SILVER HALIDE EMULSIONS WITH INCREASED SENSITIVITY

Wolfgang Müller-Bardorff, Cologne, and Wilhelm Saleck, Schildgen, Bergisch-Gladbach, Germany, assignors to Agfa Aktiengesellschaft, Leverkusen, Germany, a corporation of Germany

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5 Claims. (Cl. 96—107)

The invention relates to a method of increasing the sensitivity of photographic silver halide emulsions.

Numerous compounds are known which can be used as additives to the emulsion or developer to increase the sensitivity of a photographic layer. Many different terms are used for substances that have this effect, e.g., chemical sensitizers or activators. The best known class of substances in this field are polyethylene oxides, in particular polyethylene oxides and water-solubleonium compounds such as quaternary ammonium-, phosphonium- and sulphonium salts or combinations thereof or derivatives of thiourea, for example, thioauronium salts.

The maximum sensitivities obtained in silver halide emulsions by the addition of the above mentioned compounds presuppose that the silver halide emulsions have been activated by treatment with noble metal salts, especially gold salts.

The most commonly used combination for gold sensitization consists of gold-(III)-salts and thiocyanate or thiosulphate, although the solutions of thiocyanate are not very stable and those of thiosulphate are difficult to prepare. Reaction kinetic investigations, e.g., by Protass, Bjrrum and Kirschner (Z. Wiss. Angew. Phot. Kine 1, 55 (1956)) on gold complexes with thiocyanate ions admit of the assumption that sensitization is caused by a controlled, slow decomposition of the Au(1)-complexes in which the thiocyanate ions participate. The mechanism of this sensitization process is not certain since many reaction components are involved, such as metallic gold, Au(1)-, Au(III)- and thiocyanate ions, dithiocyanogen and additional sulphur degradation products including thiosulphate and gelatine.

Moreover, a considerable excess of thiocyanate ions is required to stabilise the gold(1)-complex and obtain maximum sensitivity of the silver halide emulsions. This method thus has numerous disadvantages. A considerable amount of the gold(III)-salts are bound by the gelatin, partly by absorption and partly by complex formation; another amount is reduced to metallic gold and lost for chemical sensitization. In addition the large excess of thiocyanate impairs the stability of the latent image after exposure.

It has now been found that a combined noble metal 55 ripening and reduction ripening of silver halide emulsions is obtained if the chemical ripening is performed with salts of noble metals of group VIII of the periodic system of the elements having an atomic weight greater than 100 and of gold, particularly platinum, palladium, iridium and more preferably gold-(III)-salts in the effective presence of o-hydroxybenzylamine compounds of the following general formula

\[
\text{R} \cdot \text{NH} \cdot \text{CH} = \text{CH} - \text{OH}
\]

In this formula, \( R \) represents a hydrogen atom or an alkyl group preferably lower alkyl up to 5 carbon atoms, which

may be substituted, for example, with hydroxyl, halogen such as Cl or Br, carboxyl, or an aryl group preferably phenyl which may be substituted, for example with alkyl, preferably lower alkoxy, halogen preferably Cl or Br, carboxyl, amine, hydroxyl, \( x \) represents a hydrogen atom, a hydroxyl group or a halogen atom preferably chlorine and bromine; and \( n \) is an integer between 1 and 5. The phenolic benzene ring in the above general formula can in addition be substituted, especially in the o-position or p-position to the hydroxy group, with lower alkyl radicals preferably those having up to 3 carbon atoms which may be substituted if desired, or with halogen atoms preferably chlorine and bromine, hydroxy or lower alkoxy groups preferably those having up to 3 carbon atoms.

The following compounds exhibit particular utility:

\[
\begin{align*}
\text{(I)} & : \quad \text{OH} \\
\text{(II)} & : \quad \text{OH} \\
\text{(III)} & : \quad \text{OH} \\
\text{(IV)} & : \quad \text{OH} \\
\text{(V)} & : \quad \text{OH} \\
\text{(VI)} & : \quad \text{OH} \\
\text{(VII)} & : \quad \text{OH} \\
\text{(VIII)} & : \quad \text{OH}
\end{align*}
\]
such compounds are already known. They can be easily reduced by reacting the appropriate substituted salicyl-
dehyde with the appropriate amine. The resulting Schiff's bases are hydrogenated in aqueous solution with
the aid of a Raney nickel catalyst. The substituent R can then be introduced with the aid of active chlorine com-
ounds in an aqueous alkaline medium. In the case of the bromobenzyl compounds, the bromine can be subse-
cutively introduced.

These o-hydroxybenzylaminocompounds are good com-
plexing agents for heavy and noble metal ions. For example, with gold(III)-chloride, gold(III)-complexes are
formed, which are often deep in color. Due to the weakly reducing effect of the compounds of the above general for-
mula, such Au(III) complexes are reduced to Au(I)-
complexes. To ensure the formation of the Au(I)-com-
plexes in every case, it is advisable to additionally add
weak reducing agents.

The process of the invention is capable of being used
with all photographic silver halide emulsions. The silver
halide can consist of silver chloride, silver bromide, silver
iodide or mixtures thereof.

An additional advantage of the process of the invention
is that the emulsions can be further sensitized with, for
example, sulphur compounds, polyalkylene oxides, water-
 soluble onium compounds or combinations of polyalkylene
oxides and onium compounds. In addition, they can be
optically sensitized by, e.g., cyanine or merocyanine
lystuffs and the like.

Basic stabilizers can be included in the emulsions, for
example, organic mercapto compounds, quaternary benzy-
chlozones, triazoles, tetrazaindolizines and the like. An-
other advantage of the process according to the invention
is that the sensitized emulsions can be used for black-
and-white photographic material or color photographic
material, it being possible in the latter case to incorporate
hydrophobic or hydrophilic color couplers into the emul-
sion layer.

As compared with a gold ripening using thiocyanate,
one of the compounds according to the invention pro-
duce the same sensitivity with half the quantity of gold,
usually with less fogging.

The mode of addition of the o-hydroxybenzylaminocom-
ounds of the invention to the emulsion is not critical.
The compounds may be added in dissolved or in solid
form, and before, during or after the addition of the noble
metal salt. Alternatively, the noble metal salt, for ex-
ample, gold(III)-chloride, may be first mixed with a
solution of the o-hydroxybenzylaminocompounds and then
added to the emulsions. Generally speaking, this addi-
tion is made before the chemical ripening of the emul-
sion. The chemical ripening is also called after-ripening.
The compounds are used in concentrations of 0.1 g. to
1 g. per kg. of emulsion (preferably 0.25-0.35 g./kg.).
The optimum quantity to be added depends on the re-
ducing power of the o-hydroxybenzylaminocompound and
the nature of the emulsion. It can easily be determined
by a few tests.

According to one preferred form of the present inven-
tion, the o-hydroxybenzylaminocompounds are used in
combination with weak reducing agents. Suitable for this

<p>| TABLE 1 |</p>
<table>
<thead>
<tr>
<th>Sensitivity increase</th>
<th>Fogging</th>
<th>Gamma</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample A</td>
<td>0</td>
<td>0.19</td>
</tr>
<tr>
<td>Sample B</td>
<td>+2 DIN</td>
<td>0.25</td>
</tr>
<tr>
<td>Sample C</td>
<td>+2 DIN</td>
<td>0.31</td>
</tr>
</tbody>
</table>

A highly sensitive gelatino-silver bromide emulsion
with a 4 mol percent of silver iodide, which contains 50
g. of silver per liter, has 35-40 mg./liter of KBr and sul-
phur compounds, etc. added thereto for the chemical
ripening in the usual way and is divided before the ripen-
ing into three samples A, B and C.

Sample A serves as standard sample. Before the ripen-
ing, the following solution is added to sample B, per
liter of emulsion:
1 ml of 0.08% gold(III)-chloride solution, mixed with
3 ml. of a 10% aqueous solution of Compound I. Before
the chemical ripening, there was added to sample C per
liter, 2 ml. of a 0.08% gold(III)-chloride solution, which
has been mixed with 3 ml. of a 10% aqueous solution of
Compound I.

After all three samples have been ripened until the
maximum sensitivity is reached, they have a wetting agent
and a stabilizer, e.g., of the azaindolizine type, added
thereto and are cast onto one of the conventional film
supports.

The three samples are exposed and developed in a nor-
mal commercial p-mercaptophenol hydroquinone de-
veloper for 10 minutes at 20°C.

The result is to be seen from Table 1.
For indicating the sensitivity, it is to be pointed out that an increase in the sensitivity of 3° corresponds to an increase by one aperture.

**Example 2**

A highly sensitive silver bromoiodide emulsion, as described in Example 1, is pretreated as in this example and divided into three samples A, B and C.

Sample A serves as standard sample.

Sample B has added thereto, per liter of emulsion, a mixture consisting of 3 ml of a 10% aqueous solution of the Compound III and 1 ml of a 0.08% gold-(III)-chloride solution.

Sample C contains, per liter of emulsion, 3 ml of a 10% aqueous solution of the Compound III, which has been mixed with 2 ml of a 0.08% gold-(III)-chloride solution.

The three samples are ripened as in Example 1, then coated as described therein onto a film support, and then the material is exposed and developed as described in Example 1.

The result is shown in Table 2.

<table>
<thead>
<tr>
<th></th>
<th>Sensitivity Increase</th>
<th>Fogging</th>
<th>Gamma</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample A</td>
<td>0°</td>
<td>0.19</td>
<td>0.55</td>
</tr>
<tr>
<td>Sample B</td>
<td>+2° DIN</td>
<td>0.12</td>
<td>0.45</td>
</tr>
<tr>
<td>Sample C</td>
<td>+3° DIN</td>
<td>0.19</td>
<td>0.55</td>
</tr>
</tbody>
</table>

**Example 3**

Three samples A, B and C of a silver halide emulsion as described in Example 1, are treated as follows:

Sample A serves as standard sample.

Sample B has added thereto, per liter of emulsion, a mixture of 3 ml of a 10% aqueous solution of Compound II and 1 ml of a 0.08% gold-(III)-chloride solution.

Sample C contains, per liter of emulsion, 3 ml of a 10% aqueous solution of Compound II, which has been mixed with 2 ml of a 0.08% gold-(III)-chloride solution.

The ripening of the three samples, the casting onto a film support, the exposure and the development is performed as indicated in Example 1.

The result is shown in Table 3.

<table>
<thead>
<tr>
<th></th>
<th>Sensitivity Increase</th>
<th>Fogging</th>
<th>Gamma</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample A</td>
<td>0°</td>
<td>0.19</td>
<td>0.55</td>
</tr>
<tr>
<td>Sample B</td>
<td>+2° DIN</td>
<td>0.20</td>
<td>0.50</td>
</tr>
<tr>
<td>Sample C</td>
<td>+3° DIN</td>
<td>0.13</td>
<td>0.55</td>
</tr>
</tbody>
</table>

**Example 4**

Three samples A, B and C of a silver halide emulsion, as described in Example 1, are treated as follows:

Sample A without additives, serves as standard sample.

Sample B has added thereto, per liter of emulsion, a mixture of 3 ml of a 10% aqueous solution of the Compound VIII, and 1 ml of a 0.08% gold-(III)-chloride solution.

Sample C contains, per liter of emulsion, 3 ml of a 10% aqueous solution of the Compound VIII, which has been mixed with 2 ml of a 0.08% gold-(III)-chloride solution.

The ripening of the three samples, the casting onto a support and the exposure and development is performed as described in Example 1.

The result is shown in Table 4.

<table>
<thead>
<tr>
<th></th>
<th>Sensitivity Increase</th>
<th>Fogging</th>
<th>Gamma</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample A</td>
<td>0°</td>
<td>0.19</td>
<td>0.55</td>
</tr>
<tr>
<td>Sample B</td>
<td>+2° DIN</td>
<td>0.20</td>
<td>0.50</td>
</tr>
<tr>
<td>Sample C</td>
<td>+3° DIN</td>
<td>0.13</td>
<td>0.55</td>
</tr>
</tbody>
</table>

**Example 5**

A highly sensitive gelatin-silver bromoiodide emulsion with 4 mol percent of silver iodide, which contains 50 g. of silver per liter, has 35-40 mg./liter of KBr and sulphur compounds etc. for the chemical ripening, is divided before ripening into 3 samples A, B and C.

Sample A serves as comparison sample. Before the ripening of sample B, the latter has added thereto the following solution, per liter of emulsion:

6 ml of a 10% solution of the Compound I, to which are added 2 ml of a 0.08% gold-(III)-chloride solution. Thereafter, 1 ml of a 10% solution of sodium hydroxyacetate is added.

Before the chemical ripening of sample C, the latter has added thereto the following mixture per liter of emulsion:

4 ml of a 0.08% gold-(III)-chloride solution are added to 6 ml of 10% solution of the compound I, the pH of the mixture is adjusted to 8 and finally 1 ml of 10% solution of sodium hydroxyacetate is added.

After ripening until the maximum sensitivity is reached, a wetting agent and a stabilizer, e.g., of the azaindolizine type are added to each of the three samples which are then cast onto a support such as paper, a cellulose acetate, a polyster preferably of polyethylene terephthalate or a polycarbonate in particular of bis-hydroxyphenyl alkane.

The three samples are exposed and developed in a commercial p-methylaminophenol hydroquinone developer for 10 minutes at 20° C.

The result is shown in Table 5.

<table>
<thead>
<tr>
<th></th>
<th>Sensitivity Increase</th>
<th>Fogging</th>
<th>Gamma</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample A</td>
<td>0°</td>
<td>0.20</td>
<td>0.55</td>
</tr>
<tr>
<td>Sample B</td>
<td>+2° DIN</td>
<td>0.27</td>
<td>0.50</td>
</tr>
<tr>
<td>Sample C</td>
<td>+3° DIN</td>
<td>0.15</td>
<td>0.50</td>
</tr>
</tbody>
</table>

**Example 6**

Three samples A, B and C of a silver halide emulsion, as described in Example 5, are treated as follows:

Sample A serves as standard sample. Before being chemically ripened, Sample B has the following solution added thereto, per liter of emulsion:

2 ml of a 0.08% gold-(III)-chloride solution are added to 6 ml of a 10% solution of compound X, the pH of the mixture is adjusted to 8 and finally 1 ml of a 10% solution of sodium hydroxyacetate is added.

Sample C has added thereto, per liter of emulsion, a mixture which is prepared as follows:

4 ml of a 0.08% gold-(III)-chloride solution are added to 6 ml of a 10% solution of compound X, the pH of the mixture is adjusted to 8 and finally 1 ml of a 10% solution of sodium hydroxyacetate is added.

The ripening of the three samples, the casting onto a support, the exposure and development is performed as indicated in Example 5.
Example 7

Three samples A, B and C of a silver halide emulsion, as described in Example 5, are treated as follows:

Sample A serves as standard sample. Before being chemically ripened, Sample B has the following solution added thereto, per liter of emulsion:

2 ml of a 0.08% gold-(III)-chloride solution are added to 6 ml of a 10% solution of compound III, the pH of the mixture is adjusted to 8 and then 1 ml of a 10% solution of sodium d-glucos-a-heptanoate is added.

Sample C has added thereto, per liter of emulsion, a mixture which has been prepared as follows:

4 ml of a 0.08% gold-(III)-chloride solution are added to 6 ml of a 10% solution of compound III, the pH of the mixture is adjusted to 8 and finally 1 ml of a 10% solution of sodium d-glucos-a-heptanoate is added.

The ripening of the three samples, the casting onto a support, the exposure and development is performed as indicated in Example 5.

The result is shown in Table 7.

<table>
<thead>
<tr>
<th>Sensitivity Increase</th>
<th>Foggng</th>
<th>Gamma</th>
</tr>
</thead>
<tbody>
<tr>
<td>sample A</td>
<td>0</td>
<td>0.20</td>
</tr>
<tr>
<td>sample B</td>
<td>+3° D.I.</td>
<td>0.13</td>
</tr>
<tr>
<td>sample C</td>
<td>+2° D.I.</td>
<td>0.10</td>
</tr>
</tbody>
</table>

Example 8

An X-ray emulsion with 2 mol percent of silver iodide, which contains 80 g of silver per liter, and which further contains bromide and sulphur compounds for the chemical ripening, is divided before ripening into two samples A and B.

Sample A is a standard sample. Before the after-ripening of Sample B the following solution is added per liter of emulsion:

3 ml of a 10% solution of the compound I are mixed with 1.4 ml of a 0.08% gold-(III)-chloride solution. The pH of the solution is adjusted to 8 and 0.15 ml of a 10% solution of sodium hydroxyacetate is added.

After these samples have been after-ripened until the maximum sensitivity is obtained, a wetting agent and a stabilizer, e.g., of the azaindolizine type, is added thereto. Thereafter the emulsions are coated onto a usual support.

The two samples are exposed and developed for 5 minutes at 20°C in an X-ray developer which yields silver images with a steep characteristic curve.

The result is shown in Table 8.

<table>
<thead>
<tr>
<th>Sensitivity Increase</th>
<th>Foggng</th>
<th>Gamma</th>
</tr>
</thead>
<tbody>
<tr>
<td>sample A</td>
<td>0</td>
<td>0.20</td>
</tr>
<tr>
<td>sample B</td>
<td>+3° D.I.</td>
<td>0.13</td>
</tr>
</tbody>
</table>

Example 9

The procedure is as indicated in Example 8, but the following solution is used with sample B: 3 ml of a 10% solution of the compound III are mixed with 1.4 ml of a 0.08% gold-(III)-chloride solution and the pH is adjusted to 8. There is then added 0.15 ml of a 10% solution of sodium hydroxyacetate. The result is shown in Table 9:

<table>
<thead>
<tr>
<th>Sensitivity Increase</th>
<th>Foggng</th>
<th>Gamma</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample A</td>
<td>0</td>
<td>0.17</td>
</tr>
<tr>
<td>Sample B</td>
<td>+3° D.I.</td>
<td>0.10</td>
</tr>
</tbody>
</table>

Example 10

A gelatino-silver bromobromodioxydine emulsion of medium sensitivity with 0.1% percent of silver iodide, which contains 45 g of silver per liter and which further contains potassium bromide and sulphur compounds, etc., for the chemical ripening is divided before after-ripening into two samples A and B.

Sample A is the standard sample.

Sample B, before the after-ripening, the following solution is added thereto per liter of emulsion:

3 ml of a 25% solution of compound I are mixed with 1 ml of a 0.08% gold-(III)-chloride solution. The pH of the solution is adjusted to 8 and 0.5 ml of a 25% solution of sodium hydroxyacetate is added.

After having ripened these samples until the maximum sensitivity is obtained, a wetting agent and a stabilizer, e.g., of the azaindolizine type, are added and they are then cast onto a usual support.

The two samples are exposed and developed for 10 minutes at 20°C in a p-methylaminophenol hydroquinone developer.

The result is shown in Table 10.

<table>
<thead>
<tr>
<th>Sensitivity Increase</th>
<th>Foggng</th>
<th>Gamma</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample A</td>
<td>0</td>
<td>0.17</td>
</tr>
<tr>
<td>Sample B</td>
<td>+3° D.I.</td>
<td>0.10</td>
</tr>
</tbody>
</table>

Example 11

A gelatino-silver chlorobromobromidine emulsion with 20 mol percent of silver chloride and 6 mol percent of silver iodide which contains 50 g of silver per liter and which further contains potassium bromide and sulphur compounds etc., for the chemical ripening is divided before after-ripening into two samples A and B.

Sample A is the comparison sample.

Sample B, before the after-ripening, the following solution is added thereto, per liter of emulsion:

7 ml of a 10% solution of compound I are mixed with 3 ml of a 0.08% gold-(III)-chloride solution. The pH of the solution is adjusted to 8 and 0.3 ml of a 10% solution of sodium hydroxyacetate is added.

After this mixture has been added to the emulsion, the samples are after-ripened up to maximum sensitivity, thereafter a wetting agent and a stabilizer, e.g., of the azaindolizine type, are added and the samples are cast onto a paper support.

The two samples are exposed and developed for 2 minutes in a developer of the following composition:

<table>
<thead>
<tr>
<th>Grams</th>
</tr>
</thead>
<tbody>
<tr>
<td>p-Methylaminophenol</td>
</tr>
<tr>
<td>Hydroquinone</td>
</tr>
<tr>
<td>Anhydrous sodium sulphite</td>
</tr>
<tr>
<td>Anhydrous sodium carbonate</td>
</tr>
<tr>
<td>Potassium bromide</td>
</tr>
<tr>
<td>Water to make 1000 ml</td>
</tr>
</tbody>
</table>
The result is shown in Table 11.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Sensitivity Increase</th>
<th>Fogging</th>
<th>Gamma</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.13</td>
<td>1.8</td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>+2.5° DIN</td>
<td>0.11</td>
<td>1.6</td>
</tr>
</tbody>
</table>

It will be clear to those skilled in this art that the practice of the invention lends itself readily to a number of useful modifications in method, material, etc. For example, the noble metal salts are not limited to the previously mentioned, because it is possible to use any suitable salt which is soluble in water or lower alcohols. Such compounds are: gold halides, such as auric chloride, or complex gold halides, such as potassium chloraurate (K₂AuCl₄) and sodium chloraurate (NaAuCl₄). However, gold compounds, such as auric sulfates, are practically as useful as the gold halides. Aurous, as well as auric compounds can be used. Complex gold salts, such as alkali metal aurous thiosulfates, alkali metal aurous sulfites (e.g., sodium or potassium aurous thiosulfate and sodium or potassium aurous sulfite). Potassium chloraurite, potassium bromoaurite, potassium iodoaurite, or the corresponding sodium, calcium, strontium, cadmium or gallium salts can also be used. Pyridinotrichloro-gold, ethylenediamine-bis-trichloro gold, diethyl-monobromo-gold and diethyl gold acetone and gold complexes with sulfur compounds such as are commonly present in gelatin, e.g., the gold thiophenamine complexes, can also be used.

Suitable salts of noble metals of the VIII group are ammonium or potassium, chloropalladate, ammonium, sodium and potassium chloroplatinate, ammonium potassium and sodium bromoplatinate, ammonium chloroplatinate, ammonium chloroplatinate, ammonium chloride, ammonium, potassium and sodium chloropalladate, ammonium, potassium and sodium chloropalladate, etc.

The after-ripening of the emulsion with the noble metal salt and the o-hydroxybenzylamine compounds is performed at an appropriate temperature particularly between 30° and 60° C. During after-ripening the pH of the emulsion is advantageously adjusted to the acid side of neutrality preferably between 5 and 7. Maintenance of the emulsion on the acid side of neutrality during coating of the emulsion is also preferred.

The noble metal salts are employed in an amount below that which produces a substantial fog. In practicizing the present invention, a quantity of the noble metal salt is employed, equivalent to between 0.1 and 50 mg of the noble metal per mole of silver halide in the emulsion.

The noble metal compounds are preferably incorporated in the emulsion in the form of their solutions in a suitable solvent such as water, methyl alcohol, ethyl alcohol or the like.

We claim:

1. In a process for producing a sensitized photographic silver halide emulsion, the step which comprises after-ripening the emulsion on the acid side of neutrality in the effective presence of at least one water-soluble salt of a Group VIII metal having an atomic weight of greater than 100 or gold and in the effective presence of an o-hydroxybenzylamine compound of the formula

\[
\text{CH}_3-\text{NR}-(\text{CH}_2)_n-x
\]

wherein R represents a substituent of the group consisting of hydrogen, alkyl, and a phenyl, x stands for a member of the group consisting of hydrogen, hydroxy and halogen and n an integer between 1 and 5.

2. A process as defined in claim 1 wherein the phenyl ring of the benzyl group of the o-hydroxy-benzylamine compounds is substituted with a radical of the group consisting of hydroxy, lower alkyl, halogen and lower alkoxy.

3. A process as defined in claim 1 wherein the emulsion is after-ripened in the further active presence of a reducing agent of the group consisting of polyhydroxy-cyclic, hydroxylic and nitrogenous bases.

4. A process as defined in claim 1, where the metal is gold.

5. A process as defined in claim 1, wherein the emulsion which is to be after-ripened additionally contains at least one member of the group consisting of sulfur compounds, oxime compounds, polyalkylene oxides and a tetrazaindolizine stabilizer.

No references cited.

NORMAN G. TORCHIN, Primary Examiner.

J. H. RAUBITSCHEK, Assistant Examiner.