

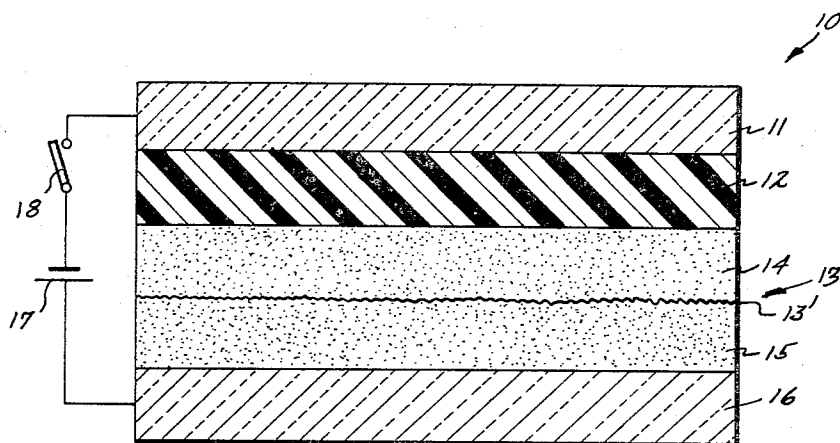
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DATA STORAGE DEVICE AND METHOD

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DATA STORAGE DEVICE AND METHOD
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In general, the present invention relates to a method and device for storing data by making permanent reproductions thereof. More particularly, the present invention involves an efficient, easily reversible process for the reproduction of a light image on a copysheet requiring a low electrical potential. As used in the present application, the term "light" refers not only to the visible portion of the radiant energy spectrum but also to all forms of radiation such as electron radiation, X-rays, ultra-violet radiation and so forth.

There are at present a wide variety of methods and devices for storing data in the form of light images. Examples of such prior art methods are the conventional silver halide film, the Xerox electrostatic photocopying system, the Ozalid photocopying system and the National Cash Register photochemical dye system. However such prior art systems have a number of limitations. Usually, once the copysheet utilized in the system is developed, it cannot be erased for reuse or can only be totally erased. Frequently, such prior art systems require chemical processing and are sensitive only to a narrow portion of the radiant energy spectrum. Also, usually the copysheet used to receive the light image presents storage problems because of sensitivity to stray radiation.

For these reasons as well as other reasons, there has been in recent years extensive research and development in the field of electrolytic development of light images on copysheets. An example of such process is disclosed by Hamm et al. in United States Patent No. 3,085,051. As discussed in the Hamm patent, the process involves exposing an electrically conductive base having a coating of photoconductive material to a light image. The image is electrolytically developed by contacting the photoconducting material with an electrolytically reducible compound and then passing current through the light exposed portions of the photoconductive material. As specifically disclosed by the Hamm patent, the electrolytically reducible may be an indium compound which is reduced to free indium upon electrolysis. Such process is typical of the presently known electrolytic development processes which utilize the decomposition of a compound at the electrode to develop the image. The copysheet developed by such processes can be erased by chemical processes such as treating with acid as described by Hamm, but the process itself is not practically reversible. Furthermore, the usual prior art electrolytic development process normally has a relatively low coulometric efficiency, e.g., about 50% and normally requires relatively high electrical potentials to develop the image, e.g., generally above about 10 volts.

Consequently, an object of the present invention is a method and device or copysheet for the easily reversible electrolytic development of light images.

Another object of the present invention is a method or copysheet for the electrolytic development of light images with substantially 100% coulometric efficiency.

Still another object of the present invention is a method or copysheet for the electrolytic development of light images which can operate with electrical potentials as low as 0.2 volt.

Other objects and advantages of the present invention will be readily apparent from the following description and drawing which illustrate a preferred exemplary embodiment of the present invention.

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In general, the present invention involves a copysheet comprising an electrolytically conductive layer consisting essentially of a first film having a dye fixed therein whose color changes at a set electrical potential and having at least one associated ion movable therein adapted to change concentration by the passage of current through the first film. The concentration change of the associated ion causes an electrical potential change in the first film which includes the set dye electrical potential. A second film contiguous to the first film has at least one counterpart ion movable therein adapted to change concentration by the passage of current through the second film opposite to the associated ion concentration change. The associated ions and counterpart ions are prevented from mixing and are related by a reversible electrochemical reaction. By exposing a photoconductive layer contiguous to the electrolytically conductive layer to a light image and passing current selectively through the light exposed portions of the photoconductive layer and the corresponding adjoining portions of the electrolytically conductive layer the color of the dye is changed in such portion to make a permanent reproduction of the light image.

In order to facilitate understanding of the present invention, reference will now be made to the appended drawing of a preferred specific embodiment of the present invention. Such drawing should not be construed as limiting the present invention which is properly set forth in the appended claims.

In the drawing:

FIG. 1 is a cross-sectional schematic view of a device embodying the present invention.

As illustrated in FIG. 1, the present invention involves a device 10 comprising a first electrically conducting layer 11 in the form of a transparent electrode. Such transparent electrode is preferably composed of commercial tin oxide glass because of its high resistance to chemical reactions. Contiguous to the first electrically conducting layer 11 is a photoconductive layer 12 formed out of a suitable material such as cadmium sulfide or zinc oxide. Adjoining the photoconductive layer 12 is an electrolytically conducting layer 13 consisting essentially of a first film 14 and a contiguous second film 15. The first film 14 is in the form of a gel such as hydroxy ethyl cellulose having a dye dissolved and fixed therein. Such dye changes color at a set electrical potential or set hydrogen activity. The first film 14 also has at least one associated ion dissolved and movable therein which is adapted to change concentration by the passage of current through the first film 14. Such concentration change of the associated ion is adapted to cause an electrical potential or hydrogen activity change in the first film 14 which includes the set dye electrical potential or hydrogen activity so that the dye color is thus changed.

The second film 15 contiguous to the first film 14 is also in the form of a gel such as hydroxy ethyl cellulose and has at least one counterpart ion dissolved and movable therein. Such counterpart ion is adapted to change concentration opposite to the associated ion concentration change by passage of current through the second film 15. The associated and counterpart ions prevented from mixing by barrier means 13 consisting of the interface between films 14 and 15 which permits the passage of current-carrying ions therethrough. Also the associated and counterpart ions are related by a reversible electrochemical reaction. Adjoining the electrolytically conducting layer 13 is a second electrically conducting layer 16 also in the form of a transparent electrode preferably composed of tin oxide glass. Completing the electrical circuit between the first electrically conductive layer 11 and second electrically conductive layer 16 is a source of electrical potential such as battery 17 and a switch 18 so that an electrical potential may be imposed as desired.

Many dyes are known which change color at a set electrical potential including the electrical potential related to a set hydrogen activity. Hence, as used in the present application, the term "set electrical potential" includes the special case of the electrical potential determined by the hydrogen activity as well as the ratio of activities such as the ferric-ferrous activity ratio. Examples of suitable dyes changing color at a set electrical potential are shown in Table 1 wherein the electrical potential for color change

TABLE 1

Indicator	P ₀ at pH=0	Color Change	
		Oxidized	Reduced
Safranine T.....	0.24	Red.....	Colorless.
Neutral red.....	0.24	Red.....	Do.
Indigo monosulfonate.....	0.26	Blue.....	Do.
Phenosafranine.....	0.28	Red.....	Do.
Indigo tetrasulfonate.....	0.36	Blue.....	Do.
Nile blue.....	0.41	do.....	Do.
Methylene blue.....	0.53	Green-blue.....	Do.
1-naphthol-2-sulfonic acid indophenol.....	0.54	Red.....	Do.
2,6-dibromophenyl indophenol.....	0.67	Blue.....	Do.
Diphenylamine (diphenylbenzidine).....	0.76	Violet.....	Do.
Diphenylamine sulfonic acid.....	0.85	Red-violet.....	Do.
Erioglaucin A.....	1.0	Red.....	Green.
Setoglucin O.....	1.06	Pale red.....	Yellow-green.
p-Nitrodiphenylamine.....	1.06	Violet.....	Colorless.
o,m'-Diphenylamino dicarboxylic acid.....	1.12	Blue-violet.....	Do.
o,o'-Diphenylamino dicarboxylic acid.....	1.26	do.....	Do.
o-Phenanthroline ferrous complex.....	1.14	Pale blue.....	Red.
Nitro-o-phenanthroline ferrous complex.....	1.25	do.....	Violet-red.

TABLE 2

pH Range	Indicator	pH Range	Indicator
0.0-2.0.....	Malachite Green Hydrochloride, yellow-bluish green.	4.8-6.4.....	Chlorophenol Red, yellow-red.
0.0-3.0.....	Eosin Y, yellow-green.	5.0-6.0.....	Hematoxylin, yellow-red.
0.0-3.6.....	Erythrosin B, orange-red.	5.2-6.8.....	Bromocresol Purple, yellow-purple.
0.1-3.2.....	Methyl Violet 2B, yellow-violet.	5.2-7.0.....	Bromophenol Red, yellow-red.
0.8-2.6.....	Crystal violet, green-blue violet.	5.7-6.8.....	Hematein, yellow-pink.
1.2-2.1.....	Diphenylamino-benzene, red-yellow.	6.0-7.6.....	Bromothymol Blue, yellow-blue.
1.2-2.8.....	m-Cresol Purple, red-yellow.	6.0-8.4.....	Naphthylamine Brown, orange-pink.
1.2-2.8.....	Thymol Blue, red-yellow.	6.8-8.0.....	Neutral Red, red-yellow orange.
1.2-2.8.....	Xylenol Blue, red-yellow.	6.8-8.4.....	Phenol Red, red-yellow.
1.2-3.0.....	Basic Fuchsin, purple-red.	6.9-8.0.....	Rosolic Acid, yellow-red.
1.3-4.0.....	Benzopurpurin 4B, blue violet-red.	7.2-8.8.....	Cresol Red, yellow-red.
1.4-3.2.....	Quinaldine Red, colorless-red.	7.4-8.6.....	Tumeric, yellow-brown.
2.0-3.0.....	Cresol Red, orange-yellow.	7.4-9.0.....	m-Cresol Purple, red-yellow.
2.4-4.0.....	Methyl Yellow, red-yellow.	8.0-9.6.....	Thymol Blue, yellow-blue.
3.0-4.6.....	Bromophenol Blue, yellow-purple.	8.0-9.6.....	Xylenol Blue, yellow-blue.
3.0-4.6.....	Chlorophenol Blue, yellow-blue.	8.3-10.....	Phenolphthalein, colorless-red.
3.0-4.6.....	Tetrabromophenol Blue, yellow-blue.	9.0-13.....	Nile Blue A, blue-pink.
3.0-6.0.....	Phenacolin, yellow-red.	9.4-14.....	Alkali Blue 6B, light blue-rose.
3.2-4.8.....	Bromochlorophenol Blue, yellow-purple.	10.0-11.....	Aniline Blue, blue-lavender.
3.4-4.9.....	Propyl Orange, red orange-yellow.	10.0-13.....	Phenacetolin, red-colorless.
3.7-5.0.....	Benzene-azo-naphthylamine, red-yellow.	10.1-12.1.....	Alizarin Yellow G, yellow-orange.
3.8-5.4.....	Bromocresol Green, yellow-blue.	11.1-12.7.....	Tropacolin O, yellow-orange.
3.8-6.6.....	Gallein, light brown-rose.	11.5-14.0.....	Orange G, yellow-pink.
4.0-7.0.....	Chrysoidin Y, orange-yellow.	11.6-14.0.....	Basic Fuchsin, red-colorless.
4.4-6.2.....	Methyl Red, red-yellow.	12.0-13.0.....	Aniline Blue, pink-orange red.
4.5-8.3.....	Azolitmin, red-blue.	12.0-14.0.....	Acid Fuchsin, red-colorless.
4.7-6.2.....	Cochineal, red-violet.	12.1-14.0.....	Clayton Yellow, yellow-red.

is given with reference to the normal hydrogen electrode potential for the dye in a solution one normal in hydrogen ions. The dyes listed in Table 1 are oxidation-reduction indicators with the standard electrical potential given when the dye is half oxidized and half reduced.

Examples of suitable dyes changing color at a set hydrogen activity are shown in Table 2 wherein the pH range for color change is given.

The associated ion of the present invention includes the one or more ion members of the oxidized or reduced portion of a reversible electrochemical oxidation-reduction system. Similarly the counterpart ion of the present invention includes the one or more ion members of the other portion of a reversible electrochemical oxidation-reduction system. Thus, if the associated ion is the reduced portion of the system, then the counterpart ion is the oxidized portion. The concentration of the associated ion in such system can be changed by changing its oxidation state. Thus, frequently the concentration of the associated ion is changed by its oxidation. Also, frequently the counterpart ion is the associated ion in a different oxidation state. In such system, when the associated ion is changed by its oxidation, the counterpart ion is the associated ion in a higher oxidation state. Examples of suitable reversible electrochemical oxidation-reduction systems are set forth in Table 3. With reference to Table 3, it should be noted that the reactions utilizing a basic solution do not directly involve the hydrogen ion. However, since in aqueous solution, the hydroxyl ion and hydrogen ion concentration are inversely proportional, the set hydrogen activity for a given dye can be produced by change in the hydroxyl ion concentration.

TABLE 3.—TYPICAL REVERSIBLE ELECTROCHEMICAL REACTIONS

Acid media:	Half cell potential, volts
(1) $\text{Cr}^{+2} \rightarrow \text{Cr}^{+3} + e$	0.41
(2) $\text{Ti}^{+2} \rightarrow \text{Ti}^{+3} + e$	0.37
(3) $\text{V}^{+2} \rightarrow \text{V}^{+3} + e$	0.255
(4) $\text{Ti}^{+3} + \text{H}_2\text{O} \rightarrow \text{TiO}^{+2} + 2\text{H}^{+} + e$	0.1
(5) $\text{Sn}^{+2} \rightarrow \text{Sn}^{+4} + 2e$	-0.15
(6) $\text{Cu}^{+} \rightarrow \text{Cu}^{+2} + e$	-0.153
(7) $\text{U}^{+4} + 2\text{H}_2\text{O} \rightarrow \text{UO}_2^{+2} + 4\text{H}^{+} + 2e$	-0.062
(8) $\text{V}^{+3} + \text{H}_2\text{O} \rightarrow \text{VO}^{+2} + 2\text{H}^{+} + e$	-0.361
(9) $\text{MnO}_4^{-2} \rightarrow \text{MnO}_4^{-1} + e$	-0.564
(10) $\text{Fe}^{+2} \rightarrow \text{Fe}^{+3} + e$	-0.771
(11) $\text{Hg}_2^{+2} \rightarrow 2\text{Hg}^{+2} + 2e$	-0.92
(12) $\text{ClO}_3^{-} + \text{H}_2\text{O} \rightarrow \text{ClO}_4^{-} + 2\text{H}^{+} + 2e$	-1.19
Basic solution:	
(1) $\text{HPO}_4^{-2} + 3\text{OH}^{-} \rightarrow \text{PO}_4^{-3} + 2\text{H}_2\text{O} + 2e$	1.12
(2) $\text{S}_2\text{O}_4^{-2} + 4\text{OH}^{-} \rightarrow 2\text{SO}_3^{-2} + 2\text{H}_2\text{O} + 2e$	1.12
(3) $\text{CN}^{-} + 2\text{OH}^{-} \rightarrow \text{CNO}^{-} + \text{H}_2\text{O} + 2e$	0.97
(4) $\text{SO}_3^{-2} + 2\text{OH}^{-} \rightarrow \text{SO}_4^{-2} + \text{H}_2\text{O} + 2e$	0.93
(5) $\text{HSnO}_2^{-} + \text{H}_2\text{O} + 3\text{OH}^{-} \rightarrow \text{Sn(OH)}_6^{-2} + 2e$	0.90
(6) $\text{SeO}_3^{-2} + 2\text{OH}^{-} \rightarrow \text{SeO}_4^{-2} + \text{H}_2\text{O} + 2e$	-0.05
(7) $\text{I}^{-} + 6\text{OH}^{-} \rightarrow \text{IO}_3^{-} + 3\text{H}_2\text{O} + 6e$	-0.26
(8) $\text{ClO}_2^{-} + 2\text{OH}^{-} \rightarrow \text{ClO}_3^{-} + \text{H}_2\text{O} + 2e$	-0.33
(9) $\text{ClO}_3^{-} + 2\text{OH}^{-} \rightarrow \text{ClO}_4^{-} + \text{H}_2\text{O} + 2e$	-0.36
(10) $\text{Br}^{-} + 6\text{OH}^{-} \rightarrow \text{BrO}_3^{-} + 3\text{H}_2\text{O} + 6e$	-0.61
(11) $\text{ClO}^{-} + 2\text{OH}^{-} \rightarrow \text{ClO}_2^{-} + \text{H}_2\text{O} + 2e$	-0.66
(12) $\text{IO}_3^{-} + 3\text{OH}^{-} \rightarrow \text{H}_3\text{IO}_6^{-2} + 2e$	-0.7
(13) $\text{Br}^{-} + 2\text{OH}^{-} \rightarrow \text{BrO}^{-} + \text{H}_2\text{O} + 2e$	-0.76
(14) $3\text{OH}^{-} \rightarrow \text{HO}_2^{-} + \text{H}_2\text{O} + 2e$	-0.88
(15) $\text{Cl}^{-} + 2\text{OH}^{-} \rightarrow \text{ClO}^{-} + \text{H}_2\text{O} + 2e$	-0.89

The basic principle of the present invention may best be described with reference to a particular simple embodiment of the present invention. In the first film 14 of the device 10, a small amount (for example 1×10^{-5} to 1×10^{-2} molar concentration; preferably, about 1×10^{-3} molar) of a dye, diphenylbenzidine is dispersed in its reduced colorless state along with a suitable concentration of a ferrous salt. For example, ferrous sulphate can be used at a concentration of about 1×10^{-3} to 1 molar but preferably a concentration of about 0.1 molar is used. In the second film 15, a similar, preferably equal, concen-

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tration of a ferric salt such as ferric sulphate is dissolved. Current is then passed through the device 10 by closing the switch 18. Under such conditions when the photoconductive layer 12 is conductive due to light impinging thereon, the ferrous ions are oxidized to ferric ions in the first film 14 while the ferric ions in the second film 15 are reduced to ferrous ions. Initially, the electrical potential in the first film 14 is about 0.55 volt which is well below the set color change electrical potential of diphenylbenzidine of about 0.76 volt. Thus, the dye remains in its reduced colorless state when added to the ferrous ion system since the dye assumes the same electrical potential as the solution in which it is dissolved. As the ferrous ions are oxidized, the electrical potential of the first film 14 increases. Thus, approximately when the concentrations (activity is usually substantially the same as concentration in dilute solutions) of the ferric and ferrous ions are equal, the standard potential of about 0.771 volt is attained. Also about at this point, diphenylbenzidine changes color since its set color change electrical potential has been exceeded. As current continues to pass through the device 10, the ferrous ions in the first film 14 are finally completely oxidized to ferric ions at which point an electrical potential of about 1.0 volt is attained in the first film 14. Also at this point, current flow stops since there are no more ferrous ions in the first film 14 to be oxidized or ferric ions in the second film 15 to be reduced. If it is then desired to reverse the color change so that the dye returns to its colorless state, the direction of the current flow may merely be reversed, the photoconductive layer 12 exposed to a light source, and the reverse reactions occur. Similar simple systems by which comparable results are attained include the dyes thionin or eriochrome used with copper, nickel, chromium and tin ions.

With the foregoing discussion in mind, the operation of the present invention as illustrated in FIG. 1 can be easily described. The photoconductive layer 12 is exposed to the light image to be reproduced while sufficient electrical potential is set up between the electrodes 11 and 16 to pass current therebetween when the resistance of the photoconductive layer 12 is reduced. Current is then passed selectively through the light exposed portions of the photoconductive layer 12 and the corresponding adjoining portions of the electrolytically conducting layer 13. As noted above such current flow changes the color of the dye but such color change occurs only at the point of current passage due to the physical immobility of the dye in the film. Thus the light image is reproduced by the dye in the first film 14. Also, merely by shutting off the electrical potential difference, the image is fixed and not influenced by further exposure.

Many other specific embodiments of the present invention will be obvious to one skilled in the art in view of this disclosure. Thus, the copysheet of the present invention may include only the electrolytically conducting layer with the device for developing the copysheet including the other elements of FIG. 1. Alternatively, the copysheet may also include the photoconductive layer and/or one or both of the electrically conducting layers. Also, the electrode spaced from the photoconducting layer by the electrolytically conducting layer need not be transparent. In addition the barrier means may consist of an ion selective membrane between the films of the copysheet.

There are many features in the present invention which clearly show the significant advance the present invention represents over the prior art. Consequently only a few of the more outstanding features will be pointed out to illustrate the unexpected and unusual results attained by the present invention. One feature of the present invention is the easy reversibility of the method and thus the easy erasability of the copysheet. Consequently the copysheets are reusable without resorting to chemical processing. Still another feature of the present invention is its high efficiency since close to 100% coulometric efficiency can be obtained. Still another feature of the present invention

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is the low electrical potential difference which may be used. Thus electrical potential differences as low as 0.2 volt can be used.

It will be understood that the foregoing description and examples are only illustrative of the present invention and it is not intended that the invention be limited thereto. All substitutions, alterations and modifications of the present invention which come within the scope of the following claims or to which the present invention is readily susceptible without departing from the spirit and scope of this disclosure are considered part of the present invention.

I claim:

1. An efficient, easily reversible process for the reproduction of a light image requiring a low electrical potential comprising:

(a) exposing to said light image a photoconductive layer on a contiguous electrolytically conductive layer, said electrolytically conductive layer consisting essentially of:

(I) a first film having a dye fixed therein whose color changes at a set electrical potential and having at least one associated ion movable therein adapted to change concentration by the passage of current through said first film with said concentration change causing an electrical potential change in said first film which includes said set dye electrical potential, and

(II) a contiguous second film having at least one counterpart ion movable therein adapted to change concentration by the passage of current through said second film, said counterpart ion concentration change being opposite to said associated ion concentration change; and

(b) preventing said associated ions from mixing with said counterpart ions, said ions being related by a reversible electrochemical reaction; and passing current selectively through the light exposed portions of said photoconductive layer and the adjoining portions of said electrolytically conductive layer to change the color of said dye in said portions.

2. A process as stated in claim 1 wherein said associated ion concentration is changed by changing its oxidation state.

3. A process as stated in claim 2 wherein said associated ion concentration is changed by its oxidation.

4. A process as stated in claim 1 wherein said counterpart ion is said associated ion in a different oxidation state.

5. A process as stated in claim 4 wherein said counterpart ion is said associated ion in a higher oxidation state.

6. An efficient easily reversible process for the reproduction of a light image requiring a low electrical potential comprising:

(a) exposing to said light image a photoconductive layer on a contiguous electrolytically conductive layer, said electrolytically conductive layer consisting essentially of:

(I) a first film having a dye fixed therein whose color changes at a set hydrogen activity and having at least one associated ion movable therein adapted to change concentration by the passage of current through said first film with said concentration change causing a hydrogen activity change in said first film which includes said set dye hydrogen activity, and

(II) a contiguous second film having at least one counterpart ion movable therein adapted to change concentration opposite to said associated ion concentration change by passage of current through said second film, and

(b) preventing said associated ions from mixing with said counterpart ions, said ions being related by a reversible electrochemical reaction; and

(c) passing current selectively through the light exposed

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portions of said photoconductive layer and the adjoining portions of said electrolytically conductive layer to change the color of said dye in said portions.

7. A copysheet adapted to be electrolytically developed reversibly and efficiently with a low electrical potential comprising an electrolytically conducting layer consisting essentially of:

- (a) a first film having a dye fixed therein whose color changes at a set electrical potential and having at least one associated ion movable therein adapted to change concentration by the passage of current through said first film with said concentration change causing an electrical potential change in said first film which includes said set dye electrical potential;
- (b) a contiguous second film having at least one counterpart ion movable therein adapted to change concentration opposite to said associated ion concentration change by the passage of current through said second film, said associated and counterpart ions being related by a reversible electrochemical reaction; and
- (c) barrier means between said films adapted to prevent the passage of said associated ions and counterpart ions therethrough but permit the passage of current-carrying ions therethrough.

8. A copysheet as stated in claim 7 wherein said electrolytically conducting layer has a contiguous photoconductive layer.

9. A copysheet as stated in claim 7 wherein said electrolytically conducting layer has a contiguous photoconductive layer and said photoconductive layer has a contiguous electrically conductive layer.

10. A copysheet adapted to be electrolytically developed reversibly and efficiently with a low electrical potential comprising an electrolytically conducting layer consisting essentially of:

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(a) a first film having a dye fixed therein whose color changes at a set hydrogen activity and having at least one associated ion movable therein adapted to change concentration by the passage of current through said first film with said concentration change causing a hydrogen activity change in said first film which includes said set dye hydrogen activity;

(b) a second film having at least one counterpart ion movable therein adapted to change concentration opposite to said associated ion concentration change by the passage of current through said second film, said associated and counterpart ions being related by a reversible electrochemical reaction; and

(c) barrier means between said films adapted to prevent the passage of said associated ions and counterpart ions therethrough but permit the passage of current-carrying ions therethrough.

11. A copysheet as stated in claim 10 wherein said electrolytically conducting layer has a contiguous photoconductive layer.

12. A copysheet as stated in claim 10 wherein said electrolytically conducting layer has a contiguous photoconductive layer and said photoconductive layer has a contiguous electrically conductive layer.

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