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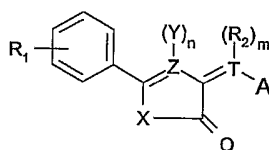
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(54) Title: NEW COMPOUNDS, A PROCESS FOR THEIR PREPARATION AND THEIR USE AS DYES AND PIGMENTS



(1)

when Z is =N- then n is 0, m is 0 or 1, when T is =N- then m is 0, A is a heterocyclic or linear or polycondensed aromatic group which is unsubstituted or substituted by hydroxy alkyl, halogen, nitro, cyano, amino, acylamino, carboxylic ester, sulfonic ester, carboxylic amide, sulfonic amide, alkoxy, aryloxy, alkylthio, arylthio or phenyl, or A together with T and R₂ can form an allycyclic or heterocyclic ring, their preparation and their use in the production of coloured plastics or polymeric colour particles.

New compounds, a process for their preparation and their use as dyes and pigments

The present invention relates to new compounds, to their preparation and to their use in the production of coloured plastics or polymeric colour particles.

The subject of the present invention are compounds of formula (1)



wherein

R_1 is hydrogen, hydroxy, halogen, nitro, cyano, amino, carboxy, carboxylic ester, sulfo, sulfonic ester, carboxylic amide, sulfonic amide, alkoxy, aryloxy, alkylthio or arylthio,

X is $-O-$, $-S-$, $-NH-$ or $-N(alkyl)-$,

Y is hydrogen or carboxylic ester,

Z is $=C-$ or $=N-$,

T is $=C-$ or $=N-$,

R_2 is hydrogen, alkyl or CN,

n is 0 or 1, when Z is $=N-$ then n is 0,

m is 0 or 1, when T is $=N-$ then m is 0,

A is a heterocyclic or linear or polycondensed aromatic group which is unsubstituted or substituted by hydroxy alkyl, halogen, nitro, cyano, amino, acylamino, carboxylic ester, sulfonic ester, carboxylic amide, sulfonic amide, alkoxy, aryloxy, alkylthio, arylthio or phenyl, or

A together with T and R_2 can form an allycyclic or heterocyclic ring.

According to the invention an alkyl is for example a straight-chain or branched C_{1-8} alkyl as methyl, ethyl, n-propyl, isopropyl, n-butyl, sec-butyl, isobutyl, tert-butyl, n-pentyl, 2-pentyl, 3-pentyl, 2,2-dimethylpropyl, hexyl, heptyl, 2,4,4-trimethylpentyl, 2-ethylhexyl or octyl, preference being given to a C_{1-4} alkyl.

According to the invention an alkylthio is for example methylthio, ethylthio, propylthio, butylthio, heptylthio or hexylthio.

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According to the invention an alkoxy is for example a straight-chain or branched C₁₋₈alkoxy, for example methoxy, ethoxy, n-propoxy, isopropoxy, n-butoxy, sec-butoxy, isobutoxy, tert-butoxy, n-pentyloxy, 2-pentyloxy, 3-pentyloxy, 2,2-dimethylpropoxy, n-hexyloxy, n-heptyloxy, n-octyloxy, 1,1,3,3-tetramethylbutoxy or 2-ethylhexyloxy.

According to the invention an aryloxy is to be understood as being for example a C₆₋₂₄aryloxy, preferably a C₆₋₁₂aryloxy radical, for example phenoxy or 4-methylphenoxy.

According to the invention an arylthio is for example phenylthio or naphthylthio.

A as aromatic group includes, for example, phenylene, naphthalene, acenaphthylene, anthracene, phenanthrene, naphthacene, chrysene, pyrene or perylene. W is preferably phenylene, naphthalene, anthracene, phenanthrene, perylene or pyrene, and most preferably phenylene or naphthalene.

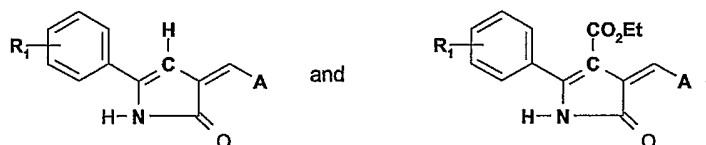
A as heterocyclic group is, for instance, pyridine, pyrazine, pyrimidine, pyridazine, indole, isoindole, quinoline, isoquinoline, carbazole, phenothiazine, benzimidazolone, benzothiazole, pyrrole, imidazole, pyrrolidine, piperidine, piperazine, morpholine or pyrazole.

Heterocyclic ring formed by A, T and R₂ can be for example pyridine, pyrazine, pyrimidine, pyridazine, indole, isoindole, quinoline, isoquinoline, carbazole, phenothiazine, benzimidazolone, benzothiazole, pyrrole, imidazole, pyrrolidine, piperidine, piperazine, morpholine or pyrazole.

According to the invention an ester is for example methyl-, ethyl-, propyl- or butylester.

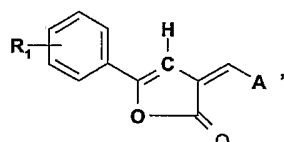
The preferred compounds of formula (1) are

a) phenyl-butyrolactams of formula

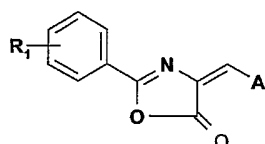


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b) phenyl-butyrolactones of formula

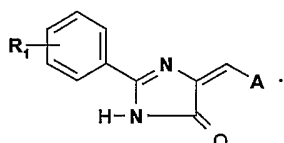


c) phenyl-oxazolones of formula



and

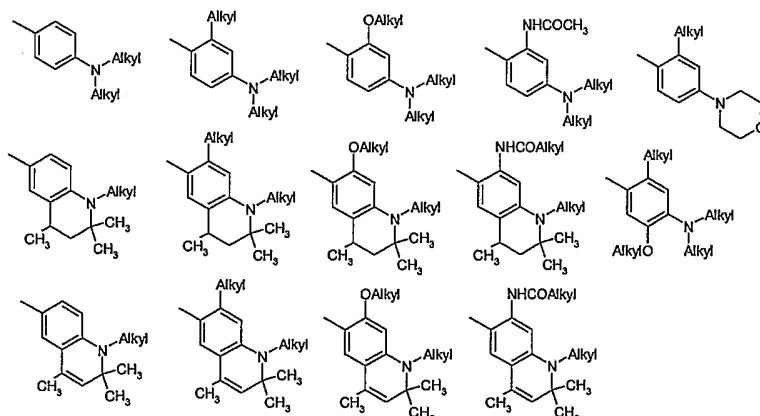
d) phenyl-imidazolones of formula



wherein R_1 and A have the meaning given under the formula (1) and Et is $-\text{CH}_2\text{CH}_3$.

A is preferred a group substituted by one or more electron donor groups as for example $-\text{OC}_1-\text{C}_{12}\text{alkyl}$, $-\text{NHC}_1-\text{C}_{12}\text{alkyl}$, $-\text{N}(\text{C}_1-\text{C}_{12}\text{alkyl})_2$ or $-\text{SC}_1-\text{C}_{12}\text{alkyl}$.

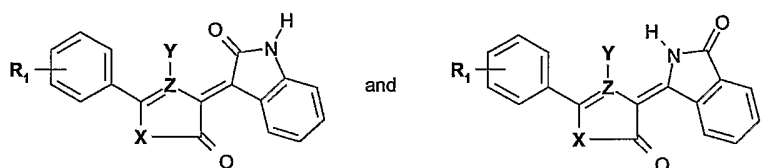
A is most preferred one of the following groups:



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In the above groups Alkyl means C₁-C₁₂alkyl, preferably C₁-C₄alkyl.

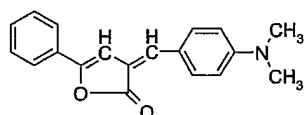
Preferred heterocycles which are formed with A, T and R₂ are:



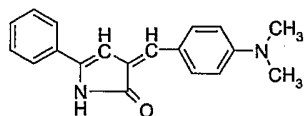
wherein R₁, X, Y and Z have the meaning given under the formula (1).

Further preferred are compounds of formula (1), wherein R₁ is hydrogen, R₂ is hydrogen, A is phenyl which is unsubstituted or substituted by nitro, methoxy, ethoxy, methylthio, chloro acetyloxy, -N(CH₃)₂ or -N(C₂H₅)₂, 1-naphthyl, 9-anthracenyl, pyrene, 2-ethylcarbonyl, isoindoline or isoindolinone, X is -O- or -NH-, Y is hydrogen or COOC₂H₅, T is =C-, Z is =C- or =N-, n is 0 or 1 and m is 1.

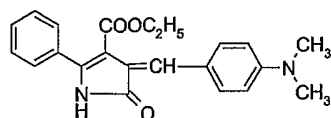
The most preferred compounds of formula (1) are the compounds of the formulae



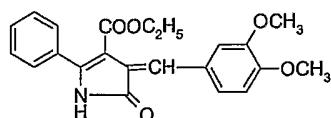
(100),



(101),



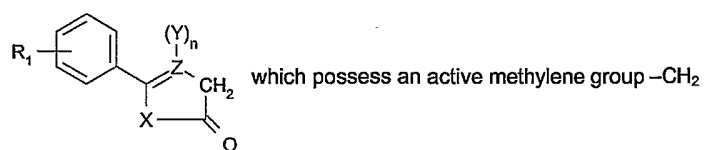
(102) and



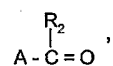
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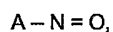
The compounds of formula (1) according to the invention are prepared, for example, by reacting a compound of formula



with a compound of formula



or with a compound of formula

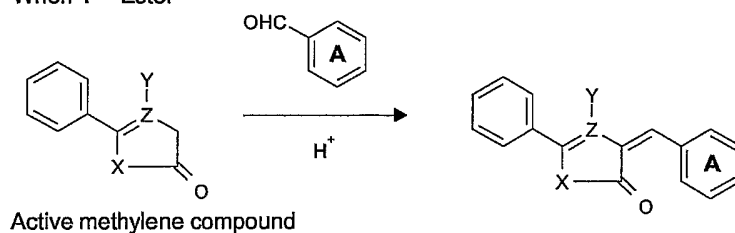


wherein R_1 , R_2 , A , X , Y , Z and n are as defined above for formula (1), at elevated temperature.

The general syntheses based on the condensation of an aldehyde, a ketone, a nitroso or imino-derivative with an active methylene compound are characterized by the following reactions schemas:

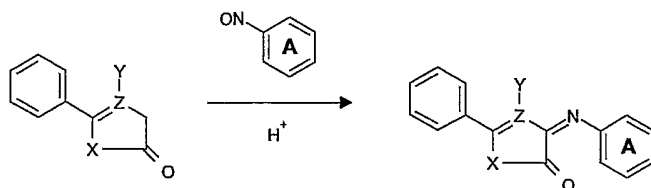
1) In case of an aldehyde derivative:

When $\text{Y} = \text{Ester}$

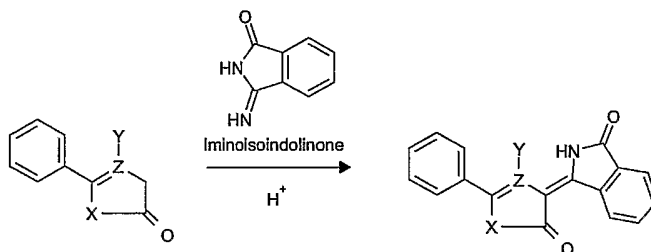


2) In case of a nitroso derivative:

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Active methylene compound

3) In case when A, T and R₂ form a heterocyclic ring:

Active methylene compound

Compounds of formula (1) can contain one or more water soluble groups as sulfonic, carboxylic or cationic groups.

Water soluble derivatives of compounds of formula (1) can be used as dyestuffs for textile application, coloration of cotton, wool, polyamide and polyacrylonitrile using all the well known dyeing processes.

Such dyes are useful for dyeing and printing manufactured natural polymer and especially synthetic hydrophobic fibre materials, especially textile materials. Textile materials composed of blend fabrics comprising such manufactured natural polymer or synthetic hydrophobic textile materials are likewise dyeable or printable with the dyes of the invention.

Useful manufactured natural polymer textile materials are especially cellulose acetate and cellulose triacetate.

Synthetic hydrophobic textile materials are especially linear aromatic polyesters, for example polyesters formed from a terephthalic acid and glycols, particularly ethylene glycol, or condensation products of terephthalic acid and 1,4-bis(hydroxymethyl)cyclohexane; polycarbo-

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nates, for example those formed from α,α -dimethyl-4,4-dihydroxydiphenylmethane and phosgene; or fibres based on polyvinyl chloride or polyamide.

The above dyes are applied to the textile materials according to known dyeing processes. For example, polyester fibres are exhaust dyed from an aqueous dispersion in the presence of customary anionic or nonionic dispersants with or without customary carriers at temperatures between 80 and 140°C. Cellulose acetate is preferably dyed at between about 65 to 85°C and cellulose triacetate at up to 115°C.

The above dyes are also useful for dyeing by the thermosol, exhaust and continuous processes and for printing processes. The exhaust process is preferred. The liquor ratio depends on the apparatus, the substrate and the make-up form. However, the liquor ratio can be chosen to be within a wide range, for example in the range from 4:1 to 100:1, but it preferably is between 6:1 to 25:1.

The textile material mentioned may be present in the various processing forms, for example as a fibre, yarn or web or as a woven or loop-formingly knitted fabric.

It is advantageous to convert the dyes into a dye preparation before use. For this, the dyes are ground so that their particle size is on average between 0.1 and 10 microns. The grinding may be effected in the presence of dispersants. For example, the dried dye is ground with a dispersant or kneaded in paste form with a dispersant and then dried under reduced pressure or by spray drying. The preparations thus obtained can be used to prepare print pastes and dyebaths by adding water.

Printing utilizes the customary thickeners, for example modified or nonmodified natural products, for example alginates, British gum, gum arabic, crystal gum, carob bean flour, tragacanth, carboxymethylcellulose, hydroxyethylcellulose, starch or synthetic products, for example polyacrylamides, polyacrylic acid or copolymers thereof or polyvinyl alcohols.

The above dyes confer on the materials mentioned, especially on polyester material, level shades having very good service fastnesses, such as in particular good light fastness, especially a very good hot light fastness, fastness to dry heat setting and pleating, chlorine fastness and wet fastness such as fastness to water, perspiration and washing; the dyeings are further characterized by good rub fastness and heat stability.

Water insoluble derivatives of compounds of formula (1) can be used as disperse dyes for coloration of PET by exhaustion or pigments for mass coloration of plastics or can be used for inks and paints. These products can also be used for coloration of wood and metals and they are also suitable as functional dyes for special applications such as optical information storage, or display devices or printed circuit boards.

The present invention relates also to a process for the production of coloured plastics or polymeric colour particles, which comprises mixing together a high molecular weight organic material and a tinctorially effective amount of at least one compound of formula (1).

The present invention further relates to the use of the compounds of formula (1) individually as colourants, especially for colouring or pigmenting organic or inorganic, high-molecular-weight or low-molecular-weight material, especially high-molecular-weight organic material. It is also possible, however, for the compositions according to the invention comprising compounds of formula (1) to be used in the form of mixtures, solid solutions or mixed crystals. Compounds of formula (1) can also be combined with colourants of another chemical class, for example with dyes or pigments, for example those selected from the group of the diketo-pyrrolopyrroles, quinacridones, perylenes, dioxazines, anthraquinones, indanthrones, flavanthrones, indigos, thioindigos, quinophthalones, isoindolinones, isoindolines, phthalocyanines, metal complexes, azo pigments and azo dyes.

The high-molecular-weight material may be organic or inorganic and may be synthetic and/or natural material. The high-molecular-weight organic material usually has an average molecular weight of 10^5 - 10^7 g/mol. It may be, for example, a natural resin or a drying oil, rubber or casein or a modified natural material, such as chlorinated rubber, oil-modified alkyd resins, viscose, or a cellulose ether or ester, such as ethylcellulose, cellulose acetate, propionate or butyrate, cellulose acetobutyrate or nitrocellulose, but is especially a completely synthetic organic polymer (duroplasts and thermoplasts) as may be obtained by polymerisation, for example by polycondensation or polyaddition. The class of polymers includes, for example, polyolefins, such as polyethylene, polypropylene, polyisobutylene, and substituted polyolefins, such as polymerisates of monomers such as vinyl chloride, vinyl acetate, styrene, acrylonitrile, acrylates, methacrylates, fluoropolymers, such as polyfluoroethylene, polytrifluoro-chloroethylene or tetrafluoroethylene/hexafluoropropylene mixed polymerisate, and copolymerisates of the mentioned monomers, especially ABS (acrylonitrile/butadiene/styrene) or EVA (ethylene/vinyl acetate). From the group of polyaddition and polycondensation resins it is possible to use, for example, condensation products of formaldehyde with phenols, the so-

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called phenoplasts, and condensation products of formaldehyde and urea or thiourea, also melamine, the so-called aminoplasts, and the polyesters used as surface coating resins, either saturated, such as alkyd resins, or unsaturated, such as maleic resins, and also linear polyesters, polyamides, polyurethanes, polycarbonates, polyphenylene oxides or silicones, and silicone resins. The mentioned high-molecular-weight compounds may be present individually or in mixtures in the form of kneadable compounds, melts or in the form of spinning solutions. They may also be present in the form of their monomers or in the polymerised state in dissolved form as film-formers or binders for paints or printing inks, such as, for example, boiled linseed oil, nitrocellulose, alkyd resins, melamine resins and urea-formaldehyde resins or acrylic resins.

Low-molecular-weight materials are, for example, mineral oils, waxes or lubricating greases.

The present invention further relates, therefore, to the use of the compounds of formula (1) for the production of inks, for printing inks in printing processes, for flexographic printing, screen printing, the printing of packaging, security colour printing, intaglio printing or offset printing, for preliminary printing stages and for textile printing, for office and home use or for graphics, such as, for example, for paper goods, for ball-point pens, felt-tip pens, fibre-tip pens, paperboard, wood, (wood) stains, metal, stamp pads or inks for impact printing processes (with impact printing ink ribbons), for the production of colourants, for paints, for use in industry or advertising, for textile decoration and industrial labelling, for roll coating or powder coating compositions or for automobile paints, for high-solids (low-solvent), water-containing or metallic paints or for pigmented formulations for aqueous paints, for mineral oils, lubricating greases or waxes, for the production of coloured plastics for coatings, fibres, plates or moulded substrates, for the production of non-impact printing material for digital printing, for the thermal wax-transfer printing process, the ink-jet printing process or for the thermal transfer printing process, and also for the production of colour filters, especially for visible light in the range of from 400 to 700 nm, for liquid crystal displays (LCDs) or charge-coupled devices (CCDs) or for the production of cosmetics or for the production of polymeric colour particles, toners, dry copy toners, liquid copy toners or electrophotographic toners.

The present invention further relates to inks comprising high-molecular-weight organic material and a colour-producing amount of the compound of formula (1).

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For example, the inks can be produced by mixing the compounds according to the invention with polymeric dispersants.

The mixing of the compounds according to the invention with the polymeric dispersant is preferably carried out by generally known mixing methods, such as stirring or mixing, and the use of an intensive mixer, such as an Ultraturax, is especially to be recommended.

When mixing the compounds according to the invention with polymeric dispersants, a water-dilutable organic solvent is advantageously used.

The weight ratio of the compounds according to the invention to ink is advantageously selected to be in the range of from 0.0001 to 75 % by weight, preferably from 0.001 to 50 % by weight, based on the total weight of the ink.

The present invention therefore relates also to a process for the production of inks which comprises mixing high-molecular-weight organic material with a colour-producing amount of the compound of formula (1).

The present invention further relates to colourants comprising high-molecular-weight organic material and a compound according to the invention of formula (1) in a colour-producing amount.

The present invention relates, in addition, to a process for the preparation of colourants which comprises mixing a high-molecular-weight organic material and a colour-producing amount of the compound according to the invention of formula (1).

The present invention further relates to coloured or pigmented plastics or polymeric coloured particles comprising high-molecular-weight organic material and compound of formula (1) in a colour-producing amount.

The present invention relates, in addition, to a process for the preparation of coloured or pigmented plastics or polymeric coloured particles which comprises mixing together a high-molecular-weight organic material and a colour-producing amount of the compound of formula (1).

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The colouring of high-molecular-weight organic substances with the colourants of formula (1) is carried out, for example, by mixing such a colourant, optionally in the form of a master batch, into those substrates using roll mills or mixing or grinding apparatus, whereby the colourant is dissolved or finely distributed in the high-molecular-weight material. The high-molecular-weight organic material with the admixed colourant is then processed according to procedures known *per se*, such as, for example, calendering, compression moulding, extrusion moulding, coating, spinning, casting or injection-moulding, whereby the coloured material acquires its final form. Admixing of the colourant can also be carried out immediately prior to the actual processing step, for example by continuously metering a powdered colourant according to the invention and a granulated high-molecular-weight organic material, and optionally also additional ingredients, such as additives, directly into the inlet zone of an extruder simultaneously, where mixing takes place just before the processing operation. In general, however, prior mixing of the colourant into the high-molecular-weight organic material is preferred, since more uniform results can be obtained.

It is often desirable for the purpose of producing non-rigid mouldings or reducing the brittleness thereof to incorporate so-called plasticisers into the high-molecular-weight compounds before shaping. There may be used as plasticisers, for example, esters of phosphoric acid, phthalic acid or sebacic acid. In the process according to the invention, the plasticisers can be incorporated into the polymers before or after the incorporation of the colourant. In order to obtain different colour shades it is also possible to add to the high-molecular-weight organic substances, in addition to the compounds of formula (1), any desired amounts of constituents such as white, coloured or black pigments.

For the colouring of paints and printing inks, the high-molecular-weight organic materials and the compounds of formula (1) optionally together with additional ingredients, such as fillers, dyes, pigments, siccatives or plasticisers, are finely dispersed or dissolved in a common organic solvent or solvent mixture. That procedure may comprise dispersing or dissolving each individual component on its own or dispersing or dissolving several components together and only then combining all the components. Processing is carried out in accordance with customary methods, for example by spraying, film-spreading or one of the many printing methods, whereupon the paint or printing ink is advantageously cured thermally or by irradiation, optionally after previous drying.

When the high-molecular-weight material to be coloured is a paint, it may be a conventional paint or a special paint, for example an automobile finish, preferably a metal-effect finish con-

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taining, for example, metal or mica particles.

Preference is given to the colouring of thermoplastics, especially also in the form of fibres, and printing inks. Preferred high-molecular-weight organic materials that can be coloured according to the invention are, very generally, polymers having a dielectric constant ≥ 2.5 , especially polyesters, polycarbonate (PC), polystyrene (PS), polymethylmethacrylate (PMMA), polyamide, polyethylene, polypropylene, styrene/acrylonitrile (SAN) or acrylonitrile/butadiene/styrene (ABS). More especially preferred are polyesters, polycarbonate, polystyrene and PMMA. Most especially preferred are polyesters, polycarbonate and PMMA, especially aromatic polyesters that can be obtained by polycondensation of terephthalic acid, such as, for example, polyethylene terephthalate (PET) or polybutylene terephthalate.

They can be used in the form of their monomers or copolymers or in the polymerised state in dissolved form as film formers or binders for paints that can be used for the decoration of metal or for decorative colour finishes, and for printing inks used, for example, in the ink-jet printing process, or also for wood stains.

Special preference is also given to the colouring of mineral oils, lubricating greases and waxes with the compounds according to the invention.

The present invention also relates to mineral oils, lubricating greases and waxes comprising high-molecular-weight organic material and a compound of formula (1), in a colour-producing amount.

The present invention also relates to a process for the preparation of mineral oils, lubricating greases and waxes, which comprises mixing high-molecular-weight organic material with a colour-producing amount of the compound of formula (1).

The present invention also relates to non-impact printing material comprising high-molecular-weight organic material and a compound of formula (1), in a colour-producing amount.

The present invention relates, in addition, to a process for the preparation of non-impact printing material, which comprises mixing together a high-molecular-weight organic material and a colour-producing amount of the compound of formula (1).

The present invention further relates to a process for the production of colour filters comprising a transparent substrate and a red, a blue and a green coating applied thereto in any de-

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sired sequence, which comprises using for the production of the red, blue and green coatings a correspondingly coloured compound of formula (1).

The different-coloured coatings are preferably arranged in such a pattern that they do not overlap over at least 5 % of their respective surface area and, most preferably, do not overlap at all.

The colour filters can be coated, for example, using inks, especially printing inks, comprising the compounds according to the invention, or, for example, by mixing a compound according to the invention with a chemically, thermally or photolytically structurable high-molecular-weight material (resist). The further production can be carried out, for example, analogously to EP-A-654 711, by application to a substrate, such as an LCD, subsequent photo-structuring and developing.

The invention further includes a transparent substrate coated with a red, a blue and a green coating each of a correspondingly coloured compound of formula (1), comprising pigmented high-molecular-weight organic material.

The sequence in which coating is carried out is not important as a rule. The different-coloured coatings are preferably arranged in such a pattern that they do not overlap over at least 5% of their respective surface area and, most preferably, do not overlap at all.

The present invention also includes colour filters comprising a transparent substrate and, applied thereto, a red, a blue and a green coating, each obtainable from a correspondingly coloured compound of formula (1).

The present invention also includes the use of compounds of formula (1) for optical information storage applications (ois).

The present invention relates, in addition, to toners comprising high-molecular-weight organic material and a compound of formula (1), in a colour-producing amount.

The present invention also relates to a process for the production of toners, which comprises mixing together a high-molecular-weight organic material and a colour-producing amount of

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the compound of formula (1).

The present invention also relates to inks or colourants for paints, printing inks, mineral oils, lubricating greases or waxes, or coloured or pigmented plastics, non-impact printing material, colour filters, cosmetics or toners comprising high-molecular-weight organic material and a compound of formula (1), in a colour-producing amount.

In a special embodiment of the process according to the invention, toners, paints, inks or coloured plastics are produced by processing master batches of toners, paints, inks or coloured plastics in roll mills or mixing or grinding apparatus.

A colour-producing amount of the compound of formula (1) means in the present invention normally from 0.0001 to 99.99 % by weight, preferably from 0.001 to 50 % by weight and especially from 0.01 to 50 % by weight, based on the total weight of the material coloured or pigmented therewith.

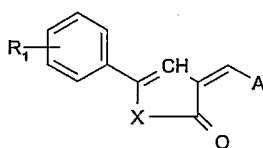
The coloured/pigmented high-molecular-weight materials obtained, such as, for example, plastics, fibres, paints and prints, are distinguished by very high colour intensity, high saturation, good fastness to overspraying, good migration-stability, good fastness to heat, light and weathering and by a high gloss and good IR reflectance behaviour.

In order to improve the light fastness properties, UV absorbers are advantageously mixed into the plastics or polymeric particles to be coloured with the compound of formula (1) according to the invention. The amount of UV absorber can vary within a wide range; advantageously there is used from 0.01 to 1.0 % by weight, especially from 0.05 to 0.6 % by weight, more especially from 0.1 to 0.4 % by weight, of a UV absorber, based on the weight of the plastics or polymeric particles.

The following Examples serve to illustrate the invention. Unless otherwise indicated, the parts are parts by weight and the percentages are percentages by weight. The temperatures are given in degrees Celsius. The relationship between parts by weight and parts by volume is the same as that between grams and cubic centimetres.

Example 1: (General method for condensation)

A stirred solution of 3-benzoyl propanoic acid (see C.F.H.Allen, Org Synth., Coll. Vol. II, 81) (10 g, 0.056 mol), 3-nitro benzaldehyde (10 g, 0.066 mol) and sodium acetate (4.8 g, 0.058 mol) in acetic anhydride (70 ml) is heated under nitrogen atmosphere at 90-95° C for 2 hours. The reaction mixture is cooled to 20° C, filtered and washed with methanol and water. The yield of the yellow product after drying is (12.5 g, 76%). m. p. 216-18°C; UV_(ethanol): $\lambda_{\max}$ 387 nm, ϵ 10179; ¹H NMR (CDCl₃): δ 6.95(1H, s), 7.52(4H, m), 7.65-8.00(4H, m), 8.30(1H, d), 8.55(1H, s). The compounds given in the Table 1 are synthesized using above procedure. In the following Tables Et means ethyl, Me means methyl, Ph means Phenyl and Ac means acetyl.

Table 1

Example	R ₁	X	A	Shade (PET)	$\lambda_{\max}$ (nm)
1	H	O	3-NO ₂ -Ph	Yellow	387
2	H	O	4-OMe-Ph	Yellow	401
3	H	O	4-SMe-Ph	Yellow	412
4	H	O	4-N,N-DiMe-Ph	Green Yellow	460
5	H	O	4-Cl-Ph	Yellow	392
6	H	O	Ph	Yellow	387
7	H	O	Naphthyl	Yellow	401

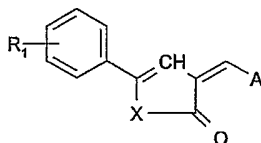
Example 8: (General Procedure for conversion of lactone to lactum)

A suspended solution of 5-phenyl-3-(3-nitro) benzylidene-lactone from Example 1 (8.0 g, 0.027 mol) in absolute alcohol (60 ml) and ammonia solution (ca. 30%, d 0.89)(5 ml) is heated to 70-80° C for 15 minutes. Remaining ammonia solution (5 ml) and potassium carbonate (2.5 g) is added to it. The reaction is continued for 2 hours at 75-80° C maintaining the ammonia level in the solution by adding ammonia solution (ca. 10 ml), followed by the addition of potassium carbonate (ca. 2.5 g). During this time, the solution becomes clear orange and within next 15 minutes, yellow precipitate is formed. After 2 hours, the solution is cooled

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to 20° C and the product is filtered and washed with water. The yield of the yellow product is 3.4 g (42%). m. p. >300; UV(ethanol): $\lambda_{\max}$ 429nm, ϵ 13062; ^1H NMR (CDCl_3): δ 6.55(1H, s), 7.42(1H, s), 7.50(3H, m), 7.62(3H, m), 7.95(2H, d), 8.22(1H, d), 8.55(1H, s). The compounds given in the Table 2 are obtained following a similar procedure.

Table 2

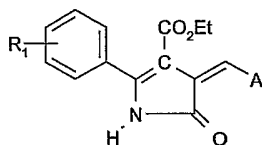


Example	R ₁	X	A	Shade (PET)	$\lambda_{\max}$ (nm)
8	H	NH	3-NO ₂ -Ph	Yellow	429
9	H	NH	4-OMe-Ph	Yellow	422
10	H	NH	4-N,N-DiMe-Ph	Orange	462
11	H	NH	4-Cl-Ph	Yellow	424
12	H	NH	Ph	Gold Yellow	416
13	H	NH	Naphthyl	Yellow	424

Example 14: (General Procedure for 4-Carboethoxy Lactum)

A stirred solution of 3-nitro benzaldehyde (3.5 g, 0.023 mol) and sodium acetate (2.5 g, 0.030 mol) in acetic anhydride (15 ml) is heated under nitrogen atmosphere to 90° C. To this 5-phenyl-4-carboethoxy-pyrroline-2-one (5 g, 0.021 mol) is added in small portions in about 10 minutes. The solution is stirred at 120-30° C for 35 minutes, cooled to 25° C and filtered. The precipitate is washed with acetic acid (15 ml) and water (200 ml). The yield of the bright yellow product is 5.7 g (73%). m. p. 199-200°C; UV(ethanol): $\lambda_{\max}$ 408nm, ϵ 10465; ^1H NMR (CDCl_3): δ 1.10(3H, t), 4.20(2H, q), 7.30-7.70 (7H, m), 8.20(3H, m), 8.35(1H, d), 9.00(1H, s). The compounds given in the Table 3 are obtained following a similar procedure.

Table 3



Example	R ₁	A	Shade (PET)	λ_{max} (nm)
14	H	3-NO ₂ -Ph	Yellow	408
15	H	4-OAc-Ph	Yellow	406
16	H	2-OAc-Ph	Yellow	402
17	H	4-OMe-Ph	Yellow	413
18	H	4-SMe-Ph	Yellow	421
19	H	4-OEt-Ph	Yellow	414
20	H	3,4-DiOMe-Ph	Yellow	416
21	H	4-N,N-DiMe-Ph	Orange	460
22	H	2-OAc-4-N,N-Et	Orange	466
23	H	1-Naphthyl	Yellow	411
24	H	9-Anthracenyl	Green Yellow	436
25	H	Pyrene	Orange	446
26	H	2-Ethylcarbazol	Yellow	438
27	H	4-Cl-Ph	Yellow	406
28	H	Ph	Yellow	401

Example 29:

1,3-Di-iminoisoindoline (3.5 g, 0.023 mol) is suspended in absolute alcohol (100 ml) and glacial acetic acid (1.5 ml). The solution is heated to 75° C under nitrogen atmosphere for 10 minutes. 5-Phenyl-4-carboethoxypyrrole-2-one (5g , 0.021 mol) is added in portions in 5 minutes. The solution is stirred at 75-80° C for 30 minutes. The precipitate is filtered hot and washed with alcohol. The yield of dull green product is 5.8 g (74%). m. p. above 300°C; UV_(ethanol): λ_{max} 459nm, ϵ 16769; ¹H NMR (DMSO): δ 1.20(3H, t), 4.30(2H, q), 7.30-7.70(7H, m), 7.90(1H, d), 8.20-8.60(2H, d), 9.30(1H, d), 10.50(1H, s). Similar procedure is followed to prepare Example 30 (see Table 4) but using triethylamine instead of glacial acetic acid to neutralize 3-iminoisoindol-1-one.hydrochloride.

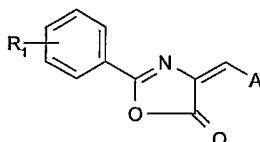
Table 4

Example	R ₁	A	Shade (PET)	$\lambda_{\max}$ (nm)
29	H	Isoindoline	Orange	459
30	H	Isoindolinone	Yellow	447

Example 31: (General Procedure for Oxazolone)

A stirred solution of hippuric acid (30 g, 0.16 mol), 1-naphthalene (23.5 g, 0.16 mol) and sodium acetate (14 g, 0.17 mol) in acetic anhydride (250 ml) is heated under nitrogen atmosphere at 90-95° C for 2 hours. The solution is cooled to 20° C, the curde product is filtered, and washed with acetic acid and water. Yield 24.5 g (49%) m. p. 164-66°C; UV_(ethanol): $\lambda_{\max}$ 399nm, ϵ 24117; ¹H NMR (CDCl₃): δ 7.55(3H, m), 7.65(3H, m), 7.87(1H, m), 7.95(1H, m), 8.15(1H, s), 8.20(2H, d), 8.30(1H, d), 9.00(1H, d). The compounds in the Table 5 are obtained following a similar procedure.

Table 5



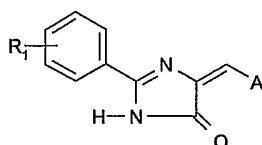
Example	R ₁	A	Shade (PET)	$\lambda_{\max}$ (nm)
31	H	1-Naphthyl	Yellow	399
32	H	4-N,N-DiMePh	Red	468
33	H	3-NO ₂ -Ph	Yellow	359
34	H	4-Cl-Ph	Yellow	364
35	H	9-Anthracenyl	Yellow	444
36	H	Pyrene	Orange	469

Example 37: (General Procedure for imidazolone)

A stirred suspended solution of Oxazolone from Example 32 (7.0 g, 0.020 mol) in glacial acetic acid (150 ml) is heated to 110° C and a slow stream of ammonia gas is passed through the solution for 4 hours at this temperature. The solution is cooled to 50° C and diluted with glacial acetic acid (100 ml). The crude product is filtered, washed with acetic acid (20 ml) and water (200 ml). Yield 3.4 g (48%). m. p. 297-297°C; UV_(ethanol): $\lambda_{\max}$ 440nm, ϵ 9811; ¹H NMR

(CDCl₃): δ 7.40-7.60(7H, m), 7.80-8.20(7H, m), 8.70(1H, s), 12.20(1H, s). The compounds given in the Table 6 are obtained following a similar procedure.

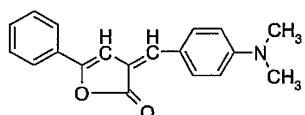
Table 6



Example	R ₁	A	Shade (PET)	λ_{max} (nm)
37	H	9-Anthracenyl	Orange	440
38	H	4-Cl-Ph	Yellow	386
39	H	1-Naphthyl	Yellow	411
40	H	Pyrene	Coffee	473

Dyeing Example 1:

1200.00 g of polyester granules (PET Arnite D04-300, DSM) are pre-dried for 4 hours at 130°C and then mixed homogeneously with 2.4 g of the compound of formula



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in a "roller rack" mixing apparatus for 15 minutes at 60 revolutions per minute.

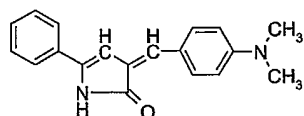
The homogeneous mixture is extruded in an extruder (twin screw 25 mm from Collin, D-85560 Ebersberg) with 6 heating zones at a maximum temperature of 275°C, cooled with water, granulated in a granulator (Turb Etuve TE 25 from MAPAG AG, CH-3001 Bern) and then dried for 4 hours at 130°C.

The resulting greenish yellow-coloured polyester granules have good allround fastness properties, especially good light fastness and high-temperature light fastness properties.

Dyeing Example 2:

1200.00 g of polyamide-6 granules (Ultramid B3K, BASF) are pre-dried for 4 hours at 75°C and then mixed homogeneously with 2.7 g of the compound of formula

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(101)

in a "roller rack" mixing apparatus for 15 minutes at 60 revolutions per minute.

The homogeneous mixture is extruded in an extruder (twin screw 25 mm from Collin, D-85560 Ebersberg) with 6 heating zones at a maximum temperature of 220°C, cooled with water, granulated in a granulator (Turb Etuve TE 25 from MAPAG AG, CH-3001 Bern) and then dried for 4 hours at 75°C.

The resulting orange-coloured polyamide granules have good allround fastness properties, especially good light fastness and high-temperature light fastness properties.

Claims

1. Compounds of formula (1)



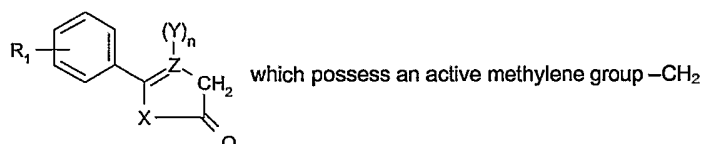
wherein

R_1 is hydrogen, hydroxy, halogen, nitro, cyano, amino, carboxy, carboxylic ester, sulfo, sulfonic ester, carboxylic amide, sulfonic amide, alkoxy, aryloxy, alkylthio or arylthio, X is $-O-$, $-S-$, $-NH-$ or $-N(alkyl)-$, Y is hydrogen or carboxylic ester, Z is $=C-$ or $=N-$, T is $=C-$ or $=N-$, R_2 is hydrogen, alkyl or CN, n is 0 or 1, when Z is $=N-$ then n is 0, m is 0 or 1, when T is $=N-$ then m is 0, A is a heterocyclic or linear or polycondensed aromatic group which is unsubstituted or substituted by hydroxy alkyl, halogen, nitro, cyano, amino, acylamino, carboxylic ester, sulfonic ester, carboxylic amide, sulfonic amide, alkoxy, aryloxy, alkylthio, arylthio or phenyl, or A together with T and R_2 can form an allycyclic or heterocyclic ring.

2. Compounds according to claim 1, wherein A is a heterocyclic or linear or polycondensed aromatic group substituted by one or more groups $-OC_1-C_{12}alkyl$, $-NHC_1-C_{12}alkyl$, $-N(C_1-C_{12}alkyl)_2$ or $-SC_1-C_{12}alkyl$.

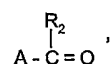
3. Compounds according to claim 1, wherein R_1 is hydrogen, R_2 is hydrogen, A is phenyl which is unsubstituted or substituted by nitro, methoxy, ethoxy, methylthio, chloro acetyloxy, $-N(CH_3)_2$ or $-N(C_2H_5)_2$, 1-naphthyl, 9-anthracenyl, pyrene, 2-ethylcarbonyl, isoindoline or isoindolinone, X is $-O-$ or $-NH-$, Y is hydrogen or $COOC_2H_5$, T is $=C-$, Z is $=C-$ or $=N-$, n is 0 or 1 and m is 1.

4. Process for the preparation of compounds of formula (1) according to claim 1, which process comprises reacting a compound of formula

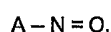


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with a compound of formula



or with a compound of formula



wherein R_1 , R_2 , A , X , Y , Z and n are as defined for formula (1) in claim 1, at elevated temperature.

5) Process for the preparation of coloured or pigmented plastics or polymeric coloured particles, which process comprises mixing together a high-molecular-weight organic material and a colour-producing amount of at least one compound of formula (1) according to claim 1.

6) Coloured or pigmented plastics or polymeric coloured particles comprising at least one compound of formula (1) according to claim 1.

7) Use of the compounds of formula (1) according to claim 1 for the production of inks, for printing inks in printing processes, for flexographic printing, screen printing, the printing of packaging, security colour printing, intaglio printing or offset printing, for preliminary printing stages and for textile printing, for office and home use or for graphics, such as, for example, for paper goods, for ball-point pens, felt-tip pens, fibre-tip pens, paperboard, wood, (wood) stains, metal, stamp pads or inks for impact printing processes (with impact printing ink ribbons), for the production of colourants, for paints, for use in industry or advertising, for textile decoration and industrial labelling, for roll coating or powder coating compositions or for automobile paints, for high-solids (low-solvent), water-containing or metallic paints or for pigmented formulations for aqueous paints, for mineral oils, lubricating greases or waxes, for the production of coloured plastics for coatings, fibres, plates or moulded substrates, for the production of non-impact printing material for digital printing, for the thermal wax-transfer printing process, the ink-jet printing process or for the thermal transfer printing process, and also for the production of colour filters, especially for visible light in the range of from 400 to 700 nm, for liquid crystal displays (LCDs) or charge-coupled devices (CCDs) or for the production of cosmetics or for the production of polymeric colour particles, toners, dry copy toners, liquid copy toners or electrophotographic toners, or for optical information storage applications (ois).