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[54] **SUPPORT FOR CHROMOGENIC GLYCOL TEST REAGENT**

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[58] **Field of Search**..... **23/253 TP**

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[57]

ABSTRACT

An improved supporting medium for a chromogenic reagent for use in an aldehyde detection test. The chromogenic reagent, preferably 3-methyl-2-benzothiazolinone hydrazone hydrochloride monohydrate (MBTH), is absorbed on a porous polymeric material. The preferred polymeric supporting medium is a film of porous polyethylene. The chromogenic reagent supported on the porous polymer finds particular utility in a test for determining the presence of glycol in engine oil.

11 Claims, No Drawings

SUPPORT FOR CHROMOGENIC GLYCOL TEST REAGENT

CROSS-REFERENCE TO RELATED APPLICATIONS

This application is related to U.S. Pat. No. 3,635,677 filed Apr. 30, 1970 and titled "GLYCOL DETECTION KIT"; and U.S. Pat. No. 3,645,696 filed Apr. 30, 1970 and titled "METHOD TO STABILIZE A CHROMOGENIC TEST REAGENT FOR GLYCOL," both patents being assigned to the assignee of the present application and having been co-pending therewith.

BACKGROUND OF THE INVENTION

The present invention relates to an improved supporting medium for a chromogenic reagent for use in an aldehyde detection test. More particularly, the invention discloses the use of a porous polymeric material, preferably a film of porous polymeric material, as the supporting medium for the chromogenic reagent. The chromogenic reagent supported on the porous polymer finds particular utility in a test for determining the presence or absence of an aldehyde derived from glycol in engine oils.

The presence of contaminants in engine oil may have damaging effects upon the performance of the engine and its internal parts. The cleanliness of the internal parts of an engine depends upon the equilibrium between the oil polluting substances and the remaining effective lubricating oil molecules. Various methods are available for testing engine oils as to their effective lubrication and cleansing properties upon engine parts. Both laboratory and field tests have shown that ethylene glycol is one of the more particularly hazardous contaminants which may enter engine oil. The ethylene glycol is derived from the anti-freeze added to the engine's cooling system. Through abusive wear of the engine and failure of sealing members within the engine glycol may enter the crankcase and lubricating system of the engine and cause pasty emulsions which may plug oil filters, pump screens, oil feed lines and passages within the engine. The presence of ethylene glycol also presents a problem in the engine combustion chamber where temperatures are high enough to convert the glycol into a varnish-like substance which causes valves to stay open, valve lifters to bend in their guards, and piston rings to stick in their grooves. Consequences of the above-mentioned malfunctions of the engine may result in extreme engine abuse, engine lifetime diminishment and eventual engine failure. The elimination of ethylene glycol in the crankcase oil is a matter of extreme importance in maintenance of the engine in that no engine oil additives appear to be available on the market to combat the incompatibility properties of glycol.

The most common method used to combat ethylene glycol contamination in engine oil and subsequent seizure of engine parts is to change the engine oil periodically so that the concentration of ethylene glycol never exceeds a maximum tolerable concentration which would damage the engine's moving members. More than about 50 p.p.m. ethylene glycol is usually considered to be excessive. What is required is an ethylene glycol testing method which will indicate the concentration of ethylene glycol after certain periodic intervals of engine-hour usage, such that the engine may be

serviced and the oil changed. This ethylene glycol test method must include a procedure by which servicing personnel may periodically check engine oil for glycol concentration without a rigorous analytical technique being required. The test must, therefore, be simple enough for layman's usage and also provide positive identification of the presence of ethylene glycol. To date, no suitable ethylene glycol test method is available for service station or industrial usage. It has been suggested that the presence of glycol in engine oil may be detected by treating a sample of oil with an oxidizer to oxidize any glycol present to aldehyde and then utilizing a chromogenic reagent adsorbed on a particulate support to detect the presence of the aldehyde. A problem with chromogenic reagent adsorbed on the surface of a particulate supporting medium is poor bonding between the reagent and the supporting medium with subsequent separation.

SUMMARY OF THE INVENTION

It is an object of this invention to provide an improved supporting medium for a chromogenic reagent for use in an aldehyde detection test.

It is another object of this invention to provide a kit for the detection of ethylene glycol in crankcase oil which utilizes a stabilized chromogenic reagent which is tightly bound within the pores of a porous polymeric material.

It is still another object of this invention to provide an improved process for the detection of glycol in oil.

Yet other objects will be apparent to those skilled in the art from this disclosure.

The foregoing objects are achieved in accordance with this invention. Broadly, this invention consists of a stable supported chromogenic reagent comprising a chromogenic aldehyde reagent selected from the group consisting of 3-methyl-2-benzothiazolinone hydrazone hydrochloride monohydrate, salicylaldehyde, p-nitrobenzaldehyde, 2-hydrazinobenzothiazole, and 2-hydrazinobenzothiazole-4-nitrobenzenediazonium fluoroborate wherein said chromogenic reagent is adsorbed on a porous polymeric supporting medium; and a method for the detection of glycol in oil comprising the steps:

- introducing a sample of the oil to be tested into an aqueous solution of an oxidizer which preferentially oxidizes glycol to an aldehyde;
- mixing the oil and aqueous solution containing the oxidizer;
- separating the aqueous and oil phases; and
- contacting a sample of said aqueous phase with the above chromogenic reagent adsorbed on a porous polymeric supporting medium.

Thus by the practice of this invention, an improved supporting medium for a chromogenic aldehyde reagent is provided. Moreover, an improved process for the detection of ethylene glycol in engine oil and an improved kit for use in said detection process are provided by the instant invention.

DETAILED DESCRIPTION OF THE INVENTION

The objects of this invention are therefor accomplished by an improved supporting medium for a chromogenic aldehyde reagent and a method wherein the supported chromogenic reagent of the invention is used to detect the presence or absence of glycol in oil. The

improved supporting medium for the chromogenic reagent is a porous polymeric material, the use of which results in a stabilized chromogenic reagent tightly bound within the pores of the porous polymeric supporting medium. Use of the supported chromogenic reagent of the invention in a test to detect the presence or absence of a glycol, especially ethylene glycol, in oil involves introducing a sample of the oil to be tested into an aqueous solution of an oxidizer which preferentially oxidizes the glycol to an aldehyde. The oil and the aqueous solution of oxidizer are thoroughly mixed, and the mixture is thereafter allowed to separate into the oil phase and the aqueous phase. A portion of the aqueous phase is then brought into contact with a chromogenic aldehyde reagent absorbed on a porous polymeric supporting medium. Whether or not the chromogenic reagent develops the appropriate color indicates the presence or absence of glycol in the oil; the intensity of the color indicates the amount of glycol present.

Most oxidizers which will transform glycols into aldehydes may be used in the present invention. However, the oxidizer must have the characteristic of not tinting the aqueous solution such that the colorimetric determination may not be used. Accordingly, standard oxidizers such as potassium permanganate and ferric sulfate would not be suitable for the present invention as they would leave a distinct color in the aqueous phase which would conceal the color change of the chromogenic aldehyde reagent. Suitable oxidizers for use in practicing the instant invention include sodium periodate, potassium periodate, hydrogen peroxide, sodium perborate, ceric nitrate, ceric sulfate, and lead tetracetate. In particular, the sodium and potassium periodates are preferred oxidizers which selectively oxidize ethylene glycol to formaldehyde without side reactions which form other oxidized compounds. In most cases, the oxidizer, in particular the periodate solution, will be about 0.001 to 1.0 molar, and preferably about 0.01 to 0.5 molar, in concentration. By use of concentrations within this range, an economic quantity of the oxidizer is utilized and strong oxidation does not take place. In most instances, between about 0.5 and about 10 milliliters, and preferably about 1 to 3 milliliters, of oxidizing solution per milliliter of oil sample will be sufficient to fully oxidize the ethylene glycol contained in an oil sample withdrawn from an engine crankcase.

In the field application of a preferred embodiment of the present invention, generally 5 to 10 milliliter oil samples are withdrawn from the engine crankcase by means of a piece of tubing or by a syringe-type instrument introduced into the crankcase through the oil dip tube. The engine oil is withdrawn and dropped into the solution of oxidizer held in a container. The container is then sealed and vigorously shaken. After thorough mixing has occurred, the solution is allowed to separate into a lower aqueous phase and an upper oil phase. The aqueous phase, containing formaldehyde resulting from the oxidation of any ethylene glycol present in the oil phase, may be used to determine whether or not formaldehyde is present and, if present, its semiquantitative concentration.

Many chromogenic aldehyde reagents exist, but the reagent used must be highly specific for formaldehyde or other aldehyde and not be influenced by other substances which may be oxidized or may be contained as additives or original components of the engine oil. Examples of chromogenic aldehyde reagents which are

specific to aldehydes and may be utilized in accordance with the present invention are 3-methyl-2-benzothiazolinone hydrazone hydrochloride monohydrate, salicylaldehyde, p-nitrobenzaldehyde, 2-hydrazinobenzothiazole, and 2-hydrazinobenzothiazole-4-nitrobenzenediazonium fluoroborate. It is pointed out that oil samples tested in accordance with the present invention, especially crankcase oils, are often acidic by nature. The chromogenic aldehyde reagent used must therefore preferably be able to respond to the presence of an aldehyde in an acidic medium when testing such acidic oils. This criterion eliminates a number of excellent chromogenic aldehyde reagents which will not perform under acidic conditions. For example, reagents such as 2-hydrazinobenzothiazole and 2-hydrazinobenzothiazole-4-nitrobenzenediazonium fluoroborate are excellent for the detection of aliphatic, aromatic, and heterocyclic aldehydes. However, these reagents require an alkaline medium and are therefore not suitable for practicing the preferred embodiment of this invention when the oil sample tested is an acidic crankcase oil.

Another criterion for preferred aldehyde reagents is the stabilization of the color after introduction of the aldehyde into the presence of the chromogenic aldehyde reagent. The color should remain stable for several minutes so that the operator may observe the reagent color against predetermined charts or a blank for a quantitative determination of the amount of glycol present in the oil sample. A study made by Sawicki, et al., *Analytical Chemistry*, 33, No. 1, 93 (1961) has indicated that 3-methyl-2-benzothiazolinone hydrazone hydrochloride monohydrate (MBTH) is an excellent reagent for the determination of formaldehyde and thus is a preferred reagent in practicing the preferred embodiment of the instant invention which involves detection of ethylene glycol in engine oil.

As with the oxidizing agents, the porous polymeric supporting media used to absorb the chromogenic aldehyde reagent should be translucent or opaque such that any color imparted from the support will not interfere with the chromogenic test. Therefore, it is a necessary criterion of the porous polymeric supporting medium that it not have an interfering background color which will interfere with the color test.

Examples of suitable porous polymeric supporting media are porous polyethylene, polypropylene, nylon, and polystyrene. The porous polymer may be particulate, i.e., powder or chips, or it may be a film such as a disk or small rectangular or square sheet. The preferred porous polymeric support is a porous polyethylene film; e.g., a disk or small rectangular or square sheet.

Means for preparing porous polymeric materials are well known, and the method of preparing a porous polymeric material is not critical to the practice of this invention. For example, polyethylene or polypropylene may be rendered porous by means of unreacted monomer contained therein. In this procedure, unreacted gaseous monomer is dissolved in the polymer which is prepared in a closed system under high pressure. Release of the pressure causes the unreacted gaseous monomer to come out of solution in the polymer with the resultant formation of open pores by the expanding gas. Porous polystyrene, for example, is commonly prepared by the impregnation of the polymer with a

volatile hydrocarbon such as pentane followed by heating, whereupon the vaporized pentane forms open pores in the polymer. Another common method of forming pores in polymers is to use carbon dioxide as a blowing, or pore-forming, agent. For example, a metal carbonate may be physically incorporated in the polymer followed by contacting the polymer-metal carbonate mixture with an acid at a temperature above the softening point of the polymer whereby the liberated carbon dioxide forms pores in the polymer. Alternatively, the carbon dioxide may be injected under high pressure into fluid polymer, for example in the barrel of a plastics extruder, and on release of the pressure the escaping carbon dioxide forms a porous polymer. Polymers may also be rendered porous by well known chemical blowing agents which, on heating, decompose to gaseous products. Also, soluble salts such as sodium chloride may be physically incorporated in the polymer and subsequently leached out by a solvent for the salt to thereby leave pores in the polymer. The foregoing merely serves to illustrate how porous polymers may be prepared, but it will be understood that the method of preparing the porous polymer is not critical to the practice of my invention.

It will be apparent to those skilled in the art that the pore size in the porous polymeric supporting medium should broadly be large enough to accommodate the chromogenic aldehyde reagent molecule and that the optimum pore size will vary depending on the size of the chromogenic reagent molecule. Generally, a pore size within the range of about 30 to 60 microns is desirable and, when the chromogenic reagent is the preferred MBTH, the preferred pore size has been found to be from about 40 to 50 microns.

In the preparation of a supported chromogenic reagent, which is generally unstable on exposure to air and thereby rendered unsuitable for the determination of aldehydes, the chromogenic reagent generally is dissolved in a solvent. In most applications, the solvent used for making the solution is deionized or distilled water. After the solution is formed it is brought into contact with a suitable porous polymeric supporting medium which will absorb the chromogenic reagent. An inert atmosphere may be maintained above the supporting medium during the absorption process so that no oxidation of the chromogenic reagent will occur during the absorption step. The absorption step may be enhanced by simultaneously drying the chromogenic reagent on the supporting medium to thereby strip off the solvent. This drying process may take from five minutes to an hour depending on the amount of solution brought into contact with the supporting medium. As mentioned above, the preferred chromogenic aldehyde reagent is 3-methyl-2-benzothiazolinone hydrazone hydrochloride monohydrate (MBTH). The MBTH solution in deionized or distilled water generally has a concentration of about 0.5 to 5 percent by weight.

The inert atmosphere during the absorption step is maintained by means of an inert gas such as nitrogen or argon. When, for example, the porous polymeric supporting medium is particulate, a column of the supporting medium and the chromogenic reagent solution may have the inert gas passed therethrough so that the inert gas is constantly in contact with the chromogenic reagent and the supporting medium. When the porous polymeric supporting medium is in the form of a film,

the film may be immersed in the solution of chromogenic reagent and an inert gas, advantageously with stirring, may be bubbled into the solution. The drying procedure for removal of the solvent from the solution of chromogenic reagent, which may be simultaneous with the absorption step, is carried out at temperatures below about 100° C so that no thermal degradation of the chromogenic reagent will occur. The drying step may be carried out in an inert atmosphere or it may be carried out under reduced pressure which, in effect, lowers the oxygen concentration. The chromogenic aldehyde reagent in effect forms a complex with the porous polymeric supporting medium so that after it is absorbed on the supporting medium it will remain stable and will not be subject to attack by atmospheric oxygen.

It has been found that when the chromogenic reagent supported on a porous polymeric supporting medium comprises about 2 to 6 weight percent of chromogenic reagent, sufficient reagent is provided to achieve the desired results, namely the detection of glycol in oil. It is preferred that the supported chromogenic reagent comprise about 3 to 5 weight percent of chromogenic reagent. The preferred reagent is MBTH.

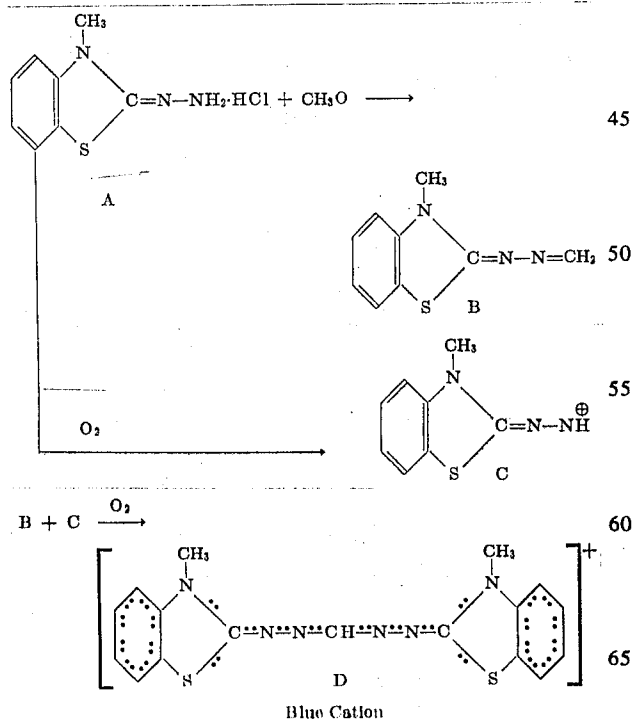
In testing a sample of oil for the presence of glycol, a mixture of the oil and the aqueous solution of oxidizer in the proportions described above are shaken vigorously for at least 1 minute and preferably for at least 2 minutes to allow intimate contact of the two phases. The oil-aqueous oxidizer mixture is allowed to stand for at least about 10 minutes and preferably for about 15 to 20 minutes to allow the phases to separate and the oxidation of glycol to be essentially completed. The aqueous phase, containing the aldehyde resulting from oxidation of the glycol, is then brought into contact with the chromogenic reagent supported on the porous polymeric supporting medium. The time for color development, depending on the concentration of the aldehyde, will be anywhere from instantaneous to about 5 to 10 minutes.

In one embodiment of this invention, chromogenic reagent absorbed on a particulate porous polymeric supporting medium is introduced into a container such as a hypodermic syringe. It has been found that about 2 cubic centimeters of the treated polymer particles in a 5 ml. syringe is an appropriate quantity. A sample of the aqueous phase is withdrawn from the container holding the oil and aqueous phases and brought into contact with the supported chromogenic reagent in the syringe by means of the syringe plunger. Generally, from about 0.5 to about 2.0 ml. of aqueous phase are used per cubic centimeter of supported chromogenic reagent. The reagent will turn color within about 10 minutes if a glycol is present in the oil.

In another embodiment of the present invention, chromogenic reagent absorbed on a porous polymeric film is sealed in a receptacle such as a plastic pillow pack or a small plastic beaker. The receptacle serves not only to protect the chromogenic reagent absorbed on the film from air and, to at least some extent, light, but it serves also as a container for carrying out the test. A piece of film about 1/16 inch thick by about 1/2 inch in diameter or 1/2 inch square has been found to be an appropriate size. A sample of the aqueous phase is withdrawn from the container holding the oil and aqueous phases and is placed in the receptacle containing the chemically treated film. The amount of aqueous

phase is not critical provided there is sufficient to thoroughly contact the treated film. As noted above, the reagent will turn color within about 10 minutes if a glycol is present in the oil.

In practicing the invention using sodium periodate as the oxidizer and MBTH as the chromogenic aldehyde reagent, the reactions occurring may be summarized as follows. Sodium periodate oxidizes the ethylene glycol to formaldehyde. The formaldehyde then contained in the aqueous phase is withdrawn and contacted with the supported MBTH chromogenic reagent. The formaldehyde reacts with the MBTH to form the azine. In addition, some MBTH is oxidized to a reactive cation which combines with the azine to form a blue dye. Therefore, the normally translucent chromogenic formaldehyde reagent absorbed on a porous polymeric support will turn a dark blue if ethylene glycol is present in the oil. A blank may be run by taking an uncontaminated sample of the oil, mixing it with the aqueous periodate solution, and then contacting the separated aqueous phase with the supported chromogenic formaldehyde reagent. The reagent turns purple to indicate that no ethylene glycol is present in the blank. Shades of color between blue and the purple color of the blank indicate the concentration of ethylene glycol. A color chart may be prepared to show the colors corresponding to various amounts of ethylene glycol in parts per million which are contained in the engine oil. With this method, the operator knows at any time what the ethylene glycol concentration is and when the oil must be changed. Each time the oil is checked, the reading is recorded until a maximum allowable concentration of ethylene glycol in the engine oil is detected. The oil is then changed and the process repeated. The equations are formulated below for the reaction of formaldehyde with 3-methyl-2-benzothiazolinone hydrazone hydrochloride (A) to form the azine (B), oxidation of (A) to a reactive cation (C), and formation of the blue cation (D).



In a preferred embodiment of the instant invention, MBTH is absorbed in the desired concentration on a film of porous polyethylene and the chemically treated film is sealed in a small plastic, disposable beaker until such time as it is used in a test for the presence of ethylene glycol in engine oil.

In the test for the presence of ethylene glycol in engine oil, an oil sample is withdrawn from the crankcase by means of a plastic syringe of about 10 ml. capacity and equipped with a suitable length of small bore plastic tubing. The oil sample is discharged into a container, for example a one ounce vial, containing approximately an equal volume of an aqueous sodium periodate solution. The mixture in the closed vial is vigorously shaken by hand for at least about 1 minute and is then allowed to stand for at least about 10 minutes to allow separation of the phases and oxidation of ethylene glycol to formaldehyde. The beaker containing the porous polyethylene film having MBTH absorbed thereon is unsealed and by means of, for example, a hypodermic syringe, about 2 to 5 ml. of the aqueous solution are added thereto. The presence or absence of ethylene glycol, and its concentration if present, can be determined by observing whether the chemically treated polyethylene film is purple, dark blue, or an intermediate color.

The present invention will be further illustrated by the following examples.

EXAMPLE I

A number of porous polyethylene squares about one-half inch on a side by about 1/16 inch thick and having an average pore size of about 45 microns are weighed and placed in a 4 weight percent aqueous solution of MBTH contained in an evaporation flask. Using an infrared lamp for heat, the water is evaporated under reduced pressure, leaving a coating of MBTH absorbed on the polyethylene squares. The chemically treated polyethylene squares are reweighed and are found to contain 3.2 weight percent absorbed MBTH.

The chemically treated polyethylene squares are individually placed in 5 ml. plastic beakers and sealed therein by welding a film of polyvinyl chloride over the mouth of each beaker. The sealed beakers may then be stored in a light proof container until ready for use.

EXAMPLE II

Motor oil compositions containing measured amounts of ethylene glycol are prepared. The motor oil compositions, respectively containing 1,000 p.p.m., 100 p.p.m. and 25 p.p.m., as well as a blank motor oil containing no ethylene glycol, are each shaken for two minutes in a 1:1 volume ratio with aqueous sodium periodate solutions of various concentrations. The oil-aqueous sodium periodate solutions are then allowed to stand for 10 or 20 minutes to allow for phase separation and oxidation of ethylene glycol to formaldehyde. A portion of the aqueous phase in every case is then placed in a beaker containing a MBTH treated polyethylene square described in Example I. Development of a blue color indicates the presence of ethylene glycol

in the motor oil while a purple color indicates a negative test for the presence of ethylene glycol. In every case, color development is detected within one minute. The results of the tests are set forth in the table.

NaIO ₄	Ethylene Glycol p.p.m.	Color of MBTH Treated Polyethylene	
		Phase Separation Time	
		10 min.	20 min.
0.10M	1000	Blue	—
	100	Purple	—
	25	Purple	—
	—	Purple	—
0.05M	1000	Blue	—
	100	Light Blue	—
	25	Light Blue	—
	—	Purple	—
0.025M	1000	Blue	—
	100	Blue	—
	25	Purple	Blue
	—	Purple	—
0.010M	1000	Blue	—
	100	Blue	—
	25	Purple	Blue
	—	Purple	—

From the Table, it is seen that, by the practice of this invention, ethylene glycol concentrations in oil that are as low as 25 p.p.m. may be detected. Thus by use of the present invention, the ethylene glycol content in motor oil may be specifically determined by means of an improved supported chromogenic reagent. The invention provides the ability to warn of ethylene glycol leakage into the engine crankcase and to avoid severe damage therefrom.

While the present invention has been described above with respect to certain embodiments thereof, it will be understood by those skilled in the art that various changes and modifications may be made without departing from the spirit and scope of the invention as set forth herein.

I claim:

1. A stable supported chromogenic reagent comprising a chromogenic aldehyde reagent selected from the group consisting of 3-methyl-2-benzothiazolinone hydrazone hydrochloride monohydrate, salicylaldehyde, p-nitrobenzaldehyde, 2-hydrazinobenzothiazole, and 2-hydrazinobenzothiazole-4-nitrobenzenediazonium fluoroborate wherein said chromogenic reagent is absorbed on a porous polymeric supporting medium having a pore size within the range of from about 30 to 60 microns.

2. The supported chromogenic reagent of claim 1 wherein the porous polymeric supporting medium is

selected from the group consisting of polyethylene, polypropylene, polystyrene, and nylon.

3. The supported chromogenic reagent of claim 2 wherein the polymer is in a physical form selected from the group consisting of particles and film.

4. The supported chromogenic reagent of claim 3 wherein said chromogenic reagent is 3-methyl-2-benzothiazolinone hydrazone hydrochloride monohydrate and the porous polymeric supporting medium is a polyethylene film having a pore size within the range of from about 40 to 50 microns.

5. The supported chromogenic reagent of claim 4 which contains about 2 to 6 weight percent of 3-methyl-2-benzothiazolinone hydrazone hydrochloride monohydrate.

6. The supported chromogenic reagent of claim 5 wherein said supported chromogenic reagent is sealed in a plastic beaker.

7. A method for the detection of glycol in oil comprising the steps:

a. introducing a sample of the oil to be tested into an aqueous solution of an oxidizer which preferentially oxidizes glycol to an aldehyde;

b. mixing the oil and aqueous solution containing the oxidizer;

c. separating the aqueous and oil phases; and

d. contacting a sample of said aqueous phase with the supported chromogenic reagent of claim 1.

8. The method of claim 7 wherein the oxidizer is selected from the group consisting of sodium periodate, potassium periodate, hydrogen peroxide, sodium perborate, ceric nitrate, ceric sulfate, and lead tetraacetate; and the supported chromogenic reagent is 3-methyl-2-benzothiazolinone hydrazone hydrochloride monohydrate supported on a porous polyethylene film having a pore size within the range of from about 40 to 50 microns.

9. The method of claim 8 wherein the oil is acidic.

10. The method of claim 9 wherein the oxidizer is about a 0.001 to 1.0 molar aqueous solution of sodium periodate, and the oil and the aqueous sodium periodate solution are mixed for about 1 to 2 minutes and allowed thereafter to undergo phase separation for about 10 to 20 minutes.

11. The method of claim 10 wherein the glycol is ethylene glycol and the supported chromogenic reagent consists of about 2 to 6 weight percent of 3-methyl-2-benzothiazolinone hydrazone hydrochloride monohydrate supported on a porous polyethylene film having a pore size within the range of from about 40 to 50 microns and wherein said supported chromogenic reagent is sealed in a plastic beaker.

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