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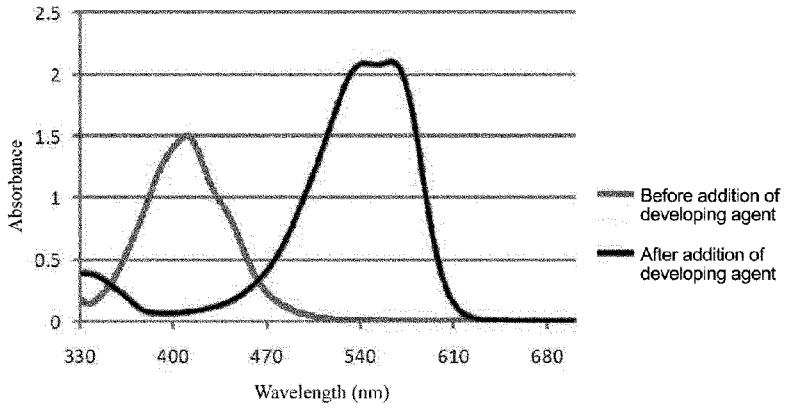
(54) **Title**: NON-AQUEOUS FLUID MARKER CONTAINING *N*-BENZYL-*N*-ETHYLANILINE AND METHOD FOR PREPARING THE SAME

[Fig. 1]



(57) **Abstract**: The present invention provides an azo dye which can be used as a marker for a non-aqueous fluid product containing *N*-benzyl-*N*-ethylaniline, and a method for preparing the same. The marker containing *N*-benzyl-*N*-ethylaniline according to the present invention exhibits high solubility in non-aqueous fluid products, has excellent storage stability, can be detected even in a small amount, and can exhibit a distinct color by reaction with an acid developer in the non-aqueous fluid product. Therefore, when the marker is added to a non-aqueous fluid product, the product can be easily distinguished from other companies' products containing no marker, which makes it possible to easily control the quality of the non-aqueous fluid product in the distribution process.

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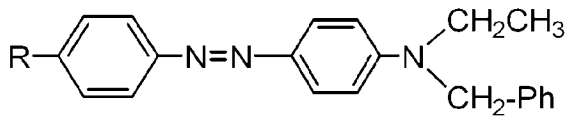
Description

Title of Invention: NON-AQUEOUS FLUID MARKER CONTAINING *N*-BENZYL-*N*-ETHYLANILINE AND METHOD FOR PREPARING THE SAME

Technical Field

[0001] The present invention relates to an azo dye represented by the following Formula 1, which can be used as a marker for a non-aqueous fluid product, and a method for preparing the same.

[0002] **Formula 1**

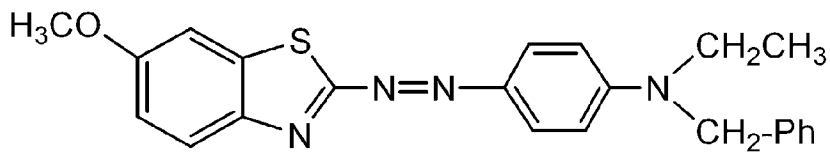
[0003] 

Background Art

[0004] A dye containing an azo group (-N=N-) as a chromophore is called an azo dye, and the azo dyes account for 50 % or more of the currently used dyes. Recently, extensive research aimed at developing a novel azo dye suitable for a variety of applications by adding functionality to the dyes has continued to progress.

[0005] According to a recent report (M. R. Yazdanbakhsh. et. al., J. Mol. Liquid. 151: 107-112 (2010)), a heteroazo disperse dye using *N*-benzyl-*N*-ethylaniline as a coupler exhibits solvatochromism. For example, a maximum absorption wavelength (λ_{max}) of a compound represented by the following Formula 2 was 503 nm in a chloroform, but it shifted to a longer wavelength of 614 nm in a glacial acetic acid by 111 nm.

[0006] **Formula 2**

[0007] 

[0008] At present, the methods for identifying the non-aqueous fluid products are as follows ; a method for exhibiting a color by adding a marker to a non-aqueous fluid product and then adding an acid or a base as a developing agent thereto, a quantitative measurement using a dye absorbing in a near infrared region, etc., are used. However, these methods have some problems in that the color may not be detected as the effect of the marker is lost over time, the developing agent used to develop a detectable color of markers may be difficult to handle as it is easily decomposed, and the good quantitative measurement may not perform as the components of the marker are slowly decomposed in water or organic solvent.

[0009] U.S. Patent No. 6,514,917, U.S. Patent No. 5,737,871, U.S. Patent No. 5,490,872, WO 1995-017483, and WO 1999-067346 disclose that a variety of azo dyes, which are easily handled and have high storage stability, can be used as markers to label petroleum products by reaction with an organic or inorganic acid. In particular, US Patent No. 6,514,917 discloses a method for quantifying a marker using an organic phosphoric or sulfonic acid as a developing agent miscible with petroleum products without extraction to an aqueous phase from the petroleum products.

[0010] The materials to be useful as markers for non-aqueous fluid products must have high solubility in a target product, have excellent storage stability, be detected even in a small amount, be easily detected, and be used in various non-aqueous fluid products. However, it is very difficult to use the existing azo dyes as a marker in terms of solubility and coloration in some non-aqueous fluid products such as petroleum products, solvents, inks, lubricants, etc.

[0011]

Summary of Invention

Technical Problem

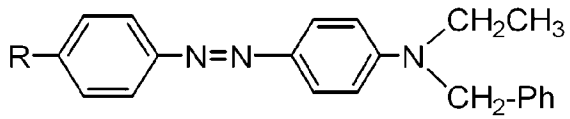
[0012] As a result of continuous studies to develop a compound which can exhibit a distinct color in a non-aqueous fluid product and facilitate quantitative measurement without extraction to an aqueous phase, the present inventors completed this invention by synthesizing several types of yellow azo disperse dyes containing *N*-benzyl-*N*-ethylaniline and these dyes could be effectively used as markers for non-aqueous fluid products.

[0013] Accordingly, an object of the present invention is to provide a novel yellow azo disperse dye for identifying a non-aqueous fluid product, and a method for preparing the same.

Solution to Problem

[0014] In one aspect, the present invention provides a non-aqueous fluid marker containing *N*-benzyl-*N*-ethylaniline represented by the following Formula 1:

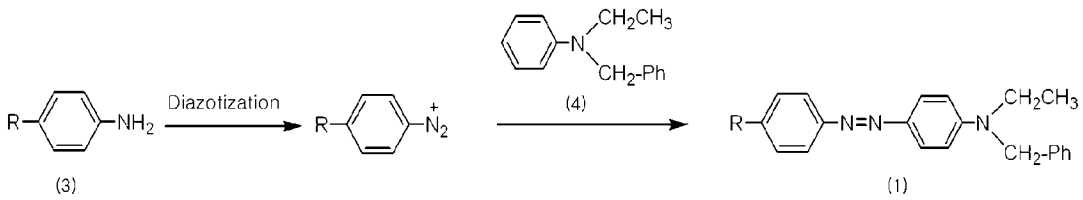
[0015] **Formula 1**

[0016] 

[0017] wherein R is a C_1 to C_{12} alkyl group, a C_1 to C_{12} alkoxy group, a C_1 to C_{12} hydroxyalkyl group, a C_1 to C_{12} hydroxyalkoxyalkyl group, a C_1 to C_{12} alkylphenyl group, a C_1 to C_{12} alkoxyphenyl group, a nitro group, a cyano group or a halogen atom.

[0018] In another aspect, the present invention provides a method for preparing a non-aqueous fluid marker, comprising: diazotizing an aniline derivative represented by the following Formula 3; and reacting *N*-benzyl-*N*-ethylaniline represented by the

following Formula 4 with the diazotized aniline derivative:

[0019] 

[0020] wherein R is a C_1 to C_{12} alkyl group, a C_1 to C_{12} alkoxy group, a C_1 to C_{12} hydroxyalkyl group, a C_1 to C_{12} hydroxyalkoxyalkyl group, a C_1 to C_{12} alkylphenyl group, a C_1 to C_{12} alkoxyphenyl group, a nitro group, a cyano group or a halogen atom.

[0021]

Advantageous Effects of Invention

[0022] In still another aspect, the present invention provides a method for identifying a non-aqueous fluid product, comprising: dissolving a non-aqueous fluid marker, which contains *N*-benzyl-*N*-ethylaniline represented by the above Formula 1, in a target product; and developing a color of the target product with an acid developer.

[0023]

Brief Description of Drawings

[0024] The above and other features of the present invention will now be described in detail with reference to certain exemplary embodiments thereof illustrated the accompanying drawings which are given hereinbelow by way of illustration only, and thus are not limitative of the present invention, and wherein:

[0025] FIG. 1 is a graph illustrating a change in UV-VIS absorption spectra before and after addition of an acid developer to a marker solution containing an azo disperse dye prepared in Example 1, in which a change in maximum absorption wavelength is shown.

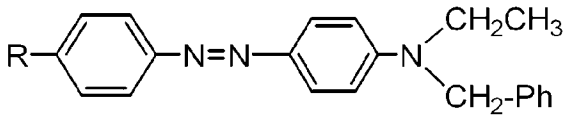
[0026]

Best Mode for Carrying out the Invention

[0027] Hereinafter, preferred embodiments of the present invention will be described in detail with reference to the accompanying drawings.

[0028] The present invention provides a non-aqueous fluid marker containing *N*-benzyl-*N*-ethylaniline represented by the following Formula 1:

[0029] **Formula 1**

[0030] 

[0031]

[0032] wherein R is a C_1 to C_{12} alkyl group, a C_1 to C_{12} alkoxy group, a C_1 to C_{12} hy-

droxyalkyl group, a C_1 to C_{12} hydroxyalkoxyalkyl group, a C_1 to C_{12} alkylphenyl group, a C_1 to C_{12} alkoxyphenyl group, a nitro group, a cyano group or a halogen atom.

[0033] The above-described substituents will be described in detail below.

[0034] The term "alkyl group" refers to a linear or branched aliphatic saturated hydrocarbon group having 1 to 12 carbon atoms. More particularly, the alkyl group may include a methyl group, an ethyl group, a *n*-propyl group, an *i*-propyl group, a *n*-butyl group, an *i*-butyl group, a *t*-butyl group, an *n*-pentyl group, an *i*-pentyl group, a *n*-hexyl group, an *i*-hexyl group, a *n*-heptyl group, an *i*-heptyl group, a *n*-octyl group, an *i*-octyl group, a *n*-nonyl group, an *i*-nonyl group, a *n*-dodecyl group, an *i*-dodecyl group, etc.

[0035] The term "alkoxy group" refers to a linear or branched aliphatic alkoxy group having 1 to 12 carbon atoms. More particularly, the alkyl group may include a methoxy group, an ethoxy group, a *n*-propyloxy group, an *i*-propyloxy group, an *n*-butoxy group, an *i* -butoxy group, a *t*-butoxy group, an *n*-pentoxy group, an *i*-pentoxy group, etc.

[0036] The term "hydroxyalkyl group" refers to a OH-alkyl group in which a hydroxyl group is bound to the aforementioned alkyl group. More particularly, the hydroxyalkyl group may include a hydroxymethyl group, a hydroxyethyl group, a hydroxy-*n*-propyl group, a hydroxy-*n*-butyl group, a hydroxy-*i*-butyl group, a hydroxy-*n*-pentyl group, a hydroxy-*i*-pentyl group, a hydroxy-*n*-hexyl group, a hydroxy-*i*-hexyl group, etc.

[0037] The term "hydroxyalkoxyalkyl group" refers to a hydroxyalkyl-O-alkyl group in which an alkoxy group is bound to the hydroxyalkyl group. More particularly, the hydroxyalkoxyalkyl group may include a hydroxyethoxymethyl group, a hydroxyethoxyethyl group, a hydroxymethoxypropyl group, a hydroxyethoxypropyl group, a hydroxyethoxybutyl group, a hydroxyethoxypentyl group, a hydroxyethoxyhexyl group, etc.

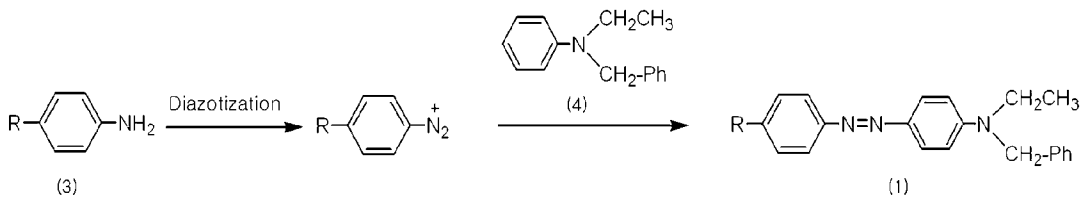
[0038] The term "alkylphenyl group" may include a methylphenyl group, an ethylphenyl group, a *n*-propylphenyl group, an *n*-butylphenyl group, etc.

[0039] The term "alkoxyphenyl group" may include a methoxyphenyl group, an ethoxyphenyl group, a *n*-propyloxyphenyl group, a *n*-butoxyphenyl group, etc.

[0040] The term "halogen atom" is a fluorine atom, a chlorine atom, a bromine atom or an iodine atom.

[0041] According to the present invention, a particularly preferred the oil marker is a compound in which R in Formula 1 is a C_1 to C_{12} alkyl group or a C_1 to C_{12} alkoxy group.

[0042] Moreover, the present invention provides a method for preparing a non-aqueous fluid marker containing *N*-benzyl-*N*-ethylaniline represented by the Formula 1. The non-aqueous fluid marker may be prepared by diazotizing an aniline derivative represented by the following Formula 3 and reacting the diazotized aniline derivative with *N* -benzyl-*N*-ethylaniline represented by the following Formula 4.

[0043] 

[0044] In the above Formulas 1 and 3, R has the same meaning as defined above.

[0045] In more detail, in the step of diazotizing an aniline derivative represented by the above Formula 3, the diazotization is performed by dispersing an aniline derivative in water or acetic acid or in a mixed solution of acetic acid and propionic acid at 0-10 °C and adding concentrated hydrochloric acid and $NaNO_2$ or HO_3SONO to react with each other. Preferably, in the mixed solution of the acetic acid and propionic acid may be mixed in a volume ratio of 5:1.

[0046] Then, the diazotized aniline derivative may be reacted with the *N*-benzyl-*N*-ethylaniline represented by the above Formula 4 to give a yellow azo disperse dye represented by the above Formula 1. The *N*-benzyl-*N*-ethylaniline may be dissolved in acetic acid or in a mixed solution of acetic acid and propionic acid in a separate container and maintained at 0-10 °C, and the prepared solution may then be added to the diazotized solution to carry out a coupling reaction at 0-10 °C. Preferably, in the mixed solution of acetic acid and propionic acid, the acetic acid and propionic acid may be mixed in a volume ratio of 5:1.

[0047] Furthermore, the present invention provides a method for identifying a target product using a non-aqueous fluid marker containing *N*-benzyl-*N*-ethylaniline represented by the above Formula 1.

[0048] The non-aqueous fluid marker containing the *N*-benzyl-*N*-ethylaniline represented by the above Formula 1 according to the present invention shows a purple color by reacting with acid developer when the non-aqueous fluid marker is dissolved in a target product. That is, when the marker according to the present invention reacts with the acid developer in the non-aqueous fluid product, a maximum absorption wavelength of the non-aqueous fluid product shifts from 410 nm to a longer wavelength of 560 nm. Therefore, the color of the non-aqueous fluid product is changed from its own color (i.e., yellow) to purple. Accordingly, it is possible to distinguish the non-aqueous fluid product including the marker according to the present invention from marker-free products. That is, when the non-aqueous fluid manufacturer dissolves the marker according to the present invention in the non-aqueous fluid product, it is possible to easily distinguish the product from other companies' products using the acid developer. Therefore, it is possible to control the quality of the product in the distribution process. The type of non-aqueous fluids to be distinguished

is not particularly limited, and the non-aqueous fluid may include gasoline, diesel, kerosene, biodiesel, bioethanol, xylene, ink, non-aqueous lubricant, solvent, and mixtures thereof. Moreover, the acid developer may include an organic acid such as dodecyl sulfonic acid, and an inorganic acid such as sulfuric acid or phosphoric acid. Preferably, the dodecyl sulfonic acid may be used as the acid developer.

[0049] Next, the present invention will be described in detail with reference to Examples, but the present invention is not limited by the following Examples.

[0050]

[0051] **EXAMPLES**

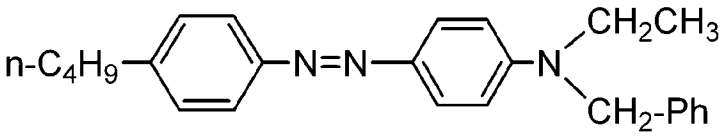
[0052] **Example 1**

[0053] 5.37 g (0.036 mol) of 4-butylaniline as a diazo compound was added to 70 ml ; of an acid mixture (where the volume ratio of acetic acid to propionic acid was 5: 1) and stirred at room temperature for about 20 minutes. Then, 7.7 ml (0.089 mol) of 35% hydrochloric acid was added to the resulting reactant and then maintained at 5-10 °C. Thereafter, 13 ml (0.039 mol) of 3*N* $NaNO_2$ was slowly added thereto so that the reaction temperature would not rise and stirred at 5-10 °C for 1.5 hours, thereby completing diazotization. Finally, a small amount of sulfamic acid was added to remove excess nitrous acid.

[0054] Meanwhile, 7.6 g (0.036 mol) of *N*-benzyl-*N*-ethylaniline as a coupler was dissolved in 20 ml of an acid mixture (where the volume ratio of acetic acid to propionic acid was 5:1) and maintained at 5-10 °C. Then, the prepared solution was slowly added to the diazotized solution and stirred at 5-10 °C for 3.5 hours. 25 g of sodium acetate (NaOAc) was added to the reaction solution and stirred at room temperature for 30 minutes. The reaction mixture was poured into 250 g of ice-water, stirred for 1 hour to produce a solid product, and filtered. In order to purify the product, the isolated product was dispersed in 100 ml of isopropyl alcohol, stirred at room temperature for 30 minutes, filtered, and then dried, thereby obtaining a marker according to the present invention as a yellow solid azo disperse dye represented by the following Formula 5.

[0055] **Formula 5**

[0056]



[0057]

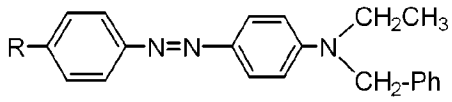
[0058] **Examples 2-5**

[0059] As yellow solid azo disperse dyes, non-aqueous fluid markers containing *N*-benzyl-*N*-ethylaniline according to the present invention were prepared in the same manner as

in Example 1, except that aniline derivatives as listed in the following Table 1 were used.

[0060] The aniline derivatives represented by the above Formula 3 prepared in Examples 1-5, the amounts of aniline derivatives used, and their synthesis yields are listed in the following Table 1.

[0061] [Table 1]



	R	Amount of diazo compound used (g)	Yield (%)
Example 1	4-C_4H_9	5.36	76.3
Example 2	4-tert-C_4H_9	5.36	78.2
Example 3	4-$C_{12}H_{25}$	9.40	41.4
Example 4	4-CH_3	3.85	69.9
Example 5	4-OCH_3	4.43	17.3

[0062]

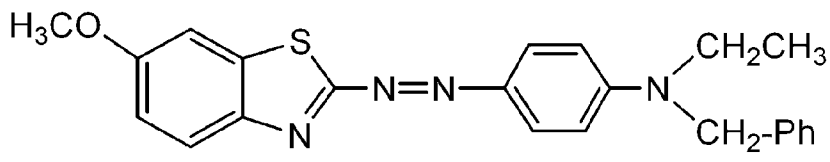
[0063] **Comparative Example 1**

[0064] 0.9 g (0.005 mol) of 2-amino-6-methoxybenzothiazole as a diazo compound was added to 10 ml of an acid mixture (where the volume ratio of acetic acid to propionic acid was 5:1) and stirred at room temperature for about 20 minutes. Then, the resulting reactant was maintained at 5-10 °C. Thereafter, 1.05 ml (0.0053 mol) of 40% HO_3 SONO was slowly added thereto so that the reaction temperature would not rise, and the resulting reactant was stirred at 5-10 °C for 1.5 hours, thereby completing diazotization. Finally, a small amount of sulfamic acid was added to remove excess nitrous acid.

[0065] Meanwhile, 1.05 g (0.005 mol) of *N*-benzyl-*N*-ethylaniline as a coupler was dissolved in 10 ml of an acid mixture (where the volume ratio of acetic acid to propionic acid was 5:1) and maintained at 5-10 °C. Then, the prepared solution was slowly added to the diazotized solution and stirred at 0-5 °C for 2 hours. 2.5 g of NaOAc was added to the reaction mixture and stirred at room temperature for 30 minutes. The reaction mixture was poured into 50 g of ice-water, stirred for 1 hour to produce a solid product, and filtered. In order to purify the product, the isolated product was dispersed 20 ml of isopropyl alcohol, stirred at room temperature for 30 minutes, filtered, and then dried, thereby obtaining 1.78 g of a red solid azo disperse

dye represented by the following Formula 2 (yield = 88.6 %).

[0066] **Formula 2**

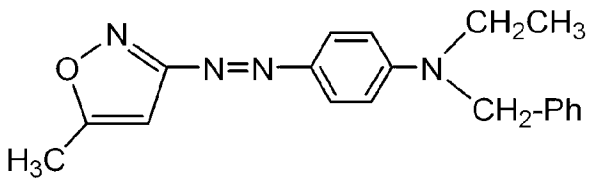
[0067] 

[0068]

[0069] **Comparative Example 2**

[0070] The compound of Formula 6 was prepared according to the method described in Comparative Example 1 using 0.49 g (0.005 mol) of 3-amino-5-methylisoxazole as diazo compound, thereby obtaining 1.20 g of a yellow solid azo disperse dye (yield = 75 %) represented by the following Formula 6.

[0071] **Formula 6**

[0072] 

[0073] The mass spectra (*m/e*) and ^{1}H NMR spectra data for confirming the respective structures of the azo disperse dyes containing *N*-benzyl-*N*-ethylaniline prepared in Examples 1-5 and Comparative Examples 1 and 2 are listed in the following Table 2.

[0074]

[Table 2]

	m/e	^{1}H NMR* (δ ppm)
Example 1	371	A; 0.92(3H, t), 1.25(3H, t), 1.36(2H, sextet), 1.62(2H, qt), 2.66(2H, t), 3.64(2H, q), 4.70(2H, s), 6.84(2H, d), 7.23-7.35(7H, m), 7.73(2H, d), 7.78(2H, d)
Example 2	371	C; 1.26(3H, t), 1.33(9H, s), 3.56(2H, q), 4.61(2H, s), 6.75(2H, d), 7.20-7.34(5H, m), 7.45(2H, d), 7.74(2H, d), 7.81(2H, d)
Example 3	483	C; 0.86(3H, t), 1.24-1.28(20H, m), 1.57-1.64(4H, m), 2.63(2H, t), 3.56(2H, q), 4.62(2H, s), 6.75(2H, d), 7.20-7.34(7H, m), 7.73(2H, d), 7.81(2H, d)
Example 4	329	C; 1.25(3H, t), 2.38(3H, s), 3.55(2H, q), 4.61(2H, s), 6.75(2H, d), 7.20-7.31(7H, m), 7.72(2H, d), 7.80(2H, d)
Example 5	345	A; 1.25(3H, t), 3.63(2H, q), 3.86(3H, s), 4.70 (2H, s), 6.83(2H, d), 7.03(2H, d), 7.21-7.35(7H, m), 7.74(2H, d), 7.78(2H, d)
Comp. Example 1	402	A; 1.30(3H, t), 3.72(2H, q), 3.89(3H, s), 4.81 (2H, s), 6.94(2H, d), 7.09(1H, dd), 7.26-7.37(5H, m), 7.50(1H, d), 7.83-7.90(3H, m)
Comp. Example 2	320	A; 1.27(3H, t), 2.44(3H, s), 3.68(2H, q), 4.76 (2H, s), 6.36(1H, s), 6.87(2H, d), 7.24-7.36(5H, m), 7.80(2H, d)
* Solvent for ^{1}H NMR ; $CDCl_3$(C), Acetone-d_6 (A)		

[0075]

[0076] **<u>Test Example 1</u>: Test for acid reactivity of yellow azo disperse dyes**

[0077] (1) Preparation of marker solution

[0078] A proper amount of each of the azo disperse dyes prepared in Examples 1-5 and Comparative Examples 1 and 2 was taken and diluted with xylene in a 25 ml volumetric flask, and 1 ml of the azo disperse dye solution was taken and diluted with xylene in a 10 ml volumetric flask to prepare a marker solution. Concentrations of the marker solutions are listed in the following Table 3.

[0079] (2) Preparation of acid developer solution

[0080] 3 g of xylene was added to 7 g of dodecylbenzene sulfonic acid to prepare a coloring agent solution.

[0081] (3) Measurement of UV-VIS molar absorption coefficients and maximum absorption wavelength

[0082] The UV-VIS molar absorption coefficients (ε) and maximum absorption wavelengths (λ_{max}) of the samples prepared in (1) were measured. Then, 10 μl of the acid developer

solution prepared in (2) was added to each sample and stirred for 1 minute. Then, the UV-VIS molar absorption coefficient (ε) and maximum absorption wavelength (λ_{max}) of each sample were measured to observe a change in color. The UV-VIS molar absorption coefficient and maximum absorption wavelength were measured using a UV-VIS-NIR spectrophotometer (UV-3101 PC, SHIMADZU). The measurement results are listed in the following Table 3 and shown in FIG. 1.

[0083] [Table 3]

	Concentration (mg /L)	Before addition of developing agent		After addition of developing agent		Wavelength shift (nm)
		λ_{max}*	ε_{max}/ $Lmol^{-1}cm^{-1}$	λ_{max}*	ε_{max}/ $Lmol^{-1}cm^{-1}$	
Example 1	17.12	410	3.261×10^4	562	4.537×10^4	152
				542	4.507×10^4	
Example 2	14.84	409	3.235×10^4	563	4.633×10^4	154
				542	4.590×10^4	
Example 3	16.64	410	3.086×10^4	562	4.488×10^4	152
				542	4.464×10^4	
Example 4	15.00	409	3.216×10^3	562	4.464×10^4	153
				539	4.424×10^4	
Example 5	16.04	409	2.850×10^4	565	1.693×10^4	67
				476	2.089×10^4	
Comp. Example 1	15.40	489	4.252×10^4	512	3.310×10^4	23
Comp. Example 2	17.28	408	3.152×10^4	451	2.052×10^4	43
* Solvent for UV measurement: Xylene						

[0084]

[0085] The alkyl group-substituted yellow azo disperse dyes prepared in Examples 1 -4 showed a distinct purple color and a maximum absorption wavelength was shifted to a longer wavelength by 150 nm or more when the dodecylbenzene sulfonic acid solution was added to the marker solution.

[0086] The methoxy group-substituted yellow azo disperse dye prepared in Example 5 had a higher absorbance at 476 nm than at 565 nm as shown in the above Table 3, when the

dodecylbenzene sulfonic acid solution was added to the marker solution. The color change of the marker solution could be observed, however the appearance of color was not distinctive.

[0087] In Comparative Example 1 using 2-amino-6-methoxybenzothiazole as the diazo compound, when the dodecylbenzene sulfonic acid solution was added to the marker solution, a maximum absorption wavelength of the azo disperse dye was shifted to a longer wavelength by 23 nm, and thus the color change of the marker solution was insignificant. Moreover, the compound of Formula 2 prepared in Comparative Example 1 was not suitable for use as a marker since the compound itself exhibited a red visible color in the xylene.

[0088] In Comparative Example 2 using 3-amino-5-methylisoxazole as the diazo compound, when the dodecylbenzene sulfonic acid solution was added to the marker solution, a maximum absorption wavelength of the azo disperse dye was shifted to a longer wavelength by 43 nm, and thus the color change of the marker solution was insignificant. The azo disperse dye was not suitable for use as a marker since the azo disperse dye exhibited a dark yellow visible color even when the acid developer was added to the marker solution.

[0089] Therefore, it is considered that the alkyl group-substituted yellow azo disperse dyes prepared in Examples 1-4, which exhibits their distinct colors by addition of dodecylbenzene sulfonic acid solution as the acid developer, can be more effectively used as an acid-reactive marker.

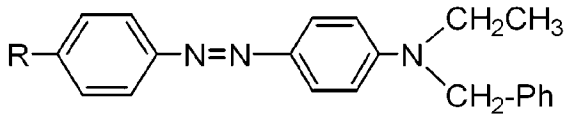
[0090] As described above, the marker containing *N*-benzyl-*N*-ethylaniline according to the present invention exhibits high solubility in non-aqueous fluid products, has excellent storage stability, can be detected even in a small amount, and can develop a distinct color by reaction with an organic acid such as dodecylbenzene sulfonic acid and an inorganic acid such as sulfuric acid or phosphoric acid in the non-aqueous fluid product. Therefore, when the marker is added to a product, the product can be easily distinguished from other companies' products containing no marker or illegally distributed products such as adulterated gasoline, which makes it possible to easily control the quality of the product in the distribution process.

[0091] The invention has been described in detail with reference to preferred embodiments thereof. However, it will be appreciated by those skilled in the art that changes may be made in these embodiments without departing from the principles and spirit of the invention, the scope of which is defined in the appended claims and their equivalents.

Claims

[Claim 1] A non-aqueous fluid marker comprising *N*-benzyl-*N*-ethylaniline represented by the following Formula 1:

Formula 1



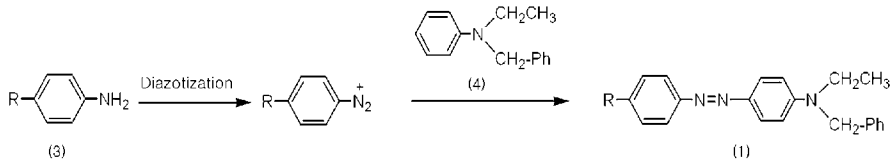
wherein R is a C_1 to C_{12} alkyl group, a C_1 to C_{12} alkoxy group, a C_1 to C_{12} hydroxyalkyl group, a C_1 to C_{12} hydroxyalkoxyalkyl group, a C_1 to C_{12} alkylphenyl group, a C_1 to C_{12} alkoxyphenyl group, a nitro group, a cyano group or a halogen atom.

[Claim 2] The non-aqueous fluid marker according to claim 1, wherein R is a methyl group, a *n*-butyl group, a *t*-butyl group, an *n*-dodecyl group or a methoxy group.

[Claim 3] A method for preparing a non-aqueous fluid marker, comprising:

diazotizing an aniline derivative represented by the following Formula 3; and

reacting *N*-benzyl-*N*-ethylaniline represented by the following Formula 4 with the diazotized aniline derivative:

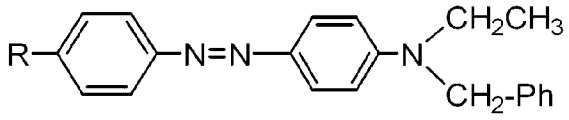


wherein R is a C_1 to C_{12} alkyl group, a C_1 to C_{12} alkoxy group, a C_1 to C_{12} hydroxyalkyl group, a C_1 to C_{12} hydroxyalkoxyalkyl group, a C_1 to C_{12} alkylphenyl group, a C_1 to C_{12} alkoxyphenyl group, a nitro group, a cyano group or a halogen atom.

[Claim 4] A method for identifying a non-aqueous fluid product, comprising:

dissolving a non-aqueous fluid marker, which contains *N*-benzyl-*N*-ethylaniline represented by the following Formula 1, in a target product; and developing a detectable color ofthe target product with an acid developer:

Formula 1



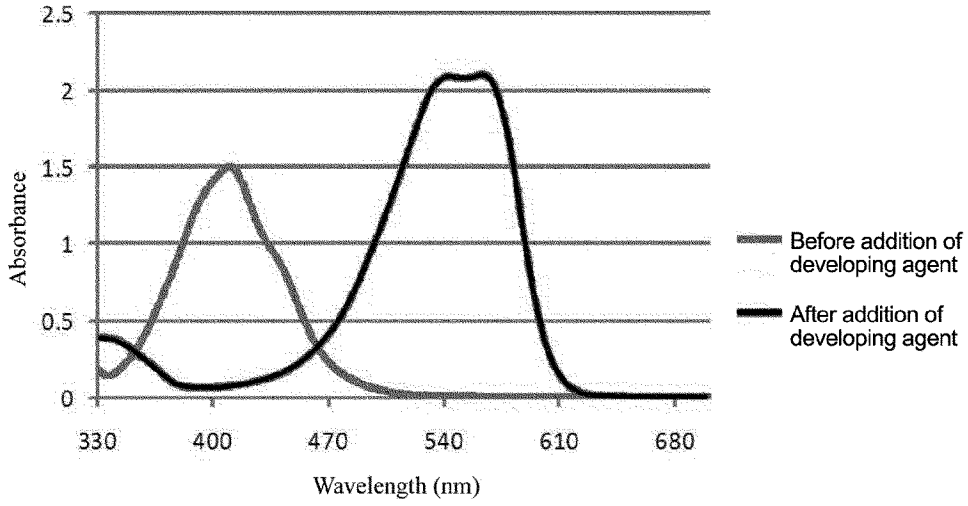
wherein R is a C_1 to C_{12} alkyl group, a C_1 to C_{12} alkoxy group, a C_1 to C_{12}

$_{12}$ hydroxyalkyl group, a C_1 to C_{12} hydroxyalkoxyalkyl group, a C_1 to C_{12} alkylphenyl group, a C_1 to C_{12} alkoxyphenyl group, a nitro group, a cyano group or a halogen atom.

[Claim 5] The method according to claim 4, wherein the target product is at least one selected from the group consisting of gasoline, diesel, kerosene, biodiesel, bioethanol, xylene, ink, and non-aqueous lubricant.

[Claim 6] The method according to claim 4, wherein the acid developer is dodecylbenzene sulfonic acid, sulfuric acid or phosphoric acid.

[Fig. 1]


2.5
2
1.5
1
0.5
0
Absorbance
330
400
470
540
610
680
Wavelength (nm)
Before addition of developing agent
After addition of developing agent

INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2012/000990

A. CLASSIFICATION OF SUBJECT MATTER

G01N 31/22(2006.01)i, G01N 21/78(2006.01)i, G01N 21/31(2006.01)i, C09B 29/34(2006.01)i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

G01N 31/22; A61Q 5/06; C09D 11/02; C09B 29/085; G01N 31/00; C09B 69/00; G01K 3/00

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Korean utility models and applications for utility models
Japanese utility models and applications for utility models

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

eKOMPASS(KIPO internal) & Keywords: KOMPASS, Google

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	YAZDANBAKHSH, M. R. et al., Journal of Molecular Liquids, 4 December 2010, Vol. 151, pages 107-112. See abstract; scheme 1; tables 1, 3; figures 1, 2, 4;	1-3
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A	US 2007-0218206 A1 (REICHERT, H. AND MULLER, B.) 20 September 2007 See entire document.	1-6
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☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents:

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents,such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
22 AUGUST 2012 (22.08.2012)	**24 AUGUST 2012 (24.08.2012)**

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

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