



## Properties of coatings based on fluorine-containing self-crosslinking latexes

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*In this study, core-shell self-crosslinking latexes comprising different levels of copolymerized perfluoroethyl groups in the shell layer of latex particles were prepared by the emulsion polymerization of 2,2,2-trifluoroethyl methacrylate, methyl methacrylate, butyl acrylate, and methacrylic acid as main monomers. The effect of crosslinking (inter-particle self-crosslinking and intra-particle crosslinking) was evaluated with the aim to spare the fluorinated monomer consumption while maintaining the desired water-resistant coating properties. For achieving the inter-particle self-crosslinking, diacetone acrylamide was copolymerized into the shell layer of latex particles to provide sites for the subsequent reaction with adipic acid dihydrazide. The intra-particle crosslinking of latex particles was introduced by copolymerizing allylmethacrylate during the synthesis. The results showed that water sensitivity of coating films was reduced most notably by the level of crosslinking, while the effect of fluorine content in particle shell composition was minor only.*

**Keywords:** Latex; Fluoropolymer; Crosslinking; Water-whitening; Characterisation and properties

### Introduction

The research on the development of more effective and more environmentally friendly fluorinated polymer products is the mainstream in the field of modern

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fluorinated materials. Consequently, many researchers have devoted themselves to investigate and develop fluorinated acrylic latexes due to their special surface properties, good adhesion to matrices, and environmental protection value. These materials have been used especially as chemical resistant coatings for versatile protective applications.

Surface properties of protective coatings have a significant effect on their integrated performance and they are usually predominated by the architecture and chemical components of the outermost surface layer. In order to obtain enhanced hydrophobicity, the outermost surface layer should contain as many fluorine groups as possible [1]. However, a relatively high market price of fluorinated monomers limits their use and the challenge is then how to minimize the amount of fluorinated monomers whereas the reasonable properties of the resulting products can be maintained. One possible strategy to achieve the increased coating hydrophobicity is to prepare latexes based on the core-shell particles consisting of fluorine-free core and fluorine-rich shell [2–6]. The results gathered in the literature reveal that while the weight fraction of fluorine-containing compounds in the polymer decreases considerably, the improved physical and chemical properties can still be maintained [7,8]. Therefore, the core-shell fluorine-containing polymer latexes with low fluorine content can keep the excellent physical and chemical properties but also decrease the production expenses of fluorine-containing polymer latexes.

The increase of the crosslinking density is an effective tool to improve water resistance and mechanical properties of water borne emulsions used as coatings and adhesives. Methods for increasing the crosslinking density include intra-particle crosslinking of latex particles achieved during polymerization and inter-particle crosslinking during the latex film formation. Among different inter-particle crosslinking reactions, the keto-hydrazide self-crosslinking has been shown to be particularly effective when utilizing the reaction between carbonyl groups in diacetone acrylamide (DAAM) repeat units and hydrazide groups from adipic acid dihydrazide (ADH) [9–12]. The water soluble ADH is added to the latex after its preparation and, as a result, the one-component self-crosslinking latex ensuring long-term storage stability and a rapid ambient crosslinking can be easily obtained. Therefore, these latexes are popular in a wide range of applications, starting from paints for the building industry, through wood paints and varnishes, up to paints for metal protection and decorative systems.

This paper focuses on the effect of crosslinking on wettability, water sensitivity, and coating properties of latex coating films originated from particles with perfluorethyl groups in the shell layer. The presented work also demonstrates the optimal composition based on the self-crosslinking latex system with the emphasis on sparing the fluorinated monomer consumption while maintaining the desired water-resistant properties.

## Materials and methods

### Materials

Latexes were synthesized of 2,2,2-trifluoroethyl methacrylate (TFEMA), methyl methacrylate (MMA), butyl acrylate (BA), methacrylic acid (MAA), allyl methacrylate (AMA), and diacetone acrylamide (DAAM); all the monomers being purchased from Sigma-Aldrich. Disponil FES 993 (BASF, Czech Republic) was used as the surfactant and ammonium persulfate (Penta, Czech Republic) utilized as the initiator of the polymerization reaction. Adipic acid dihydrazide (ADH) served as the crosslinker when purchased from Sigma-Aldrich. All the chemicals were used as received without any further purification.

### Preparation and characterization of latexes

Two series of acrylic latexes of core-shell particles were synthesized by the semi-continuous non-seeded emulsion polymerization comprising a variable content of acrylic monomers (see Table 1). The core/shell weight ratio of the particles was 1:1, representing a shell thickness of about 20 % of the particle radius.

**Table 1** *Composition and characteristics of latexes differing in the way and the extent of crosslinking and TFEMA concentration in monomer feeds forming the shell layer of latex particles.*

Sample	Composition of monomer feeds [wt. %]		Particle size in the water phase [nm]		Gel content [wt. %]
	Core	Shell	Core	Core-shell	
<i>Series A: Self-crosslinking</i>					
A1	0/44/53/3/0/0	0/39.5/52.5/3/5/0	80.2 ± 0.8	103.8 ± 0.6	77.8
A2	0/44/53/3/0/0	15/27/50/3/5/0	114.4 ± 0.6	143.1 ± 0.9	81.5
A3	0/44/53/3/0/0	30/14/48/3/5/0	126.4 ± 0.3	158.5 ± 1.1	79.1
A4	0/44/53/3/0/0	45/1/46/3/5/0	103.2 ± 0.6	128.1 ± 0.4	78.9
<i>Series B: Self-crosslinking and inter-particle crosslinking</i>					
B1	0/43.4/52.6/3/0/1	0/39/52/3/5/1	80.0 ± 0.9	101.4 ± 0.5	98.6
B2	0/43.4/52.6/3/0/1	15/26/50/3/5/1	84.7 ± 0.7	102.2 ± 0.7	99.1
B3	0/43.4/52.6/3/0/1	30/13/48/3/5/1	88.5 ± 0.5	112.4 ± 0.6	98.2
B4	0/43.4/52.6/3/0/1	45/0.5/45.5/3/5/1	86.4 ± 0.4	110.9 ± 0.3	98.9

The nature of acrylic monomers forming core and shell layers was chosen to achieve a calculated  $T_g$  (using the Fox equation [13]) of approximately 10 °C. In the case of both series, TFEMA was copolymerized with the monomers designed to form the shell layer of latex particles and its concentration was gradually increased. In the series A, the latexes were designed to provide the keto-hydrazide self-crosslinking during the film-formation. The series B was represented by latexes being able to produce highly crosslinked coating films by means of the combination of both the inter-particle crosslinking and the keto-hydrazide self-crosslinking. The inter-particle crosslinking of latex films was achieved by covalent crosslinking of emulsion copolymers during their synthesis using AMA. For the keto-hydrazide self-crosslinking, a constant amount of DAAM was copolymerized into the shell layer of latex particles to provide sites for the subsequent interfacial covalent crosslinking by the reaction with ADH being added during latex coating formulation.

The latexes were produced in a 700 mL glass reactor by the semi-continuous non-seeded emulsion polymerization under starved conditions in the nitrogen atmosphere at 85 °C. This procedure ensured relatively homogeneous latex particles of statistical copolymers. The reactor charge was put into the reactor and heated to the polymerization temperature. Then, the monomer emulsion was fed into the stirred reactor at feeding rate about 2 mL min<sup>-1</sup> in two steps (1. core preparation, 2. shell preparation). Afterwards, during 2 hours of hold period at 85 °C, the polymerization was completed. A recipe for emulsion polymerization is shown in Table 2.

**Table 2** *Specification of conditions for emulsion polymerization*

	Reactor charge	Monomer emulsion (Core)	Monomer emulsion (Shell)
Water [g]	120	124	124
Disponil FES 993 [g]	0.5	7.4	7.4
Ammonium persulfate [g]	0.4	0.4	0.4
Monomers [g]	–	100	100

The pH was adjusted to 8.5 with an ammonia solution. To ensure the keto-hydrazide self-crosslinking of latexes, a 10 wt. % aqueous solution of ADH in the amount corresponding to the molar ratio ADH:DAAM = 1:2, was added to the latex under agitation. The content of solids in the final latexes was about 35 wt. %.

The average particle sizes of latex particles in the water phase were determined by dynamic light scattering experiments performed using a Coulter instrument (model "N4 Plus", Coulter; UK). All the DLS measurements were conducted at 25 °C.

The degree of crosslinking introduced into latex coating films was evaluated based on gel content changes. Specimens were prepared by pouring and drying the latexes on silicone substrates. Such specimens were first air-dried at room temperature (RT,  $23 \pm 1$  °C) for a month and then vacuum-dried at 30 °C for 2 weeks. The gel content was determined according to CSN EN ISO 6427 using a 24-hour extraction with THF in a Soxhlet extractor. Around 1 g of the dried latex sample was put into a thimble. After extraction, the thimble was dried in an oven at 75 °C for 12 h, cooled in a desiccator overnight, and the gel content calculated from the initial and final weights, assuming that the gelled material remained captured in the thimble.

### Preparation and characterization of coatings

The self-crosslinking latexes were applied on glass and low carbon steel panels using a blade applicator. The thickness of wet coating films was 120 µm. No coalescing agents were needed. Coatings were air-dried at RT and  $45 \pm 5$  % relative humidity (RH) for 7 days. The resulting coating films were evaluated for their gloss, hardness, adhesion, impact resistance, water absorption water-whitening, and wettability. The gloss of coating films cast on glass panels coated with a black matte paint (RAL 9005) was determined by a micro TRI-gloss µ instrument (BYK-Gardner, Germany) using a gloss-measuring geometry 60°. The hardness of coatings was measured by a pendulum hardness tester "Persoz" (BYK-Gardner, Germany) according to CSN EN ISO 1522. The adhesion of coating films toward a glass substrate was determined according to CSN ISO 2409 using a cross-cut tester (Elcometer Instruments, UK). The impact resistance was evaluated using an Elcometer 1615 Variable Impact Tester (Elcometer Instruments, UK) following CSN EN ISO 6272; all experiments being carried out at RT.

For evaluating the water absorption of latex coating films, the respective specimens were prepared by pouring the latexes into a silicone mold. Films were allowed to dry out at RT for a month. The water absorption by latex films was measured by immersing the samples of the approximate dimensions  $20 \times 20 \times 1$  mm<sup>3</sup> into distilled water at RT. The water absorption,  $A$  is given by  $A = 100 (w_t - w_0)/w_0$ , where  $w_0$  is the sample weight before immersion and  $w_t$  is the sample weight after water immersion for a given time period. The swollen films were carefully removed from water, and water from the film surface wiped by touching the polymer with a filter paper.

The water whitening of dried coating films (120 µm wet thickness) draw-downed on glass panels was evaluated objectively by measuring the change in transmittance at a fixed wavelength (500 nm, near the green light that is most sensitive to the human eye) using a ColorQuest XE Spectrometer (Hunterlab,

USA). Before the measurement, coating films were allowed to dry out at RT and 33% RH for two weeks. After that, the films were immersed in distilled water at RT for 1, 2, 3, 4 and 24 hours, respectively, followed by the immediate measurement of the transmittance of the exposed coating film area. The extent of water whitening,  $W = 100 (T_0 - T_t)/T_0$ , where  $T_0$  is the sample transmittance before exposure to distilled water and  $T_t$  is the sample transmittance after performing the immersion test.

Wettability studies had involved the measurements of water contact angles using an optical tensiometer Attension Theta (Biolion Scientific, Finland) by the sessile drop method. An automatic micropipette was used to accurately dispense the volume (1  $\mu$ L) of water. The time dependence of the water drop flowing (the change of contact angle) was recorded by a video camera and the steady-state contact angle value subtracted in the time period of 10 s. Measurements were made at 22 °C and humidity of  $45 \pm 5$  %. A OneAttension software (Biolion Scientific, Finland) was used to image data recorded and for the determination of water contact angles. Five measurements were performed for each sample and the accuracy of the constant angle measurement estimated to be  $\pm 1^\circ$ .

For the wettability measurements, the coating films were draw-downed on glass panels using a blade applicator. Their wet thickness was 120  $\mu$ m and no coalescing agents were used. The coating samples were dried for two weeks in a hermetic chamber with fixed RH of 53 % (using saturated solution of magnesium nitrate). The humidity in the chamber was checked by a thermohygrometer (T2041, Aldrich), when ranging within  $\pm 2$  %.

## Results and discussion

### Properties of latexes

Latexes with negligible amount of coagulum (0–0.2 %) were synthesized by the semi-continuous non-seeded emulsion polymerization process with varying amount of TFEMA in shell compositions. All the prepared latexes were stable for over 12 months. To prove the core-shell morphology of the latex particles, the average diameters of core particles (for samples taken after finishing the core polymerization step) and the resulting core-shell particles were determined using DLS (see Table 1). When comparing the results between core and core-shell particles for the individual samples, they were in a good accordance with the calculated dimension of the shell thickness being about 20 % of the particle radius.

The level of crosslinking introduced into latex particles and coating films was assessed according to gel content values (see Table 1). It was shown that the latexes of series A were found to produce crosslinked polymer materials of gel content around 80 wt. % owing to the keto-hydrazide crosslinking, whereas the latexes of series B provided almost completely crosslinked materials (gel content about 99 %) due to covalent inter-particle and intra-particle crosslinking reactions.

### Properties of coatings

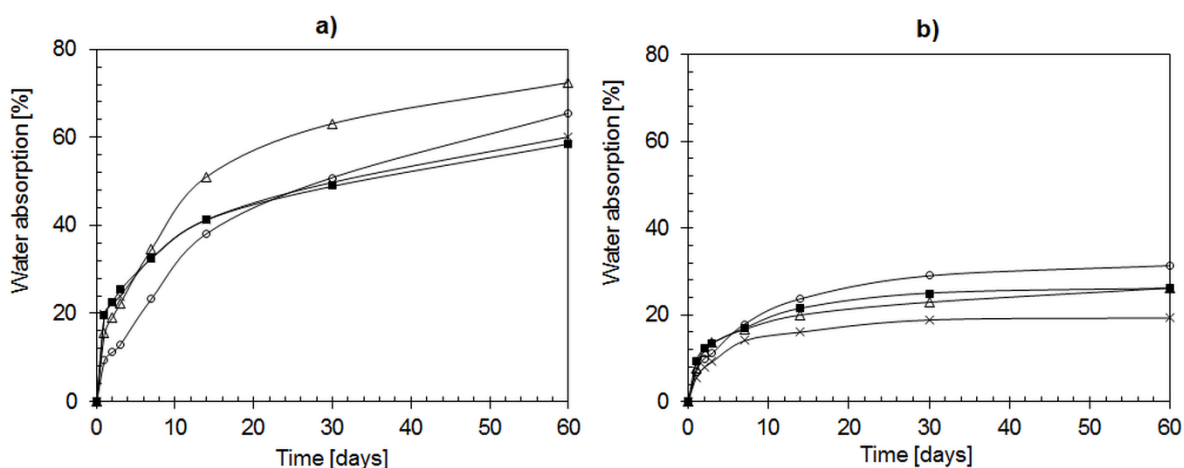
The effect crosslinking and TFEMA content in the shell layer of latex particles on final coating properties is presented in Table 3. All the latexes were able to form high-quality transparent films. Dry film thickness of the coatings was approximately 60  $\mu\text{m}$ . It has been shown that coating films of series B had exhibited increased hardness, which can be attributed to the increased network density of the resulting polymer materials due to intra-particle crosslinking. Further, all the tested coatings also exhibited a high gloss and superior adhesion to the glass and steel substrates. Also, the impact resistance of the resulting coatings was shown to reach maximum values suggesting us the formation of crosslinked and elastic polymer materials in which sufficient coalescence of latex particles had been achieved before the inter-particle self-crosslinking took place.

**Table 3** Comparison of properties of coating films draw-downed from latexes differing in the extent of crosslinking and concentration of fluorine groups in shell layer of latex particles

Sample	TFEMA content in shell [wt. %]	Gloss [%]	Hardness [%]	Adhesion [degree of flaking]		Impact resistance [cm]
				Glass	Steel	
<i>Series A: Self-crosslinking</i>						
A1	0	86.3 $\pm$ 0.5	29.7 $\pm$ 1.3	0*	0*	above 100*
A2	15	79.9 $\pm$ 0.6	32.3 $\pm$ 1.0	0	0	above 100
A3	30	81.3 $\pm$ 0.5	30.7 $\pm$ 0.9	0	0	above 100
A4	45	787.8 $\pm$ 0.8	30.1 $\pm$ 0.6	0	0	above 100
<i>Series B: Self-crosslinking and inter-particle crosslinking</i>						
B1	0	95.3 $\pm$ 0.9	39.7 $\pm$ 1.0	0	0	above 100
B2	15	92.1 $\pm$ 1.1	40.8 $\pm$ 0.7	0	0	above 100
B3	30	85.6 $\pm$ 0.9	40.5 $\pm$ 0.4	0	0	above 100
B4	45	88.9 $\pm$ 0.1	38.9 $\pm$ 1.1	0	0	above 100

\* Maximal attainable value (representing the best property).

Water sensitivity of latex coating films was evaluated based on their water absorption and water whitening. The results of water absorption measurements are illustrated in Fig. 1. It is obvious that similar water absorption trends with respect to the fluorine content can be observed for both series of latex coatings, when water uptake has been found to be reduced with the increasing level of TFEMA units in the particle shell in the case of performing long-term water absorption experiments. The results of short-term water absorption revealed that fluorine-containing coatings exhibited a greater water uptake in comparison with the fluorine-free samples. This finding indicated a worse coalescence of the fluorine-containing latex particles, assuming that the increased hydrophobicity of particles surface could induce the formation of larger interstitial areas and/or channels filled with highly hydrophilic components (surfactant and initiator). Thus, a more porous film structure was probably formed and, thus, the initial speed of water absorption facilitated.



**Fig. 1** Water absorption into latex coatings

a) coating films of series A, b) coating films of series B;

○ curves – materials synthesized without TFEMA in shell layer of latex particles,

■ curves – materials synthesized using 15 wt. % of TFEMA in shell monomer composition,

△ curves – materials synthesized with 30 wt. % of TFEMA in shell monomer composition and

× curves – materials synthesized using 45 wt. % of TFEMA in monomer composition of the shell layer of latex particles.

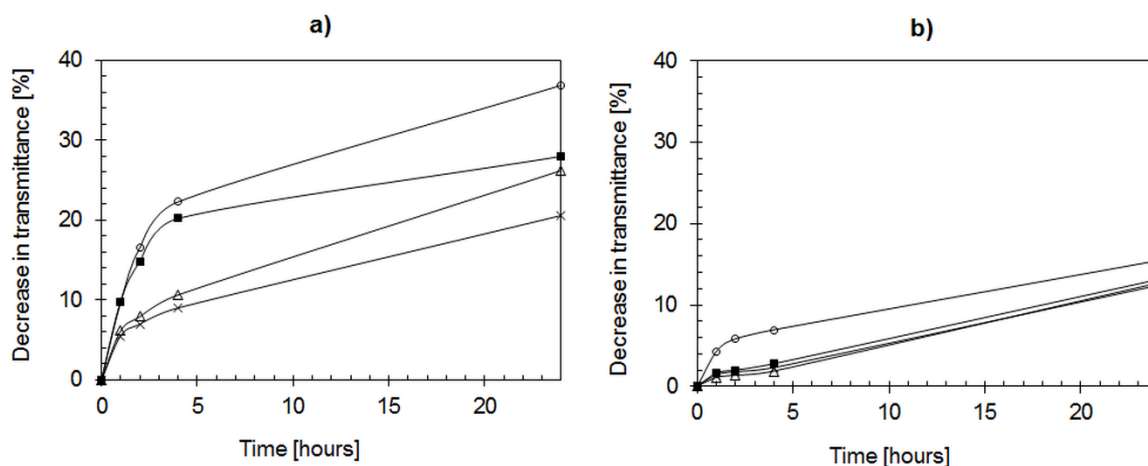
The standard deviations of performed measurements did not exceed 3 %.

Also, it has been shown that water absorption of latex films was affected most notably by the level of crosslinking with a relation: the more crosslinked film, the smaller water uptake. This phenomenon could be seen clearly especially in the case of long-term water absorption experiments. The reason for the suppressed water swelling in the case of crosslinked latex films was well described in literature. Kessel et al. [14] studied the competing effects of keto-hydrazide crosslinking and interdiffusion in waterborne systems finding that crosslinking inhibited particle flattening and retarded interdiffusion and



entanglements formation. As the result, a porous film structure was formed and the extraction of ionically-charged components by water facilitated, which led to the reduction of the osmotic pressure (as the main driving force for water absorption). Moreover, the decreased water uptake of crosslinked latex coatings was also associated with the enhanced stiffness of crosslinked polymer, restricting the influx of water and not allowing water domains to grow.

Furthermore, water whitening of dried coating films was evaluated according to changes in transmittance (Fig. 2). The phenomenon of water whitening is believed to be caused by water entrapped inside the coating film. The opacity comes from water scattering centers of appropriate size within the film. It is well known that the size and number of the water domains are responsible for the water whitening effect and both can be restricted by the stiffness of the polymer. Therefore, films suffering from high water swelling are prone to enhanced water whitening as well [15,16]. Also, in this case, the experiments confirmed that results of water whitening exhibited similar trends to water absorption results, i.e. water whitening was found to be reduced with the increasing level of TFEMA units in particle shell and primarily, by the level of crosslinking. Thus, latex compositions combining self-crosslinking and intra-particle covalent crosslinking (series B) gave rise to more water-whitening resistant coatings. The effect of fluorine content in particle shell composition was shown to be only minor in the case of both series.



**Fig. 2** Water whitening expressed in terms of transmittance decrease for coating films cast on glass panels after their immersion in distilled water.

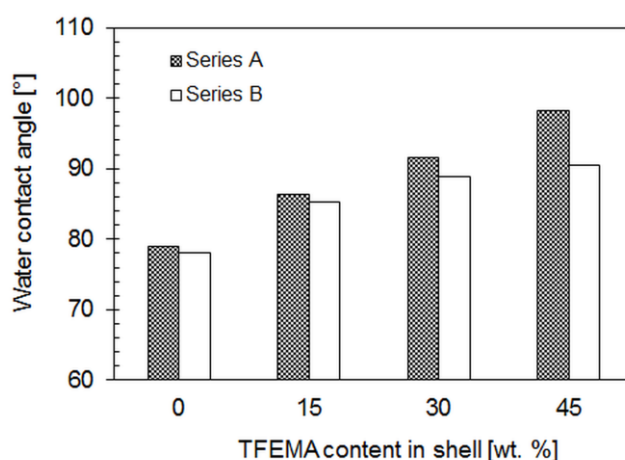
Transmittance values were measured at 500 nm.

a) coating films of series A, b) coating films of series B;

○ curves – materials synthesized without TFEMA in the shell layer of latex particles,  
 ■ curves – materials synthesized using 15 wt. % of TFEMA in shell monomer composition,  
 △ curves – materials synthesized with 30 wt. % of TFEMA in shell monomer composition and  
 × curves – materials synthesized using 45 wt. % of TFEMA in monomer composition of shell layer of latex particles.

The standard deviations of performed measurements did not exceed 5 %.

Next, the combined effects of crosslinking and TFEMA content on coating wettability were also evaluated and the results of water contact angles measurements are presented in Fig. 3. It was confirmed that the wettability of coatings decreased by increasing the content of TFEMA in the shell layer of latex particles and showing the effect by the level of crosslinking: more crosslinked latex films of series B manifested a lower effect of fluorine content in the particle shell on coating water-repellent properties compared to the coatings of series A. This phenomenon can be attributed to the fixation of latex polymer by crosslinking, which hindered the orientation of higher amount of perfluoroethyl groups towards the film-air interface during film-formation.



**Fig. 3** Dependence of wettability of latex coatings on TFEMA content in the shell layer of latex particles

## Conclusion

The present study deals with the properties of coatings based on self-crosslinking latexes differing in perfluoroethyl group concentration in the shell layer of latex particles. The effect of crosslinking was evaluated with the aim to spare the fluorinated monomer consumption while maintaining the desired water resistant properties. As shown, the combination of keto-hydrazide inter-particle self-crosslinking with covalent intra-particle crosslinking was an effective tool to provide coatings with enhanced resistance to water whitening, while the effect of fluorine content in particle shell composition was minor only. It can be concluded that the intra-particle crosslinked self-crosslinking latexes comprising 15 wt. % of 2,2,2-trifluoroethylmethacrylate in shell layer of latex particles provide transparent coatings with excellent mechanical properties and high water whitening resistance, which may find a potential use in the coating industry as clear coats or binders for exterior and interior material protection.

## Acknowledgement

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