

Analog Image Processing in Modern Color Photography

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In the context of this article, analog image processing comprises methods to chemically control and improve image quality in color negative films during photographic processing. The chemical control applies to the generation of image dyes and to the optical interaction of partial dye images. The image dyes are generated in two consecutive steps: silver reaction and coupling reaction. Yield- and distribution-parameters of these reaction steps are relevant for image-quality aspects. In this review the various chemical methods to achieve image processing (e. g. inhibition, (dye-) amplification, dye spread) are discussed.

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Introduction

Image processing is usually seen as a unique feature of digital photography, but in fact it is state of the art in modern color negative films. It is just that the term “image processing” is rarely used in the context of chemical photography. Some striking similarities between digital, chemical and visual image processing have been described.¹

The imaging chain as shown in Fig. 1 has an analog branch and a digital branch, both of which include a processing step. In the analog branch this is the step transforming the latent image into the developed dye image. In this processing step the latent image is not only made visible but at the same time a lot of chemical reactions occur with the sole purpose of improving image quality. Therefore the term “processing” has a two-fold meaning here: it is image processing by means of chemical processing. The chromogenic processing of a color negative film basically consists of two consecutive reactions:

- Silver reaction: the latent image is transformed by reaction with the developer into a silver image and a coupled distribution of oxidized developer.
- Coupling reaction: the oxidized developer in turn reacts with the coupler to the final dye image.

The variety of intended image processing reactions and some unwanted side reactions make the actual reaction scheme of the development process much more complex, as depicted in Fig. 2.

For a given latent image all image-properties (contrast, sharpness, granularity, color-rendition) are completely determined by the printer-relevant dye densities. Any mechanism to influence these dye densities is a po-

tential avenue for chemical image processing. The dye densities can be controlled either

- by reactive interference with the formation of the dyes, the controlling parameters being the yield and distribution in the silver reaction and in the coupling reaction;
- or by optical interaction of different, partial dye images.

Silver Reaction

Controlling the silver yield plays a dominant role in chemical image processing, whereas the silver distribution is completely predetermined by the distribution of the latent image under standard processing conditions. Silver yield is defined here as the amount (mass) of developed silver per developable grain.

Control of Silver Yield

The yield can either be decreased (inhibition) or increased (amplification).

In particular the image-wise inhibition of the silver development in a sort of feedback-loop is the basis for the improvement² of various aspects of image quality (left part of Fig. 3):

- inhibition promotes partial development of the individual grains; this improves granularity;
- diffusion of an inhibitor across an edge creates an edge effect thereby enhancing sharpness;
- diffusion of an inhibitor to adjacent layers creates interimage-effects compensating for unwanted side-absorptions of sensitizers and/or of image dyes; this enhances color accuracy and color brilliance.

Amplification, in contrast, deteriorates these image properties; it is only the speed of a film which gains from amplification (right part of Fig. 3).

Inhibition. An image-wise retardation of silver development can be achieved by inhibitors released either from the developing silver halide crystals (Br^- , I^-) or

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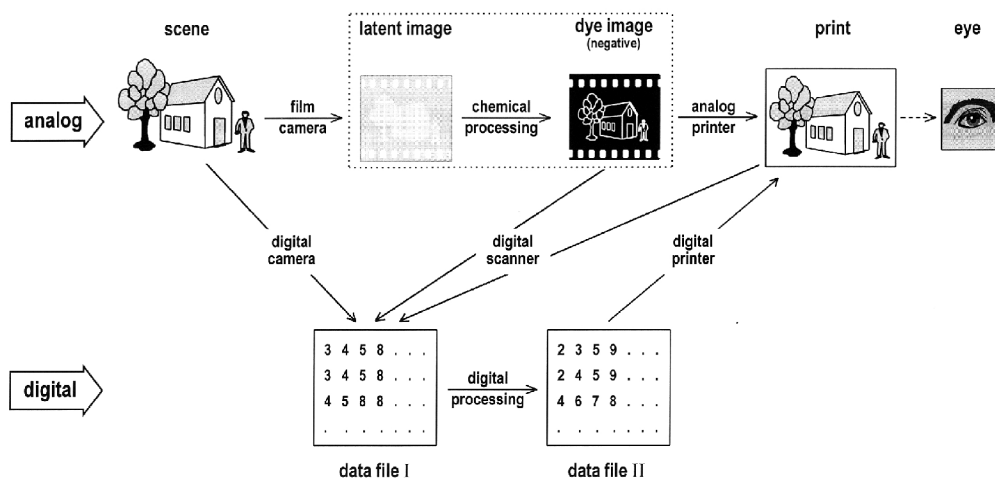
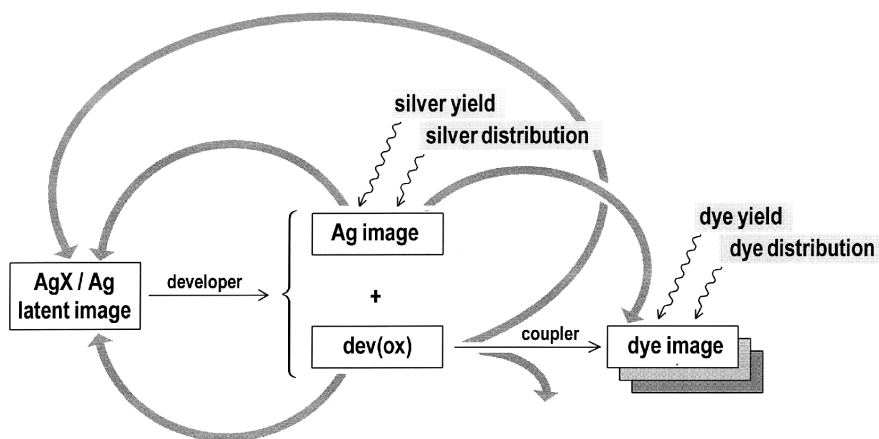


Figure 1. Imaging chain with analog and digital branch.



Chemical (analog) image processing:
 imagewise control of dye densities during processing by
 - reactive interference with formation of dyes
 - optical interaction between partial dye images

Figure 2. Chromogenic processing (black lines) consisting of silver reaction and coupling reaction; shaded lines and boxes indicate image processing interactions.

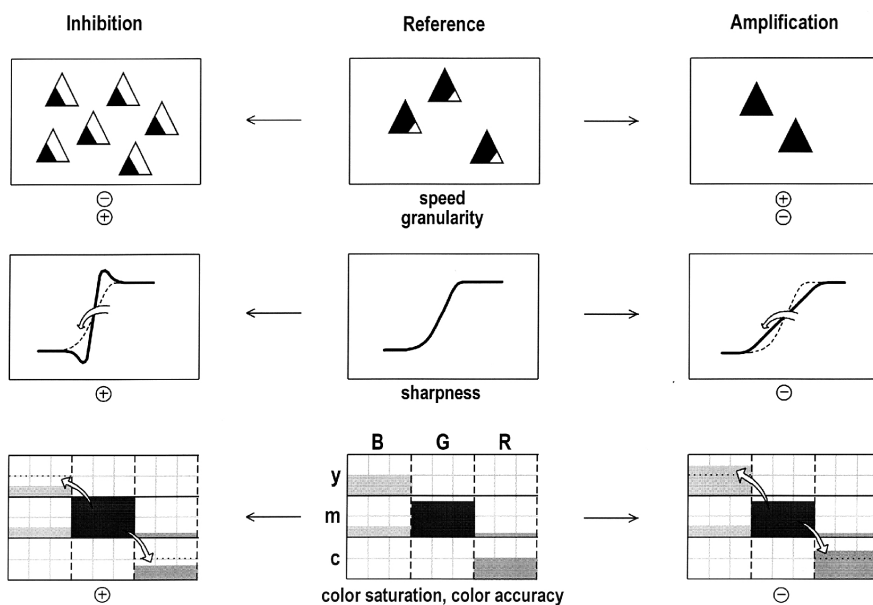


Figure 3. Effect of inhibition and amplification of silver development on various image properties; y, m, c denote different partial layers; B, G, R denote spectral absorption bands of image dyes.

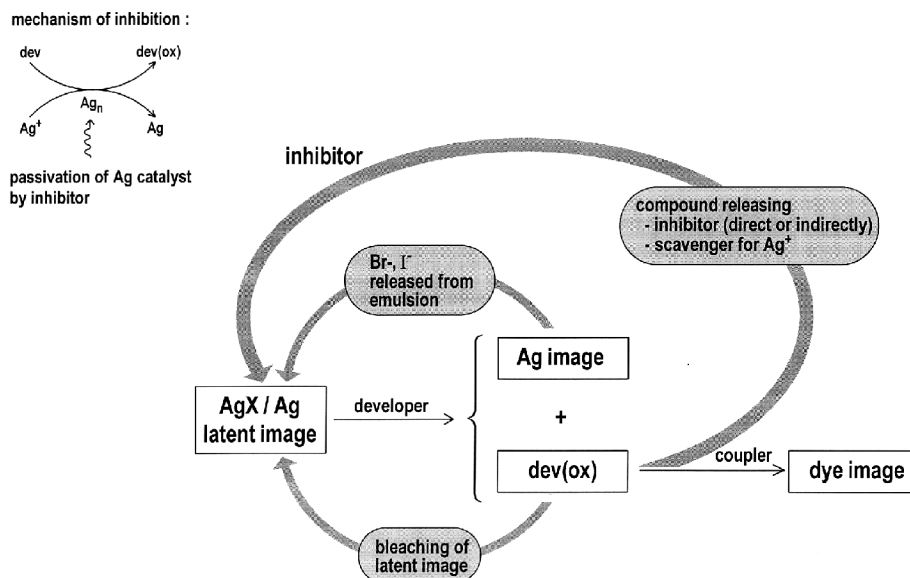


Figure 4. Methods to achieve an inhibition of the silver development; explanation of y, m, c and B, G, R see Fig. 3.

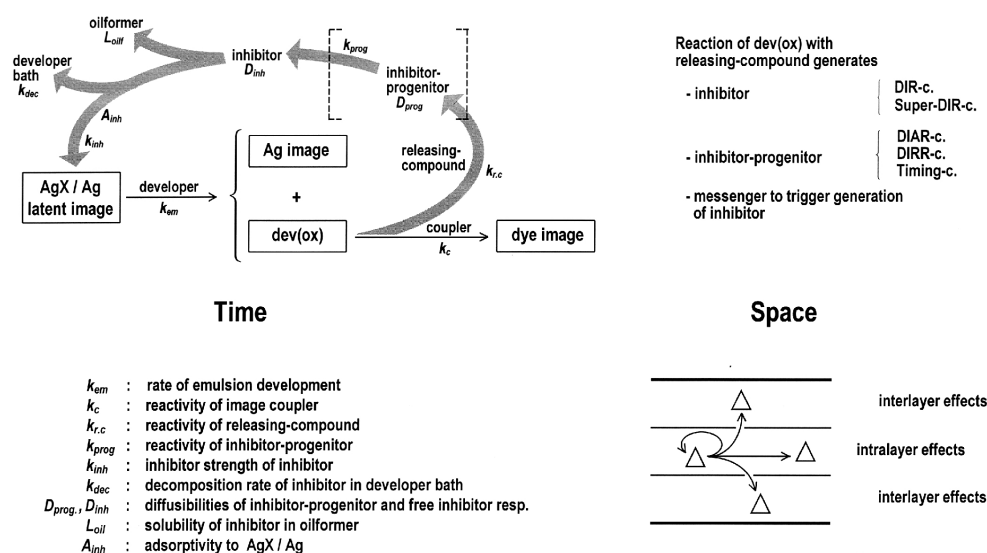


Figure 5. Kinetic aspects regarding the application of inhibitor releasing compounds.

from special releaser-compounds by reaction with the oxidized developer (see Fig. 4). The action of a typical inhibitor is to passivate the silver speck on a developing grain poisoning its catalytic activity.³ Therefore, even small amounts of an inhibitor can be very effective in retarding further development. Two more mechanisms of retardation are indicated just for completeness:

- the release of a scavenger for Ag⁺: this mechanism cannot be very effective as it does not act on the catalyst⁴;
- bleaching of latent image by oxidized developer: this is an unwanted side-reaction which degrades the signal/noise-ratio.

Optimization of inhibition effects requires the inhibitor to interfere at a certain time (during the development process), at a certain point, and with a certain power. This implies a fine tuning of the various kinetic and diffusion processes involved, as summarized in Fig. 5. In classical Development-Inhibitor-Releasing (DIR)-cou-

plers the inhibitor is released directly. In order to achieve high diffusibility even with strongly adsorbing inhibitors or to delay the release of the free inhibitor, releasing-compounds with an indirect release of the inhibitor have been devised. In these cases the primary reaction of the oxidized developer with the releaser-compound produces an inhibitor-progenitor which in turn releases the inhibitor (DIAR-c: Development-Inhibitor-Anchimeric-Releasing-coupler; DIRR-c: Development-Inhibitor-Releaser-Releasing-coupler).^{5,6} The reaction sequence can be even further extended by the insertion of more consecutive reaction-steps (1 or 2) into the reaction chain leading to the free inhibitor.⁷⁻¹⁰ A different approach is the image-wise release of a messenger compound which initiates the generation of an inhibitor in a distant layer of the film.¹¹

As there are different requirements for the realization of short-range effects (improvement of granularity) and of long-range effects (interimage effects, edge effects) the

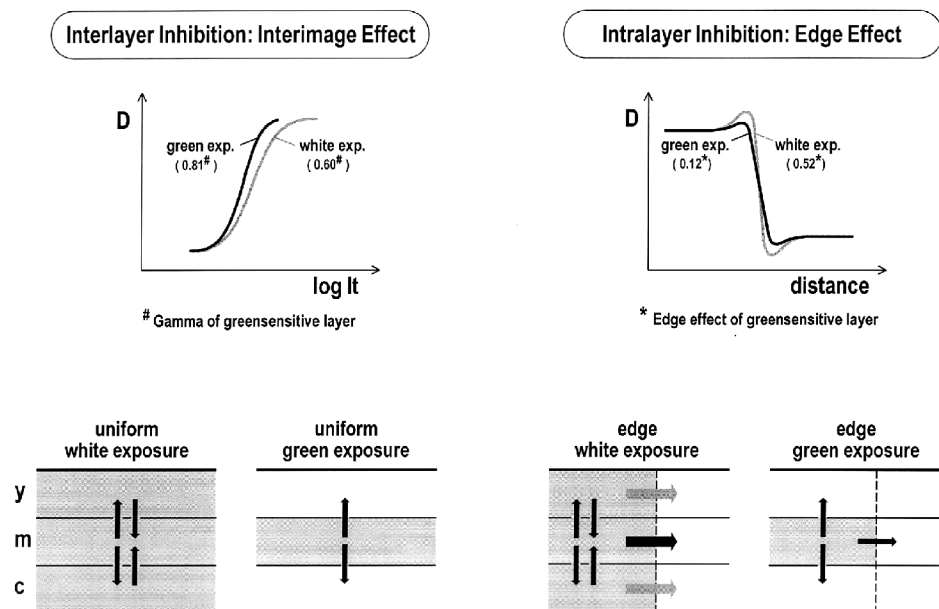


Figure 6. Interimage and edge effects generated by inhibition; the edge effect is defined as the density increase resulting from an exposure through a narrow slit (30 μm width) compared to a homogeneous exposure (experimental film; measurements at density $D = 0.8$; y, m, c indicate different partial layers;).

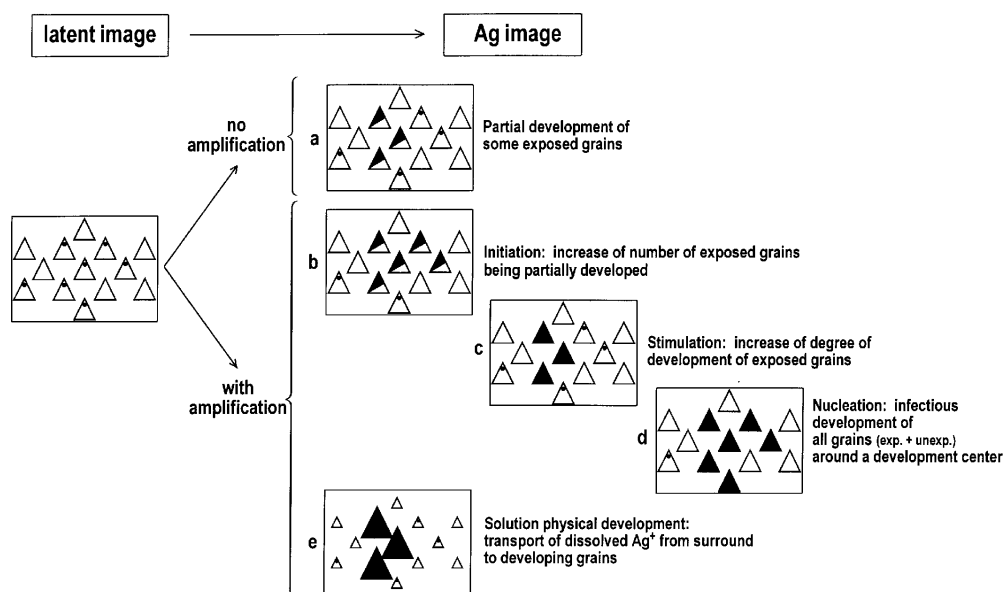


Figure 7. Mechanisms of amplification of silver development.

overall optimization of a color negative film obviously needs a full set of different releaser-compounds.^{5,12}

The impact of inhibitor diffusion is manifest in interimage and edge effects (Fig. 6).

Interimage Effects. In an experimental color negative film the gamma of the green sensitive layer to white exposure was 0.60, jumping to 0.81 on green exposure. In the whitelight exposed film there is no net diffusion of inhibitor out of the green sensitive layer, in contrast to the situation under green exposure.

Edge Effects. Due to the higher net concentration of inhibitor in the green sensitive layer of the whitelight ex-

posed film, diffusion of inhibitor across the edge is more pronounced in this case (creating an edge effect of 0.52) than in the green-exposed film (edge effect 0.12).

Amplification. An increase of the silver yield can be realized according to one of the following mechanisms (Fig. 7):

- initiation: increase in number of exposed grains being (partially) developed;
- stimulation: increase in degree of development of exposed grains;
- nucleation: infectious development of all grains (exposed and unexposed) around a development center;
- solution physical development: transport of Ag^+ to developing grains through the solution phase.

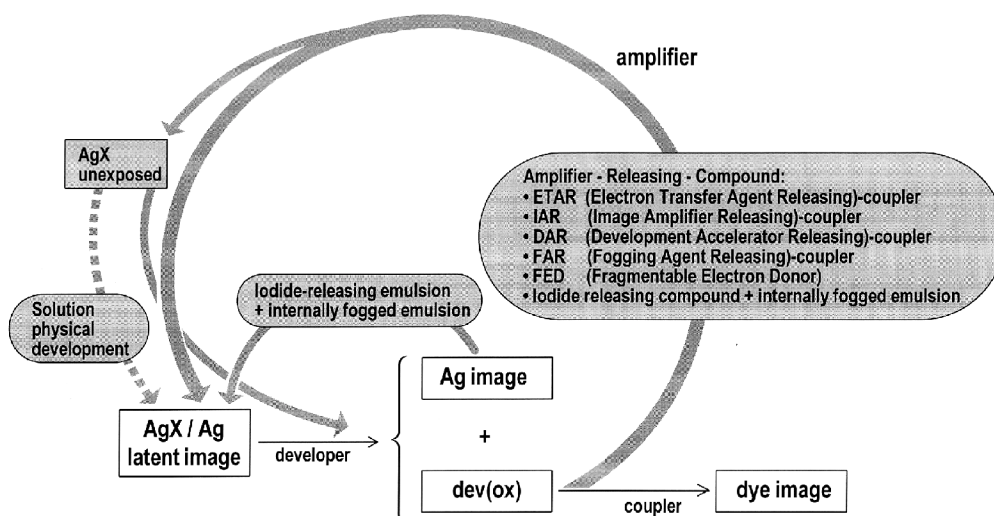


Figure 8. Methods to achieve amplification of silver development.

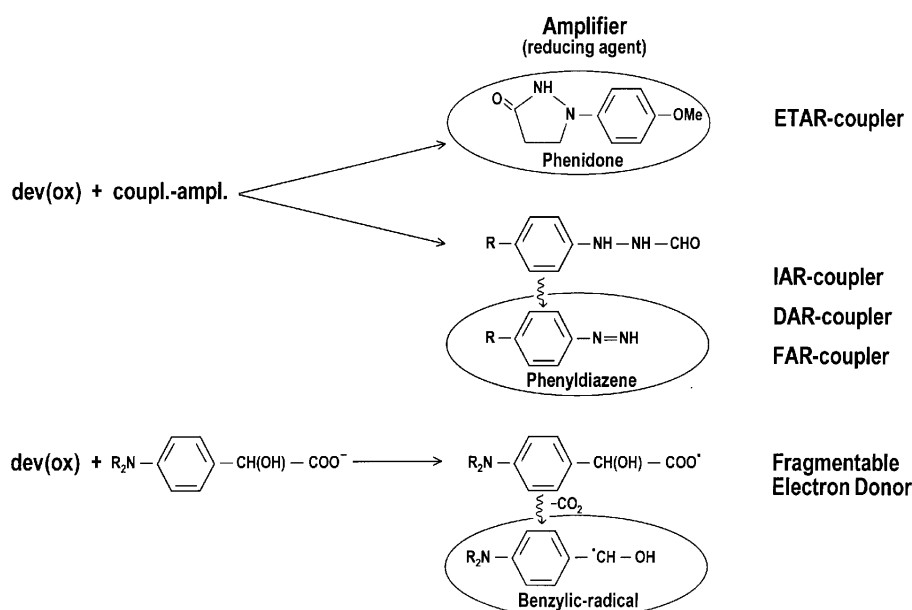


Figure 9. Chemical reactions to generate amplifying agents.

These mechanisms are explained in more detail in Fig. 8.

In solution physical development the developed silver acts as catalyst for the reaction of dissolved Ag^+ with developer. The amplification is limited only by the supply of the reaction partners (and not by the crystal size). Solution physical development always contributes to the total development to an extent which depends on the presence of silver halide solvents (complexing compounds) in the developer, and on the solubility of the silver halide crystals (iodide content; size).

Other methods for amplification or acceleration (the terms are used synonymously here) correspond to the methods for inhibition. Namely, the amplifying agent is either released from the developing emulsion crystal or from a special releaser-compound.

In the first case the amplifying agent can be iodide in combination with an internally fogged emulsion having

a thin surface-shield of AgCl/AgBr . Released iodide breaks up the shield of the fogged emulsion grains making the hidden silver specks accessible to the action of the developer.¹³ In this way a form of infectious development is achieved.

In the second case the amplifying agent is generated in a reaction of the oxidized developer with a releaser-compound which can be:

- iodide-releasers in combination with an internally fogged emulsion; the amplification mechanism is the same as if the iodide is released from the emulsion,¹³
- fragmentable electron donors: these compounds fragment after oxidation by oxidized developer to generate a reducing (fogging) agent according to the scheme of Fig. 9. (If the fragmentable electron donor is adsorbed to the surface of a silver halide crystal it may even be oxidized by a positive hole generated on exposure. The resulting reducing agent can inject an

extra electron into the conduction band of the exposed crystal enhancing the sensitivity; this is a mode of chemical sensitization¹⁴).

- amplifying agent releasing couplers (ETAR-, IAR-, DAR-, FAR-couplers); the amplifying agent has reducing properties.

ETAR-couplers release typical black and white-developers, eg., a Phenidone-type-developer.¹⁵ The terms IAR-, DAR- and FAR-couplers do not designate chemically different classes, but different strategies their inventors had in mind:

- image-amplifiers are to counteract latent-image-bleaching,¹⁶
- development accelerators are to enhance the development process,¹⁷
- fogging agents, by definition, are to make adjacent grains developable irrespective of exposure.^{17,18}

For some typical DAR-couplers it has been demonstrated that the released acylhydrazines do not act discriminatively but instead make both exposed and unexposed grains developable.¹⁹⁻²¹ We therefore assume that each of these types of amplifying agent simultaneously reacts along all three pathways (initiation, stimulation, nucleation), depending on the local redox potential during development and the reducing power of the agent.

Control of Silver Distribution

The primary spatial distribution of the developed silver is completely determined by the latent image distribution. But theoretically there are means to influence the silver distribution to a certain extent by transformation into a secondary distribution, eg.,

- silver redistribution: a first black and white developed silver image is rehalogenated and then (after fogging) color developed. The rehalogenation step can alter the primary distribution.
- application of silver-amplifying agents: amplification by use of, eg., FAR-couplers, not only influences silver yield but also silver distribution.

Coupling Reaction

The coupling process transforms the silver image via the oxidized developer into the final dye image. The controlling parameters are dye yield (dye density per mass of silver) and dye distribution (dye spread; characterized by the diameter of the dye cloud).

The effects of dye yield and dye spread on sharpness and granularity are described by Eqs. 1 through 3.²²⁻²⁶

$$MTF_{Ag/dye} = f(1/\phi) \quad (1)$$

$$MTF_{dye} = MTF_{Ag(opt.)} \cdot MTF_{Ag(chem.)} \cdot MTF_{Ag/dye} \quad (2)$$

$$\sigma_{D(dye)} = \sigma_{m(Ag)} \cdot \frac{dD_{(dye)}}{dm_{(Ag)}} \cdot \sqrt{\int MTF_{Ag/dye}^2(n) \cdot B(n) \cdot dn} \quad (3)$$

MTF:	modulation transfer function;
ϕ :	diameter of dye cloud
$MTF_{Ag(opt.)}$:	MTF for exposure effects (eg., scattering) on silver image
$MTF_{Ag(chem.)}$:	MTF for development effects (eg., edge effects) on silver image
$MTF_{Ag/dye}$:	MTF for transformation of silver image to dye image
MTF_{dye} :	MTF of final dye image
$\sigma_{m(Ag)}$:	granularity of silver image (based on silver mass \underline{m})

$\sigma_{D(dye)}$:	granularity of dye image (based on dye density \underline{D})
$\frac{dD(dye)}{dm(Ag)}$:	differential dye yield;
v :	spatial frequency
$B(v)$:	factor describing the influence of the geometry of the scanning aperture on the measurement of granularity

These equations are derived for a monolayer film with no nonlinear processes intervening in the transformation of the silver image into the dye image. The equations are taken as an approximative description for a real color negative film with interlayer effects and nonlinear effects (eg., the $MTF_{Ag/chem}$ is known to depend on exposure and therefore is not independent of $MTF_{Ag/opt.}$). We assume that the impact of dye spread and of dye yield is correctly described at least in a qualitative way.

Equations 1 through 3 imply, that sharpness is decreased by extended dye spreading (increase of ϕ) and is not dependent on dye yield, whereas granularity gains from dye spreading and low dye yield.

Control of Dye Yield

The oxidized developer can react along different, competitive channels (Fig. 10):

- reaction with image coupler via a leuco-dye to form dye
- reaction with competing compounds (competing with the image coupler)
- decomposition (both the oxidized developer and the leuco-compound decompose to a certain extent²⁷).

Accordingly the dye yield can be influenced by reactivity and equivalency of the coupler, by its concentration and location, eg., in a layer adjacent to the emulsion, and also by the addition of competing compounds (special purpose couplers or scavengers which can be added or generated in a reaction of oxidized developer with a scavenger releasing coupler²⁸).

Typical strategies for the reduction of granularity in multilayered color negative films are low coupler concentrations in high speed sublayers and the addition of scavengers; scavengers for oxidized developer are also built into interlayers separating layers of different sensitization to prevent crosstalk between different color channels.

Application of silver saving 2-equivalent-couplers (or even 1-equivalent dye formation²⁹) increases dye yield but incurs a penalty in granularity.

An increase in dye yield is realized in silver catalyzed dye amplification processes.^{29,30} In these processes an external oxidizing agent, eg., H_2O_2 as depicted in Fig. 11, is added to the developer; the silver system only generates the catalyst for the redox reaction. The increase of dye yield is connected with a deterioration of granularity. Therefore such a dye amplification process is not applicable for film processing, but for paper processing only. Recently the advances in dye amplification chemistry with respect to stability of processing solutions and design of an appropriate color paper have been discussed.^{31,32}

Control of Dye Distribution

Dye spread will decrease granularity and at the same time sharpness. Therefore dye spread is an ambiguous tool and has to be carefully optimized with regard to perceived image quality.

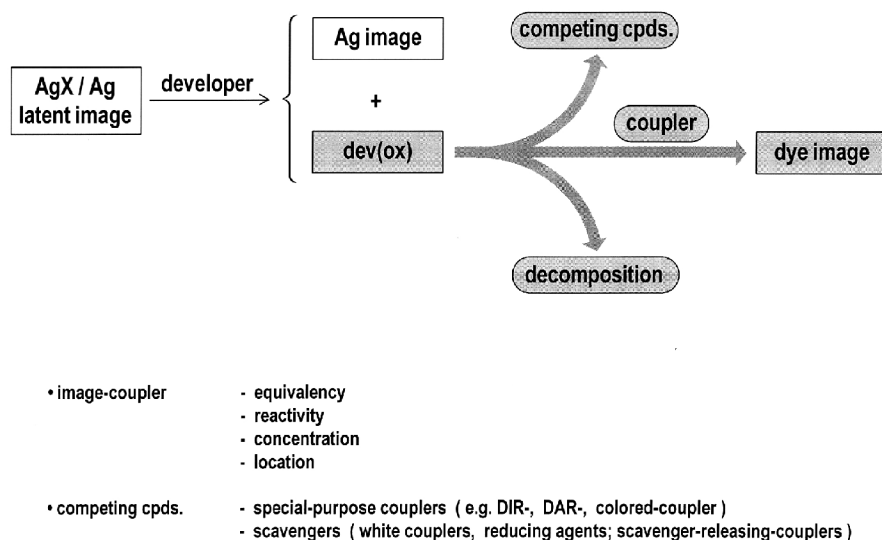


Figure 10. Reaction channels for oxidized developer.

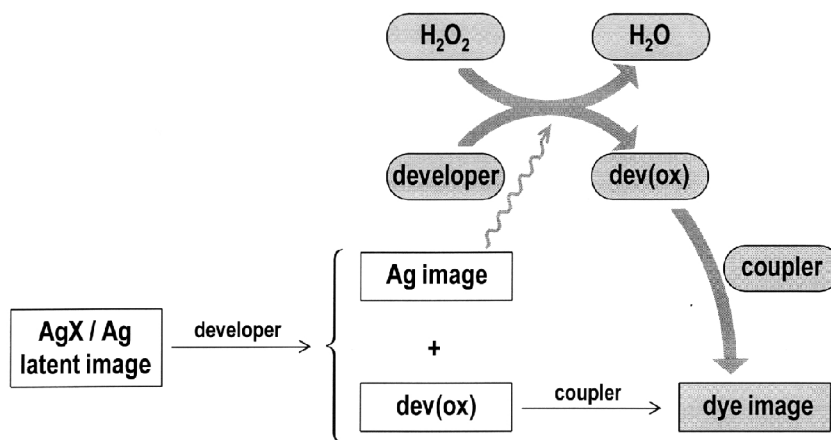
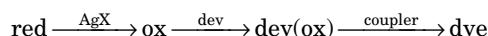


Figure 11. Silver-catalyzed dye amplification.

A general description of the coupling process includes an intermediate, which can be the leuco-dye, but may also stand for other structures. All three image carriers - dev(ox), intermediate(s), dye - are potential candidates to achieve a wider dye-distribution (see Fig. 12).

Spreading of Oxidized Developer

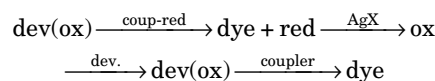
- Low concentration of coupler or a long distance to the coupler^{33,34} not only decreases dye yield (see Fig. 12) but also enhances diffusion of the oxidized developer.
- Admixture of certain polymers, e. g., polyurethane in relative high concentrations,^{35,36} it is assumed that at high concentrations (35 - 50% of total binder) the polymer forms a coherent phase within the gelatin matrix facilitating diffusion of the oxidized developer.³⁷
- Addition of a reducing agent (red; e. g., a black and white developer) to the color developer (dev). The reducing agent is oxidized by exposed silver halide and crossoxidizes the color developer.^{24,38}



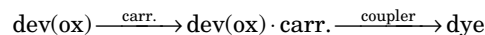
(image-carrying, diffusing species are typed in bold letters).

Spreading of a Coupling-Intermediate

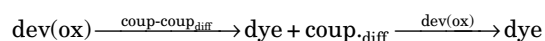
- Incorporation of reducing agent releasing couplers (ETAR-, DAR-, IAR-, FAR-couplers) not only affects silver yield but also dye spreading as both intermediates (red, ox) contribute to image diffusion.^{39,40}



- Dev(ox)-carrier: the idea is to let the oxidized developer react with a carrier compound (carr.) to form a diffusible product which eventually reacts with the coupler.⁴¹



- Diffusible coupler releasing coupler: in a first step a diffusible coupler (coup_{diff}) is released (image-wise) which subsequently reacts to form an immobile dye.⁴²



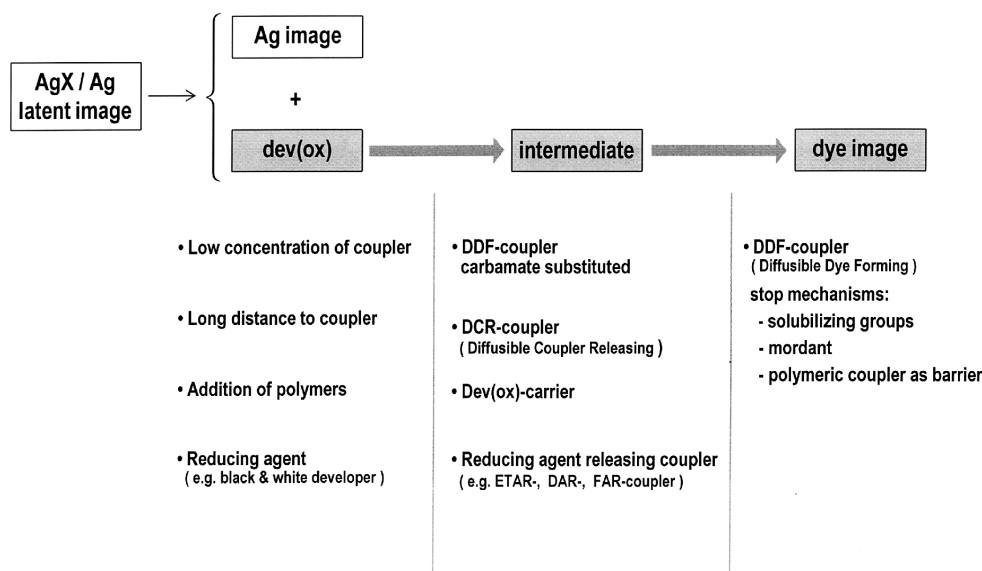
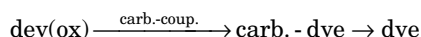


Figure 12. Control of dye distribution.

As the diffusible coupler needs further reaction with oxidized developer for dye formation, it seems, however, that this mechanism does not expand the image diffusion beyond the diffusion of the oxidized developer.

- Diffusible dye forming coupler with carbamate substituent: the dye generated in the coupling reaction is diffusible but only for a brief time period as the carbamate substituent (carb.) enabling diffusion decomposes under alkaline processing conditions.⁴³



Spreading of Final Dye. Diffusion of image dyes can easily be achieved by applying couplers with relative small ballast groups (historically the problem with chromogenic coupling was to *avoid* diffusion of couplers and dyes !), but the result is uncontrollable dye diffusion particularly under humid conditions.⁴⁴ A few mechanisms have been proposed for a stop mechanism: solubilizing groups (limiting dye diffusion to the processing time spent in the developer bath),^{45,46} a mordant for the diffusing dye, and additional polymeric couplers (dyes) as a barrier for diffusion.⁴⁷

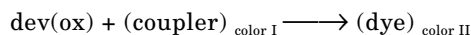
Optical Interaction of Partial Dye Images

The routines of chemical image processing discussed so far relate to modulation of the chemical formation of image dyes.

A different approach to chemical image processing is the optical interaction of different, partial dye images.

The combination of a negative image with a corresponding positive partial image constitutes optical inhibition as shown in Fig. 13.

Colored couplers⁴⁸ undergo a color shift in the coupling reaction.



In this example a yellow colored coupler is incorporated in the green sensitive layer together with the magenta coupler. In the coupling reaction the image coupler forms the magenta dye which has a pronounced yellow side absorption. This unwanted increase of yellow density is compensated for by a corresponding decrease of yellow density in the reaction of the colored coupler. The resulting uniform yellow density can be filtered off during printing. The overall effect is the elimination of the unwanted yellow side absorption of the magenta image dye. This optical interaction between magenta and yellow partial images (optical interimage effect) is equivalent to chemical interaction based on inhibitive messenger compounds (chemical interimage effects; as shown in Figs. 3 and 6).

Unsharp Colored Couplers. Not only interimage effects but also edge effects can be generated optically. In this case the negative image is to be superimposed by an unsharp positive image. This principle, generally known as unsharp masking, led to the proposal to incorporate the unsharp mask into the film.⁴⁹ Fig. 13 illustrates unsharp masking of a magenta partial image. The positive image is realized by application of a magenta colored coupler which couples from magenta to colorless; the unsharpness is realized by appropriate diffusibility of the colored coupler. The density profile of the combination of negative image and positive mask exhibits an edge effect. The concept of unsharp colored couplers to create edge effects is of theoretical interest only, it has not been applied in commercial films yet.

Noncomplementary Couplers. Addition of noncomplementary couplers leads to optical amplification (see Fig. 14). For example, the addition of a magenta coupler to the standard yellow coupler in the blue sensitive layer enhances the side absorption of the yellow dye. Small quantities can be used to fine tune the interimage effects and can, to a certain extent, improve speed and sharpness.⁵⁰

The ultimate limit in this respect is an admixture of all three image dyes in one layer giving complete color desaturation as applied in chromogenic black and white films.

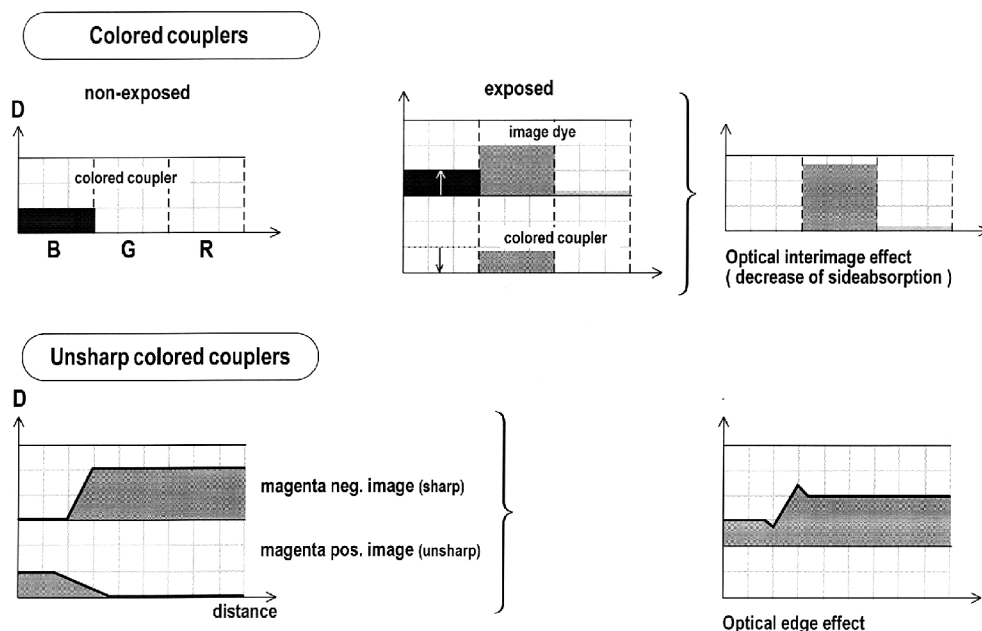


Figure 13. Optical inhibition by colored couplers resulting in optical interimage effects or optical edge effects (B, G, R denote spectral absorption bands of the dyes).

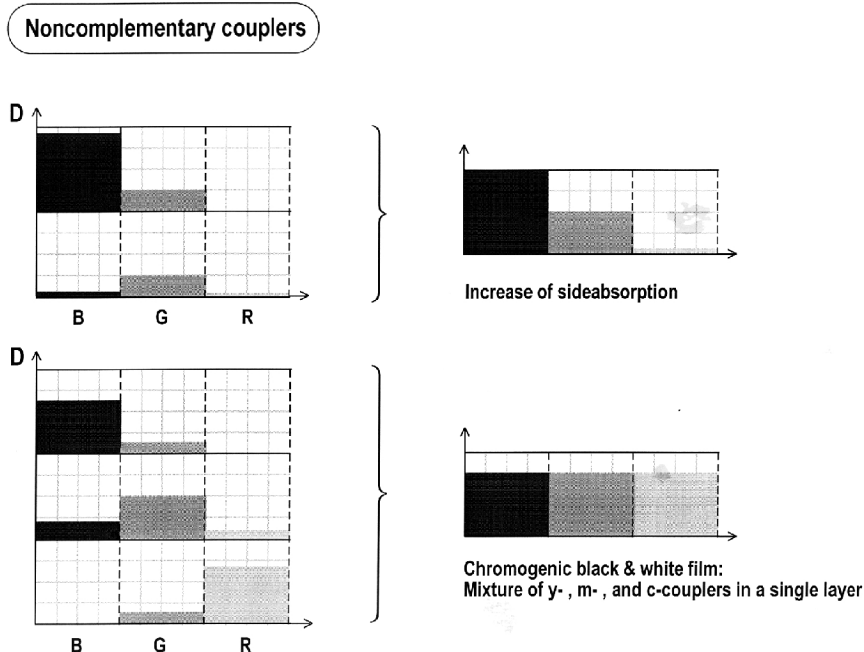



Figure 14. Optical amplification by non-complementary couplers (B, G, R denote spectral absorption bands of the image dyes)

This ends our trip along the obscure routes of chemical image processing. There are probably still further new routes and new mechanisms to be discovered. It remains an open question whether in the future era of digital printers, chemical image processing will be substituted by digital image processing (simplifying the chemistry of films), or whether chemical image processing will be just supplemented by additional digital image processing. 

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