume (1–25 mL) of aqueous acrylamide solution of known concentration (0.25–2.5 mol L^{-1}) was added under N_2 cover. The pH of the reaction medium was adjusted to desired levels using small volumes of dilute acetic acid or dilute caustic soda, if required.

The test tube or conical flask was then placed in a thermostated bath maintained at a specific temperature (40–70°C) and graft copolymerization was allowed to proceed over a specified time period (1–6 h). The gross polymer product was then completely precipitated out by steeping in water–methanol (1 : 3 v/v) mixture (20–250 mL) and then only in methanol (5–60 mL). The product was finally dried in vacuum at 45°C to a constant weight. The free polymer (polyacrylamide) was isolated from the gross product by extraction with distilled water under reflux in a sohxlet apparatus for 72 h. The residue was then thoroughly washed with hot water, dried in air, and finally in vacuum at 45°C. The dried residue expectedly containing the DAC–polyacrylamide graft copolymer and possibly some ungrafted

DAC also is termed the apparent graft copolymer. Percent grafting, grafting efficiency, and total conversion were calculated in the usual manner.⁵

Measurement of Tensile and Other Properties of Fabric Samples

The fabric tenacity and breaking elongation (%) were evaluated¹² using a Zwick 1445 CRT Universal tensile testing machine as per IS: 1969–1968. Test specimens of 5.0×2.0 cm were used for this purpose. The initial modulus (i.e., the modulus at 1% elongation) was calculated from a load-elongation master curve. Stiffness of the fabric samples was measured¹² with the help of a Cantilever Type SAS-MIRA stiffness Tester as per IS: 6490–1971.

Dyeability Studies

Cotton cellulose (DAC) fabrics both ungrafted and grafted with polyacrylamide were subjected to dyeing in a dye bath using a reactive dye and an acid dye as follows.

Dyeing with the Use of a Reactive Dye

Procion brilliant red M 8B dye (CI reactive red 11) obtained from Atic Industries, Gujrat was used for this study. For 1 g of fabric, 0.01 g of the dye was used in 100 mL of aqueous 5% Na_2SO_4 solution. The dyeing was done at 60°C for 1 h after which alkali fixation was achieved on addition of aqueous NaOH solution in two equal installments over a 30-min time period so as to give an overall 1% NaOH concentration in the final solution. The dyed sample was then squeezed, neutralized with dilute acetic acid solution, and soap washed in a separate bath as per the ISO-I washing method.¹³ The soap-washed sample was further washed with distilled water and dried.

Dyeing with the Use of an Acid Dye

Atul crocein scarlet MOO (CI Acid red—73) obtained from Atic Industries, Gujrat was used for this purpose. For 1 g of fabric sample, 0.01 g of the dye was used in 100 mL aqueous 5% Na_2SO_4 solution and a requisite volume of 2% HCOOH solution was added to adjust the pH to 4–4.5. The dyeing was carried out for 1 h at 100°C, after which the dyed sample was squeezed, neutralized with dilute NaOH solution, and then soap washed as per the ISO-I washing method¹³ and dried in air.

Table I Graft Copolymerization of Acrylamide on DAC Initiated by K₂S₂O₈ in Limited Aqueous System following Pretreatment Technique: Effect of Variation of Initiator Concentration

K ₂ S ₂ O ₈ (mol L ⁻¹)	Gross Polymer (g)	Apparent Graft Copolymer (g)	Total Conversion (%)	Grafting (%)	Grafting Efficiency (%)
0.008	0.380	0.210	90	5	5.5
0.02	0.390 (0.380)	0.224 (0.211)	95 (90)	12 (5.5)	12.6 (6.1)
0.04	0.390	0.242	95	21	22.1
0.06	0.396	0.274	98	37	37.7

Acrylamide = 0.2 g, temperature = 60°C, time 3 h, pH 5, water = 3 mL, cotton fiber (DAC, copper no. = 4) = 0.2 g. Data in parentheses give results of corresponding grafting experiment done without pretreatment by photoexposure.

Measurement of Dye Uptake

The dye uptake, i.e., depth of shade levels of a dyed cotton fabric sample (ungrafted or grafted) was evaluated and expressed in terms of K/S value at the respective λ_{\max} employing a Macbeth color Eye 2020+ reflectance spectrophotometer using the equation of Kubelka and Munk¹³:

$$K/S = \frac{(1 - R_{\lambda_{\max}})^2}{2R_{\lambda_{\max}}} \propto C_D$$

where K = coefficient of absorption, S = coefficient of scattering, $R_{\lambda_{\max}}$ = reflectance of dyed sample at λ_{\max} , C_D = the dye uptake level in the dyed sample. The effect of polyacrylamide grafting in influencing the dye uptake level was assessed by comparing the observed K/S values for grafted and ungrafted fabrics.

RESULTS AND DISCUSSION

General Observations

Preliminary experiments with scoured cotton cellulose (copper number 0.25) and oxycellulose or

DAC (copper number 4.0) each soaked with K₂S₂O₈ initiator solution and the subsequent attempt to graft copolymerize acrylamide following exposure to light for 30 min (pretreatment technique) or without prior photo-exposure revealed that initial photoexposure or pretreatment of the fiber/fabric significantly favored grafting over homopolymerization of acrylamide. Percent grafting was relatively low and homopolymerization of acrylamide was relatively high causing lower grafting efficiency when the specified pretreatment was not effected. The presence of polyacrylamide residue in the grafted cotton (DAC) fiber is evidenced from the appearance of a strong absorption band at 1650 cm⁻¹ where two close absorption peaks for C=O stretching, and mesomeric C—N stretching are believed to be coupled with N—H deformations.

A relatively high degree of gelled products was usually obtained somewhat early during polymerization causing more difficulty in analysis and product evaluation if the pretreatment technique was bypassed. Limited oxidation of cellulose also favored relatively high levels of polyacrylamide grafting much as in earlier observations for similar grafting using methyl methacrylate and other monomers.⁵

Table II Graft Copolymerization of Acrylamide on DAC Initiated by K₂S₂O₈ in Limited Aqueous System following Pretreatment Technique: Effect of Variation of Monomer Concentration

Acrylamide (mol L ⁻¹)	Gross Polymer (g)	Apparent Graft Copolymer (g)	Total Conversion (%)	Grafting (%)	Grafting Efficiency (%)
0.5	0.364	0.209	77.0	4.5	5.5
1.0	0.383	0.220	86.0	10.2	11.5
1.5	0.393	0.237	90.6	18.6	19.2
2.0	0.392	0.243	90.0	21.7	22.6
2.5	0.395	0.268	91.5	34.0	35.0

K₂S₂O₈ = 0.006 g, temperature = 60°C, time 3 h, pH 5, cotton fiber (DAC, copper no. = 4) = 0.2 g, acrylamide = 0.213 g (volume of water adjusted).

Table III Graft Copolymerization of Acrylamide on DAC Initiated by $K_2S_2O_8$ in Limited Aqueous System following Pretreatment Technique: Effect of Variation of pH

pH	Gross Polymer (g)	Apparent Graft Copolymer (g)	Total Conversion (%)	Grafting (%)	Grafting Efficiency (%)
2	0.356	0.198	78.0	Low/measurable acidic degradation	
3	0.374	0.243	86.8	21.0	34.2
4	0.391	0.250	95.6	25.0	26.1
5	0.393	0.232	96.7	16.0	16.5
7	0.380	0.216	90.0	8.2	9.0
10	0.390	0.231	95.0	15.5	16.3

Acrylamide = 0.2 g, $K_2S_2O_8$ = 0.006 g, temperature = 60°C, time 3 h, cotton fiber (DAC, copper no. = 4) = 0.2 g.

The bulk of the studies presented here were therefore done using DAC of a low degree of oxidation (copper number 4.0) and the grafting of acrylamide was effected following the pretreatment technique. Results of graft copolymerization of acrylamide on the cotton substrate, mostly DAC, are presented and discussed under two major heads: studies of the graft copolymerization process and studies of the effect of grafting on properties of cotton-DAC fabrics.

Graft Copolymerization

Effect of Variation of Initiator Concentration

It may be seen from Table I that an increase in initiator, $K_2S_2O_8$ concentration results in a significant increase in percent grafting and grafting efficiency whereas total conversion for 3 h of grafting remains level at a high mean value ($94 \pm 4\%$). Concentration of $K_2S_2O_8$ beyond 0.04 mol L⁻¹ led to a high level of grafting at a relatively fast rate causing ready formation of a gel-like product. The grafted fibers get highly embedded and stuck in the thick gel of polyacrylamide thus formed, rendering their isolation and analysis very difficult.

Effect of Variation of Monomer Concentration

Examination of Table II reveals that the initiator and DAC content and other conditions remaining fixed, increase in acrylamide monomer concentration by lowering the liquid volume of the graft copolymerization system as described in Table II results in a progressive increase in percent grafting and grafting efficiency. The percent conversion level, however, remained at a high level (> 77%) showing a slow increasing trend followed by a leveling off trend with increase in monomer concentration. Here, changes in the substrate (DAC)/liquor ratio consequent to adjustment of volume of water should also be viewed as a contributing factor.

Effect of Variation of pH of the Reaction Medium

Table III shows that polymerization under neutral condition, i.e., at pH 7, is associated with a distinctive character exhibiting high conversion but significantly low percent grafting and grafting efficiency. Changeover to a slightly lower or higher pH led to some increase in percent conversion and substantial (1.8- to 2.0-fold) enhancement in percent

Table IV Graft Copolymerization of Acrylamide on DAC Initiated by $K_2S_2O_8$ in Limited Aqueous System following Pretreatment Technique: Effect of Variation of Temperature

Temperature (°C)	Gross Polymer (g)	Apparent Graft Copolymer (g)	Total Conversion (%)	Grafting (%)	Grafting Efficiency (%)
40	0.232	0.209	16	4.5	28.12
50	0.300	0.220	50	10.0	20.00
60	0.382	0.230	90	15.0	16.66
65	0.396	0.232	98	16.0	16.3
70	0.396	0.320	98	60.0	61.0

Acrylamide = 0.200 g, $K_2S_2O_8$ = 0.006 g, pH 5, time 3 h, cotton fiber (DAC, copper no. = 4) = 0.2 g.

Table V Graft Copolymerization of Acrylamide on DAC Initiated by $K_2S_2O_8$ in Limited Aqueous System following Pretreatment Technique: Effect of Variation of Time of Polymerization

Time (h)	Gross Polymer (g)	Apparent Graft Copolymer (g)	Total Conversion (%)	Grafting (%)	Grafting Efficiency (%)
1.5	0.280	0.205	40.0	2.5	6.0
2	0.320	0.210	60.0	5.0	8.3
2.5	0.352	0.222	76.0	11.0	14.4
3	0.377	0.234	88.5	17.0	19.2
4	0.398	0.238	96.5	19.0	19.6
5	0.403	0.301	100	50.7	50.7
6	0.400	0.318	100	58.0	58.5

Acrylamide = 0.2 g, $K_2S_2O_8$ = 0.006 g, temperature = 60°C, water = 3 mL, pH 5, cotton fiber (DAC, copper no. = 4.0) = 0.2 g.

grafting and grafting efficiency. A fairly low pH (pH < 3), i.e., a fairly high acidic condition is likely to cause notable or relatively severe hydrolytic degradation, mass loss, and hence weakening of the cellulose chains, more so over extended time periods of graft copolymerization, rendering the grafted fiber less suitable or unsuitable for textile application. Graft copolymerization of acrylamide over a pH range 3 and 4 resulted in relatively high percent grafting and grafting efficiency.

Effect of Variation of Temperature of Polymerization

Percent conversion increased sharply from 16 to 90% with the increase in temperature from 40 to 60°C after which it remained at a level as high as 98% (Table IV). Polymerization for 3 h at 70°C as in Table IV caused the whole polymer product to gel substantially, which on analysis showed very high percent grafting and grafting efficiency.

Effect of Variation of Time of Polymerization

Table V shows that percent conversion expectedly increased with time of polymerization with a leveling off trend to the maximum possible conversion after nearly 4 h of polymerization. The percent grafting and grafting efficiency, however, follow a steadily increasing trend up to about 4 h, beyond which there was a sudden jump in the values of these parameters (Table V). It is also interesting to note that the gross polymer product substantially becomes a gel when polymerization time is extended beyond 4 h.

Odd Effects and Mechanism

It may be said that whenever the polymerization of the water-soluble monomer AAm (giving a water-soluble polymer) was allowed to proceed relatively fast to a high conversion (90–95%) by increasing the initiator concentration, monomer concentration, temperature, or time of polymerization or by lowering of volume of solvent water, the reaction system

Table VI Graft Copolymerization of Acrylamide on DAC Initiated by $K_2S_2O_8$ in Limited Aqueous System following Pretreatment Technique: Effect of Variation of Substrate (Fabric) : Monomer Ratio (w/w) on Physical Properties of Fabric

Substrate/Monomer Ratio	Grafting (%)	Bending Length (cm)	Initial Modulus (N/mm)	Fabric Tenacity (N/mm)	Elongation at Break (%)
1/0.00	0.0	1.59 (1.52)	5.90 (5.70)	2.49 (2.50)	21.22 (20.45)
1/0.25	4.7	2.90	11.58	2.80	11.80
1/0.50	6.5 (3.6)	4.30 (2.97)	18.22 (10.70)	3.61 (2.50)	10.52 (16.31)
1/1.00	9.0	4.95	45.24	5.53	9.12
1/1.50	11.2	5.20	40.00	5.88	11.71

Cotton fabric (DAC, copper no. 4.0) = 2.5 g, $K_2S_2O_8$ = 50 mg, time 3 h, pH 10, temperature = 60°C, water = 5.0 to 30 mL. Values in the parenthesis indicate corresponding data for use of scoured cotton fabric in place of DAC fabric.

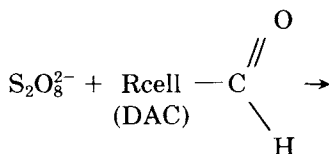
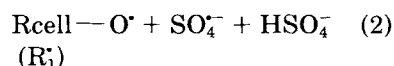
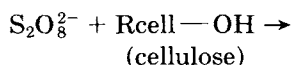
Table VII Graft Copolymerization of Acrylamide on DAC Initiated by $K_2S_2O_8$ in Limited Aqueous System following Pretreatment Technique: Effect of Variation of pH

pH of the Grafting Medium	Grafting (%)	Bending Length (cm)	Initial Modulus (N/mm)	Fabric Tenacity (N/mm)	Elongation at Break (%)
4	14.8	5.85	9.53	2.32	8.90
5	12.6	4.00	14.80	2.76	18.61
7	8.0	4.27	20.6	4.5	12.11
10	9.0	4.95	45.25	5.53	9.12
(Ungrafted DAC)	0.0	1.59	5.98	2.49	21.22

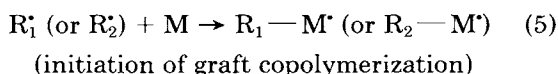
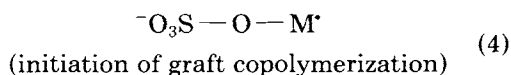
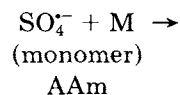
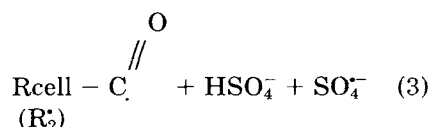
Cotton fabric (DAC of copper no. 4.0) fabrics = 2.5 g, $K_2S_2O_8$ = 50 mg, fabric : monomer (AAm) ratio = 1 : 1 (w/w), temperature = 60°C, time 3 h.

turned more into a swollen gel. Independent polymerization of AAm in the absence of cellulose or DAC under comparable conditions also substantiated this point. Polyacrylamide formed became progressively less soluble in water, presumably as a consequence of an uncertain gel effect or autoacceleration effect. Analysis of data in Tables I, II, IV, and V reveals that this additional uncertain effect becomes very prominent if the percent conversion in the limited aqueous medium goes beyond 96–98%. At this stage, high degrees of branching and crosslinking involving the polyacrylamide growing chain ends bound to the cellulose chains or otherwise come into play leading to large-scale immobilization and insolubilization of the polymer formed. The overall fall-out of this turn of events is reflected in the sudden jump in the percent grafting and grafting efficiency parameters (Tables IV and V).

The mechanism of graft copolymerization in the present case may be described by the following sequence of reactions:

**Table VIII Graft Copolymerization of Acrylamide on Cotton Fabrics Initiated by $K_2S_2O_8$ in Limited Aqueous System following Pretreatment Technique: Effect of Polyacrylamide Grafting on Dye Uptake (K/S Value) and Moisture Regain**

Grafting (%)	K/S Value of Dyed Cotton Fabrics		Moisture Regain (%)
	Reactive Dye (λ_{\max} 540 nm)	Acid Dye (λ_{\max} 480 nm)	
A. fabric used (DAC, copper number = 4.0)			
0.0	2.46	2.02	8.0
3.5	2.49	2.50	12.6
4.7	2.78	2.56	13.4
6.0	3.14	2.66	14.2
9.0	3.28	2.75	15.3
12.6	2.27	2.85	12.2
B. fabric used (scoured cotton, copper number = 0.24)			
0.0	2.30	1.83	—
3.6	2.37	2.24	—



Reaction (1) describes the primary radical generation step, reaction (4) shows initiation of homopolymerization of acrylamide, reactions (2) and (3) show the possible means of radical generation on cellulose and DAC chains by a redox mechanism, the cellulose/DAC chain units activating the decomposition of $\text{S}_2\text{O}_8^{2-}$ to radicals, and reaction (5) shows the major means of polyacrylamide grafting on cellulose/DAC chain segments. Between reactions (2) and (3), the latter is faster in view of higher reducing power of the aldehyde groups than the alcohol groups. At very high conversions, the gel effect assumes high significance and branching by chain transfer involving growing polyacrylamide chain radical ends and other polyacrylamide chains (causing true branching) or cellulose/DAC chain segments (causing further grafting), and coupling or crosslinking of two growing chains as in reactions (4) and (5) become increasingly/critically consequential leading to sudden rise in percent grafting and hence grafting efficiency.

Studies of Effect of Polyacrylamide Grafting on Properties of Cotton Cellulose/DAC Fabrics

Influences of variation of DAC (fabric) : monomer ratio (w/w) are shown in Table VI. Relevant data indicate that with progressive increase in monomer content from 0.25 to 1.5 g per gram of DAC fabric, grafting level increased from 4.7 to 11.2%. Progressive increase in percent grafting rendered the DAC fabrics increasingly stiff as shown by bending length data. The initial modulus value was also enhanced with enhancement of percent grafting with a leveling off trend beyond about 9% grafting corresponding to 1 : 1 (w/w) DAC-monomer ratio. Higher percent grafting led to an increasing trend in fabric tenacity. Elongation at break suffered a sharp fall initially on a low degree of grafting and it remained reasonably

level around 9–11% on further increase in percent grafting (Table VI).

Analysis of data in Table VI and those in Table VIII showing influence of variation of percent grafting on dye uptake (particularly using a reactive dye) and on moisture regain clearly reveals that grafting of polyacrylamide to a level of nearly 9% produced the most balanced effect in imparting prominently improved mechanical properties and moisture absorption on DAC fabric and at the same time rendering it more receptive to reactive and acid dyes. Polyacrylamide if incorporated by grafting to above 9–10% offers a prominent masking effect on the base polymer (DAC), thereby rendering it less available to dyes and moisture. This explains well a falling trend in dye uptake (using reactive dye) and moisture regain for percent grafting > 9%.

As for effect of variation of pH of the polymerization medium, data in Table VII reveal that polyacrylamide grafting effected in the pH range 7–10 produced optimum results in respect to mechanical properties of the fabric. Weakening of cotton cellulose/DAC due to hydrolytic degradation and mass-loss occurred substantially for graft copolymerization at $\text{pH} \leq 3$ (Table VIII).

CONCLUSION

Controlled low degrees (9–10%) of grafting of polyacrylamide on cotton fabric of low degree of oxidation (dialdehyde cellulose, DAC, copper number 4.0) induced by $\text{K}_2\text{S}_2\text{O}_8$ in a limited aqueous system at 60°C results in improved property parameters for the fabric, viz. dyeability with reactive and acid dyes, tenacity, and moisture regain. Concomitantly, the fabric gains much in stiffness (bending length) and modulus. Such modification enhances the prospect of the use of relevant grafted fabrics in such applications as stiff and durable lining for apparels, tarpaulins, canvas, beltings, and soft luggages.

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