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IMAGE-RECEIVING MATERIAL FOR DYE DIFFUSION THERMAL TRANSFER

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(56) Prior Art Documents
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(57) Claim

1. An image-receiving material for dye diffusion thermal transfer comprising a resin-coated base paper and an image-receiving layer formed on the front side of said base paper and comprised of a dye-receiving resin, wherein the dye-receiving resin is a combination of at least one acrylate copolymer of monomers containing polar groups and an oxidized polyethylene.

13. A process for the manufacture of an image-receiving material for dye diffusion thermal transfer with a receiving layer containing dye-receiving resin applied to the front side of a resin-coated base paper, wherein the receiving layer comprising a combination of at least one acrylate copolymer containing polar groups and an oxidized polyethylene is applied as an aqueous composition onto the front side of the resin-coated base paper in a one-step process.

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COMPLETE SPECIFICATION

(ORIGINAL)

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Complete Specification for the invention entitled:

"IMAGE-RECEIVING MATERIAL FOR DYE DIFFUSION THERMAL TRANSFER"

The following statement is a full description of this invention including the best method of performing it known to us/~~me~~:-

The invention relates to an image-receiving material for dye diffusion thermal transfer as well as a process for its manufacture.

A system of dye diffusion thermal transfer ("D2T2") has been developed in recent years which makes possible the reproduction of an electronically created picture in the form of a "hardcopy".

The principle of D2T2 is that, with regard to the basic colours cyan, magenta red, yellow and black, the digital picture is encoded into electrical signals which are then transmitted to a thermal printer and translated into heat.

The dye of the donor layer of a dye transfer band/sheet which is in contact with the receiving material sublimates under the effect of heat and diffuses into the receiving layer.

As a rule, a receiving material for dye diffusion thermal transfer consists of a support material with a receiving layer applied to its front side. Other layers may be applied onto the front side additionally such as barrier-, release-, adhesive- and protective layers.

The necessity of such additional coatings is required by the demands placed upon the receiving material. These may be:

- a smooth surface
- heat and pressure stability
- light stability (no yellowing)
- good dye solvency
- good anti-scratch and abrasion characteristics
- "anti-blocking" characteristics (no sticking)

Either plastic foils such as polyester film or coated paper may serve as the support material.

The main component of the receiving layer is, as a rule, a thermoplastic resin showing an affinity to the dye contained in the dye transfer band. Materials suitable for such may be linear polyester, e.g. polyethylene terephthalate, polybutylene terephthalate or acrylic resins, e.g. polymethylmethacrylate, polybutylmethacrylate, polymethylacrylate etc.. Furthermore such materials as polystyrene, polycarbonate, polyvinyl pyrrolidone, ethyl cellulose, polysulfone and other polymers may be used as dye-receiving resins.

The American Patents US 4 748 150 and US 4 774 224 show that polycarbonate may be used as a receiving layer on a polyethylene-coated base paper. Moreover an intermediate layer is applied between the support material and the receiving layer. This intermediate layer is a vinyliden-chloride-copolymer and serves to improve adhesion between the receiving layer and the support material.
The above-mentioned receiving sheet has shown itself to be disadvantageous as the polycarbonate used shows a strong tendency to yellowing and in time affects the transferred picture negatively. A further disadvantage is that both coats must be applied using solvent agents which can lead to health-, and safety-problems.

The problem of pressure sensitivity of the receiving sheet when in contact with the heating head has been dealt with in the European Patent Application EP 0 288 193. This pressure sensitivity makes itself shown in a reduction of the surface gloss of the layer or in the phenomenon "strike-through" in which an impression of the picture can be seen on the reverse side of the receiving sheet.

The problem is solved by applying a release layer based on silicone with a SiO_2-additive onto a polyester receiving layer which has been coextruded onto a polyester support material. The disadvantage of the above-mentioned is that, probably as a result of a reaction between the reacting groups of the silicone compounds and the diffused dyes in the receiving layer, the picture is blurred. Furthermore, the similarity to a photo, as required by the market, is missing with pictures produced in this manner.

It is furthermore a fact that dye issuing from the dye-donor band and diffused into the receiving layer tends to pale under the influence or light. This problem has been dealt with in the US Patent 4 775 657 in so much as the receiving layer, consisting of polycarbonate, is coated with a protective coating of polyester or polyurethane. A disadvantage of material so produced is the pressure sensitivity of the receiving sheet as well as the necessity of several work operations and the necessity of using organic solvents during coating.

The Patent Application EP 0 261 970 describes a receiving layer containing a linear saturated polyester as a binding agent and a silica coupled silane copolymer as a release agent (anti-blocking additive).

The object of this invention therefore is to provide a receiving material for dye diffusion thermal transfer procedures which does not show the disadvantages as mentioned above, i.e. it must exhibit good heat and light proof characteristics as well as being impervious to pressure and demonstrates good flatness and anti-blocking characteristics. Moreover, the receiving material represents a further improvement of colour density and colour gradation compared to the receiving sheets already on the market.

The object of the invention is accomplished by coating ~~the~~ a front side of a ~~polyolefine~~ resin-coated base paper with a receiving coating mass which, as a dye-receiving resin, contains a combination of at least one acrylate-copolymer containing polar groups and oxidized polyethylene.

It was surprising to find that the utilization of the above-mentioned combination created a receiving material which not only met the requirements as previously listed but at the same time enabled a high colour density of the printed image as well as improving colour gradation.

In a preferred embodiment of the invention an acrylate-copolymer whose polar groups are carboxyl-, metal-combined carboxyl groups and/or nitrile groups was utilized. Zinc-combined carboxyl groups are especially to be preferred in the metal-combined groups.

Acrylnitrile and/or methacrylic acid take part in the structure of the acrylate copolymer using in the combination according to the invention and the amounts of these monomers in the copolymer are between 10 an 40 mol.-%. In a preferred embodiment the amounts of these monomers are between 25 and 35 mol-%.

The acrylate copolymer compound may, additionally, contain styrene in an amount of up to 40 mol.-%.

The weight relationship of the acrylate copolymer to the oxidized polyethylene in the combination according to the invention may be between 99:1 and 30:70. The best results, as far as colour density and colour gradation are concerned, were obtained with acrylate copolymer/oxidized polyethylene weight relationship of between 70:30 and 40:60 (see example 2, table 2).

The receiving layer for the receiving sheet according to the invention may contain, as well as the dye-receiving resin, fine-particled silica or Al_2O_3 as a matting agent or further additives, such fluorine tenside as wetting agents, dispersing agents, dye-couplers, UV stabilisers, pigments and other auxiliary agents.

The coating mass for the receiving layer may be applied using any of the usual procedures for coating and dosing such as roll-, gravure-, nipp-coating, air brushing or wire bar onto a substrate as for instance polyethylene-coated paper.

The receiving coating may be applied from an aqueous form in a one-step operation.

The coating weight of the receiving layer may be between 0,3 - 15 g/sq.m. but 1 - 10 g/sq.m.is preferrable.

The base paper or support material is coated with a resin; for example, a polyolefine such as polyethylene, or a polycarbonate. The support material may be coated on one but preferably both sides. The side of the resin coated base material which is subsequently coated with the image-receiving layer is referred to throughout this specification and claims as the "front side".

As a support material, a paper with at least one side coated with a polyolefine such as polyethylene is preferred, wherein this polyolefine layer applied in accordance to the available coating technology has a basis weight of more than 5 g/sq.m., preferably between 7 - 25 g/sq.m.

The polyolefine layer may contain pigments and other additives.

The invention is illustrated in the following examples although this in no way sets limits.

EXAMPLES

Example 1

The front side of a polyethylene-coated base paper *) was coated with an aqueous dispersion of the following content:

Product	content, %-weight			
	1A	1B	1C	1D**)
Acrylate-Copolymer I, 40 % aqueous dispersion	96,0			96,0
Acrylate-Copolymer II, 40 % aqueous dispersion		96,0		
Acrylate-Copolymer III, 38 % aqueous dispersion			96,0	
Fluorine tenside, 1 % in water	4,0	4,0	4,0	4,0
Coating weight, g/sq.m.	5,0	5,0	5,0	5,0

**) no titan dioxide in the polyethylene coating

other test conditions:

- Machine speed : 130 m/minute
- Drying temperature : 110 ° C
- Drying time : 10 sec.

*) The base paper with the basis weight of 180 g/sq.m. was coated on both sides with polyethylene.

The back side of the base paper was coated with clear polyethylene, a mixture of LDPE and HDPE (35 % HDPE with a density of d = 0,959 g/cm^3, MFI = 8; 28 % HDPE with d = 0,950 g/cm^3, MFI = 7; 20 % LDPE with d = 0,934 g/cm^3, MFI = 3; 17 % LDPE with d = 0,915 g/cm^3, MFI = 8) at a coating weight of between 14 - 15 g/sq.m.

The front side was coated with a mixture of pigmented polyethylene (19 % HDPE with d = 0,959 g/cm^3, MFI = 8; 20 % LDPE with d= 0,934 g/cm^3, MFI = 3; 13,3 % LDPE with d = 0, 915 g/cm^3, MFI = 8; 26,7 % LDPE with d = 0,924 g/cm^3, MFI = 4,5; 21 % TiO_2 masterbatch with a 50 % TiO_2 content) at a coating weight of 15 g/sq.m.

The acrylate copolymers were copolymers in whose structure polar group containing monomers of the following content were used:

Acrylate-copolymer I (e.g. Primal HG-44 from Rohm & Haas Ltd.)	-	35 mol.-%
Acrylate-copolymer II (e.g. Maincote HG-54 from Rohm & Haas Ltd.)	-	30 mol.-%

An acrylate/styrene-copolymer (e.g. NeoCryl SR-205 from Polyvinyl-Chemie Ltd. Holland) containing zinc-combined carboxyl-groups was used as acrylate-copolymer III.

A fluorine tenside (e.g. FT-248 from Bayer AG was used as a wetting agent.

The receiving material was printed on using the dye diffusion thermal transfer method and subsequently analysed. The results may be seen as compiled in table 1.

This example is designed to show the exceptional suitability of the above-mentioned acrylate copolymers as components of the inventions's receiving layer.

Example 2

A support material as in example 1 except for the front side being polyethylene coated at 7 g/sq.m., was coated with an aqueous dispersion of the following content:

Product	Content, per cent weight -						
	2A	2B	2C	2D	2E	2F	2G
Acrylate-copolymer I 40 % aqueous dispersion (as example 1)	91,7	61,4	41,4	-	41,4	41,4	-
Acrylate-copolymer III 38 % aqueous dispersion (as in example 1)	-	-	-	-	-	-	42,4
Oxidized polyethylene 30 % aqueous dispersion (e.g. Südranol 340, from Süddeutsche Emulsions-chemie GmbH)	5,1	35,0	55,2	96,0	55,2	55,2	53,7
1 % Fluorine tenside in water (as example 2)	3,2	3,6	3,4	4,0	3,4	3,4	3,9
coating weight g/sq.m.	5,0	5,0	5,0	5,0	10,0	0,5	5,0

All other rest conditions were identical to example 1.

The results of the tests of the following printed pictures are to be seen in table 2.

Example 3

A support material as in example 1 was coated with an aqueous dispersion of the following content:

Produkt	Content, per cent weight		
	3A	3B	3C
Acrylate-copolymer I 40 % aqueous dispersion (as example 1)	53,8	48,4	42,7
Oxidized polyethylene 30 % aqueous dispersion (as example 2)	27,6	32,3	28,4
Silica 15 % in water (e.g. Syloid ED 50, from Grace GmbH)	11,1	--	--
Al_2O_3, 63 % slurry (e.g. Martifin OL-008, from Martinswerk)	--	15,3	13,5
Titan dioxide, 40 % in water (e.g. Rutil RN40 from Kronos Titan)	4,1	--	--
UV-absorber, 15 % in water (e.g. Tinuvin 213 from Ciba-Geigy AG)	--	--	11,8
Fluorine tenside, 1 % in water (as example 1)	3,4	4,0	3,6
coating weight g/sq.m.	5,0	5,0	5,0

All other test conditions were identical to example 1.

The test results are compiled in table 3.

Comparative Examples

V1. The test was carried out as example 1. The receiving layer was applied in an aqueous form of the following content:

Product	Content, per cent weight	
	V1 A	V1 B
Acrylate-copolymer IV, 50 % aqueous dispersion	96,0	--
Acrylate-copolymer V, 40 % aqueous dispersion	--	96,0
Fluorine tenside, 1 % in water (as example 1)	4,0	4,0
coating weight, g/sq.m.	5,0	5,0

The acrylate copolymers were copolymers in whose structures polar groups containing monomers of the following content were used:

Acrylate copolymer IV - 9 mol.-%
(e.g. Primal P 376 from
Rohm & Haas Company)

Acrylate copolymer V - 7 mol.-%
(e.g. Primal WL 91 K from
Rohm & Haas Company)

The receiving material so produced was then printed upon by means of dye diffusion thermal transfer and then analysed. The results are compiled in table 4.

V2. The test was carried out as example 1. An acryl resin such as polyethyl-acrylate (e.g. Plextol B 408 from Röhm Ltd.) was used as dye-receiving resin.

The printed pictures so produced (hard copy) were tested and the results of the test may be seen in table 4.

V3. For comparison purposes, Hitachi image-receiving material as can be found on the market, was used. The results may be seen in table 4.

Testing of the Image Receiving Material Produced as per Examples 1 - 4

The receiving material produced underwent dye diffusion thermal transfer.

A Hitachi colour video printer model VY - 25 E together with Hitachi dye ribbon was used. The technical details of the video printer are as follows:

Video memory	:	Pal 1-full-image memory
Printed image	:	64 colour shade image
		image elements: 540 : 620 dots
Printing time	:	2 minutes/print

The prints so produced (hard copies) were investigated for their colour density and anti-blocking characteristics.

The density measurements were taken before and after a 24-hour exposure of the prints to a Xenon-lamp. The loss of density thereby caused was measured using Δ d (%) as an evaluation of the light stability.

The equipment used here was an Original Reflection Densitometer SOS-45. The measurements were taken in five colour gradations from F1-F5 for the basic colours cyan, magenta, yellow and black whereby the values for F1, F3 and F5 are given in the tables. The number of possible colour gradations from 0-7 is likewise to be found in the tables.

At the same time comparative measurements were taken from receiving materials from the market.

The results to be found in tables 1-4 show that the receiving material manufactured according to the invention and the images printed on it reflect higher values of colour density and colour gradation in every colour range.

The light stability (Δ d-values) also show better values from the material produced according to the invention as do the comparison materials used.

Table 1: Characteristics of the Printed Image Receiving Material Produced according to Example 1

Example	F	Colour density d								Gradation	"anti-" blocking characteristics
		cyan		magenta		yellow		black			
		a	b	a	b	a	b	a	b		
1 A	F 1	1,54	1,40	1,29	1,19	1,43	1,39	1,39	1,46	6	good
	F 3	0,24	0,12	0,23	0,13	0,23	0,20	0,28	0,22		
	F 5	0,09	–	0,09	–	0,08	–	0,11	–		
1 B	F 1	1,64	1,49	1,31	1,24	1,48	1,45	1,47	1,52	6	good
	F 3	0,28	0,16	0,22	0,15	0,25	0,23	0,29	0,24		
	F 5	0,11	–	0,09	–	0,08	–	0,11	–		
1 C	F 1	1,38	1,25	1,15	1,06	1,14	1,11	1,30	1,29	6	good
	F 3	0,15	0,08	0,18	0,10	0,15	0,13	0,21	0,18		
	F 5	0,03	–	0,06	–	0,04	–	0,08	–		
1 D	F 1	1,51	1,40	1,26	1,15	1,42	1,36	1,37	1,38	6	good
	F 3	0,27	0,17	0,23	0,14	0,21	0,18	0,28	0,24		
	F 5	0,14	–	0,12	–	0,8	–	0,13	–		

a - before exposure to Xenon-lamp
b - after 24 h-exposure to Xenon-lamp

Table 2 Characteristics of the Printed Image Receiving Material Produced according to Example 2

Example	F	Colour density d												Gradation	"anti-" blocking characteristics
		cyan			magenta			yellow			black				
		a	b	d %	a	b	d %	a	b	d %	a	b	d %		
2 A	F1	1,72	1,43	16,8	1,36	1,24	8,8	1,55	1,49	3,9	1,49	1,51	0	7	good
	F3	0,30	0,23	23,3	0,24	0,20	16,7	0,29	0,22	24,1	0,33	0,26	21,2		
	F5	0,12	–	–	0,10	–	–	0,12	–	–	0,13	–	–		
2 B	F1	1,89	1,57	16,9	1,48	1,40	5,4	1,66	1,46	12,1	1,61	1,65	0	7	good
	F3	0,36	0,30	16,7	0,29	0,25	4,0	0,36	0,32	11,1	0,38	0,32	15,8		
	F5	0,17	–	–	0,14	–	–	0,17	–	–	0,17	–	–		
2 C	F1	2,09	1,78	14,8	1,62	1,53	5,6	1,75	1,44	17,7	1,73	1,76	0	7	good
	F3	0,51	0,44	13,7	0,40	0,32	20,0	0,42	0,39	11,9	0,48	0,44	8,3		
	F5	0,27	–	–	0,21	–	–	0,22	–	–	0,25	–	–		
2 D	F1 F3 F5	–	–	–	–	–	–	–	–	–	–	–	–	–	stuck

Table 2 Continuation

Example	F	Colour density d: cyan a	cyan b	cyan d %	magenta a	magenta b	magenta d %	yellow a	yellow b	yellow d %	black a	black b	black d %	Gradation	"anti-" blocking characteristics
2 E	F1	2,05	1,80	12,2	1,65	1,56	5,5	1,73	1,44	16,8	1,75	1,76	0	7	good
	F3	0,50	0,44	12,0	0,40	0,30	10,0	0,41	0,38	7,3	0,50	0,48	4,0		
	F5	0,26	–	–	0,20	–	–	0,20	–	–	0,25	–	–		
2 F	F1	2,01	1,78	11,4	1,62	1,55	4,3	1,75	1,53	12,6	1,73	1,75	0	7	good
	F3	0,48	0,43	10,4	0,38	0,32	15,8	0,40	0,37	7,5	0,48	0,45	6,3		
	F5	0,25	–	–	0,21	–	–	0,20	–	–	0,24	–	–		
2 G	F1	1,84	1,56	15,2	1,52	1,44	5,3	1,35	1,22	9,6	1,75	1,75	0	7	good
	F3	0,44	0,40	9,1	0,42	0,39	7,1	0,42	0,36	14,3	0,48	0,45	6,3		
	F5	0,18	–	–	0,19	–	–	0,20	–	–	0,25	–	–		
Comparison (Hitachi)	F1	1,70	1,36	20,0	1,43	1,15	19,6	1,51	1,21	19,9	1,69	1,39	17,8	6	good
	F3	0,28	0,21	25,0	0,35	0,26	25,7	0,43	0,33	23,3	0,44	0,33	25,0		
	F5	0,09	–	–	0,03	–	–	0,09	–	–	0,08	–	–		

Table 3 Characteristics of the Printed Image Receiving Material Produced according to Example 3

Example	F	Colour density d: cyan a	cyan b	cyan d %	magenta a	magenta b	magenta d %	yellow a	yellow b	yellow d %	black a	black b	black d %	Gradation	"anti-" blocking characteristics
	F1	1,78	1,41	20,7	1,48	1,38	6,8	1,58	1,36	13,9	1,57	1,47	6,4		
3A	F3	0,30	0,26	13,3	0,28	0,25	10,7	0,30	0,25	16,7	0,35	0,28	20,0	7	good
	F5	0,11	-	-	0,12	-	-	0,10	-	-	0,13	-	-		
	F1	1,79	1,51	15,6	1,41	1,34	4,9	1,57	1,43	8,9	1,52	1,55	0,0		
3B	F3	0,39	0,29	25,0	0,32	0,26	18,8	0,35	0,31	11,4	0,39	0,35	10,0	7	good
	F5	0,17	-	-	0,14	-	-	0,17	-	-	0,17	-	-		
	F1	1,77	1,46	17,5	1,41	1,33	5,7	1,56	1,41	9,6	1,53	1,55	0,0		
3C	F3	0,39	0,25	35,9	0,33	0,26	21,2	0,36	0,32	11,1	0,40	0,35	12,5	7	good
	F5	0,18	-	-	0,15	-	-	0,18	-	-	0,20	-	-		
	F1	1,70	1,36	20,0	1,43	1,15	19,6	1,51	1,21	19,9	1,69	1,39	17,8		
Comparison	F3	0,28	0,21	25,0	0,35	0,26	25,7	0,43	0,33	23,3	0,44	0,33	25,0	6	good
(Hitachi)	F5	0,09	-	-	0,03	-	-	0,09	-	-	0,08	-	-		

Table 4 Characteristics of the Printed Image Receiving Material Produced according to Examples V1 - V3

Example	F	Colour density d												Gradation	"anti-" blocking characteristics
		cyan			magenta			yellow			black				
		a	b	d %	a	b	d %	a	b	d %	a	b	d %		
V1 A		-	-	-	-	-	-	-	-	-	-	-	-	-	stuck
V1 B		-	-	-	-	-	-	-	-	-	-	-	-	-	stuck
V2	F1	1,45	1,17	19,3	1,34	1,24	7,5	1,36	0,53	61,0	1,42	1,46	1,4	6	good
	F3	0,21	0,10	52,3	0,18	0,11	38,9	0,14	0,02	85,7	0,24	0,18	25,0		
	F5	0,10	-	-	0,10	-	-	0,05	-	-	0,10	-	-		
V3 Hitachi-receiving material	F1	1,70	1,36	20,0	1,43	1,15	19,6	1,51	1,21	19,9	1,69	1,39	17,8	6	good
	F3	0,28	0,21	25,0	0,35	0,26	25,7	0,43	0,33	23,3	0,44	0,33	25,0		
	F5	0,09	-	-	0,03	-	-	0,09	-	-	0,08	-	-		

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:-

1. An image-receiving material for dye diffusion thermal transfer comprising a resin-coated base paper and an image-receiving layer formed on the front side of said base paper and comprised of a dye-receiving resin, wherein the dye-receiving resin is a combination of at least one acrylate copolymer of monomers containing polar groups and an oxidized polyethylene.

2. An image-receiving material according to claim 1, wherein the polar groups contained in the acrylate copolymer are of the carboxyl-, metal-combined carboxyl groups, and/or nitrile groups.

3. An image-receiving material according to claim 2, wherein the metal-combined carboxyl groups are zinc-combined carboxyl groups.

4. An image-receiving material according to any one of claims 1 to 3, wherein the monomers which contain the polar groups and participate in the structure of the acrylate copolymers are acrylnitrile and/or methacrylic acid and the amount of these monomers contained in the copolymer is between 10 and 40 mol.-%.

5. An image-receiving material according to claim 4, wherein the content of monomers is between 25 and 35 mol.-%

6. An image-receiving material according to any one of claims 1 to 5, wherein the acrylate copolymer additionally contains styrene as a monomer and its content is between 0 and 40 mol.-%.

7. An image-receiving material according to claim 1, wherein the relationship of acrylate-copolymer to oxidized polyethylene is between 99 to 1 and 30 to 70.

8. An image-receiving material according to claim 7, wherein the relationship of acrylate-copolymer to oxidized polyethylene is between 70 to 30 and 40 to 60.

9. An image-receiving material according to any one of claims 1 to 8, wherein the receiving layer contains additional additives such as pigments, matting agents, wetting agents and other auxiliary agents.

10. An image-receiving material according to any one of claims 1 to 9, wherein the coating weight of the receiving layer is between 0,3 and 15 g/sq.m.

11. An image-receiving material according to claim 10, wherein the coating weight of the receiving layer is between 1 and 10 g/sq.m.

12. An image-receiving material according to any one of claims 1 to 11, wherein the resin coating of the base paper is a polyolefine coating with the coating weight of more than 5 g.sq.m.

13. A process for the manufacture of an image-receiving material for dye diffusion thermal transfer with a receiving layer containing dye-receiving resin applied to the front side of a resin-coated base paper, wherein the receiving layer comprising a combination of at least one acrylate copolymer containing polar groups and an oxidized polyethylene is applied as an aqueous composition onto the front side of the resin-coated base paper in a one-step process.

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