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Spectral Evaluation of Laboratory-Made Silver Halide Emulsions for Color Holography

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Ultra-fine-grain silver halide emulsions with average grain size of around 10 nm were prepared in our laboratory for true color holographic recording. They were spectrally sensitized with various types of cyanine dyes, and their absorption spectra were measured to study the efficiency of sensitization. Their photographic and holographic characteristics were evaluated and discussed in connection with the results of absorption spectrum measurements. Adequately sensitized emulsions showed M- or J-bands in their absorption spectra at the wavelengths of laser beams for exposure. Dye molecules showing no bathochromic shifts of M-bands in emulsion sols contributed very little to spectral sensitization and seemed situated away from silver halide particles. Each of the red, green, and blue holographic gratings recorded on the laboratory-made emulsion plates showed a higher than 50% diffraction efficiency. But some of the best sensitized emulsions did not show the optimum results in diffraction efficiencies.

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Introduction

Extremely high-resolution and low-noise recording materials are necessary for fabricating high-quality reflection holograms. Several materials have been developed for this purpose, for example, silver halide emulsions, dichromated gelatins, and photopolymers. These materials could be panchromatic by adding suitable dyes, and a true color hologram can be recorded in such spectrally sensitized materials. Kubota[1] demonstrated that the sandwich technique enabled the fabrication of true color reflection holograms by using dichromated gelatin plates for green and blue recordings and commercially available silver halide emulsion plates for red recording. Recently, Bjelkhagen et al.[2,3] succeeded in recording color reflection holograms in panchromatic single-layer silver halide emulsion plates, PFG-03C,[4] from Russia. In any case, the properties of commercially available materials are fixed and it is hard for users to modify the characteristics of these materials as they like.

Of the materials mentioned above, silver halide emulsions have the advantage of higher sensitivity than the other materials as well as easy handling. We have studied, by absorption spectrum analysis, the nucleation and growth of silver halide particles prepared by mixing silver ion solutions with halide ion solutions in a rapid solution mixing device.[5,6] Using the same mixing device as in that study, we prepared ultra-fine-grain silver halide emulsions[7] for holography in our laboratory. Emulsion preparation in users' laboratories enables them to modify the characteristics of recording materials, for example, the thickness of the emulsion layer, the balance between sensitivity and resolving power, and even optimization of the spectral sensitivity to the wavelengths of laser beams.

It is impossible to perform direct measurements of the absorption spectra of dyed ordinary photographic emulsions, because they are opaque owing to scattering by the silver halide grains. In such cases, the reflectance spectra have been measured to study the adsorbed dyes on silver halide particles,[8,9] and the Kubelka-Munk equation was applied[10,11] to get the corresponding absorption spectra. However, if the particles in emulsions are smaller than about 30 nm in diameter, the absorption spectra of dyes adsorbed on these particles can be measured directly without the disturbance of scattering. Sahyun and colleagues[12] showed the absorption band of dye-silver ion complex from deconvolution of the absorption spectrum of AgBr nanosol with dye and discussed the role of such complexes in spectral sensitization. The particles in emulsions for high-quality reflection holography are so fine that the absorption spectra of both emulsion sols and coatings on glass plates can be measured directly. In this article, the photographic and holographic characteristics of plates prepared with laboratory-made silver halide emulsions were evaluated and discussed in connection with the results of absorption spectrum measurements.

Experimental

Preparation of Silver Halide Emulsion. All the reagents except for the gelatin and dyes were purchased from Waken-yaku Co., Ltd. High-restraining-power alkaline ossein gelatin (P-4261, Nitta, Osaka, Japan) was used to prepare all the emulsions. The sensitizing dyes were supplied by our request from the Japanese Research Institute for Photosensitizing Dyes Co., Ltd.[13] The reagents were used without further purification. Ultra-fine-grain silver halide emulsions were prepared according to the following procedures.

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1. Mixing (emulsification). Equal volumes (10 ml) of the three solutions, listed in Table I, were mixed with a new two-step three-solution rapid mixing device[14] (Otsuka Electronics, Osaka, Japan) at 38°C. The new mixing device prepared a much larger volume of emulsion sol by a single operation than the previous one,[15] and was modified to operate more easily in a dark room. Solutions A and B were mixed at the first mixing step, then Solution C was combined with the mixture of A and B at the second step. The nominal overall mixing time was less than 100 ms. The concentration of each ingredient in the final mixture became one third of the values in Table I (except for gelatin, because both Solutions B and C contained it).
2. Freezing. After the mixing, the emulsions were put immediately in a freezer for the sake of instant gelation and stored for 24 h at –18°C.
3. Washing. The frozen emulsions were crushed and washed with distilled water of lower than 5°C to remove soluble salts. The freezing of emulsions promoted the removal of useless salts in the washing process.
4. Ripening. The washed emulsion gels were kept at 38°C to ripen. The ripening time was varied from 10 to 120 min. Shorter ripening time was more favorable for preventing the emulsion grains from growing. The warming of emulsions, however, was needed to make chemical sensitizers effective. At the beginning of ripening, 0.05 ml of 2.35×10^{-3} mol dm^{-3} $Na(AuCl_4)$ and 0.3 ml of 6.2×10^{-2} mol dm^{-3} $Na_2S_2O_3$ were added. At 5 min before the end of ripening, 4 ml of 4.0×10^{-4} mol dm^{-3} sensitizing dye was also added to 20 ml of the emulsion sol.

Immediately after ripening, the prepared emulsion sols were cooled to store at 5°C or coated directly on glass plates and dried in clean air at 25°C. Emulsions I to IV (6 ml) or Emulsions V to VIII (2.4 ml) were coated on a 4 × 5-in. glass plate to make a coating about 10 μm thick after drying. Chemically sensitized emulsion VII, ripened for 15 min at 38°C, was used for the experiments of spectral sensitization. The average grain diameter of emulsion VII was about 10 nm and was most suitable for photographic and holographic characterization experiments.

Absorption Spectrum Measurements. A small portion of the prepared emulsion sol was filled into a quartz cell for absorption spectrum measurements. Direct absorption spectrum measurements of emulsion sols could be performed with a conventional spectrophotometer (UV-2500 PC, Shimadzu, Kyoto, Japan). For evaluation of the spectra of emulsion sols, the absorption spectra of aqueous and 50% aqueous ethanol solutions of sensitizing dyes were also measured. All the measurements were performed at room temperature.

Photographic and Holographic Characterization of Emulsion Plates. Photographic characteristics of laboratory-made silver halide emulsion plates were evaluated by the following processes:

- Exposure: 633 nm (He-Ne, 50 mW) laser beams. Exposure times were varied. The beam intensity on the exposure plane was about 1.25 mW cm^{-2}.
- Development: CW-C1,[16a] 5 min at 15°C.
- Fixation: Fujifix, 5 min at 15°C.

TABLE I. Concentrations of Solutions for Emulsion Preparation Before Mixing

	Solution A	Solution B	Solution C	
Emulsion	$AgNO_3$	Gelatin	KX*	Gelatin
	mol dm^{-3}	wt%	mol dm^{-3}	wt%
I	0.076	2.4	0.07	2.4
II	0.146	2.4	0.14	2.4
III	0.216	2.4	0.21	2.4
IV	0.286	2.4	0.28	2.4
V	0.076	6.0	0.07	6.0
VI	0.146	6.0	0.14	6.0
VII	0.216	6.0	0.21	6.0
VIII	0.286	6.0	0.28	6.0

* KBr + KI (Br: I = 9: 1)

After fixation, the processed plates were washed and dried and the optical densities of the exposed parts of the plates were measured with a Sakura PDA 25 densitometer.

Color holographic characteristics of the plates were evaluated as follows:

- Exposure: 488 nm (blue), 515 nm (green) (Ar-ion, 200 mW) and 633 nm (red) (He-Ne, 50 mW) laser beams. Exposure times were varied. The intensity of each beam on the exposure plane was about 1.25 mW cm^{-2}. The interference fringes were recorded by introducing the object beam from the emulsion side and the reference beam from the glass side to the plate. The total intensity of the two beams from different directions was about 2.5 mW cm^{-2}. The angle between object and reference beams was so set as to give interference fringes of 5090 lines/mm spatial frequency for 633-nm light.[17]
- Development: CW-C1, 5 min at 15°C.
- Bleaching: Ferric EDTA,[16b] 7 min at 15°C.

Reflectance spectra of holographic gratings recorded on processed plates were measured with the same spectrophotometer as that used for absorption spectrum measurements, but the spectrophotometer was equipped with an integrating sphere. The reconstructed spectra indicate the wavelength selectivity which means the dependence of diffraction efficiency on wavelength. We regarded the peak values of reflectance in these spectra as diffraction efficiencies of recorded holographic gratings.

Results and Discussion

Absorption Spectra of Emulsion Sols without Sensitizing Dyes. Figure 1 shows the absorption spectra of emulsion sols without dyes. The optical path length was 1 mm. The steep rise of absorbance at wavelengths shorter than 500 nm, observed in each spectrum, was due to crystalline silver iodobromide.[18] The intensity of the absorption band was proportional to the silver halide concentration of emulsion sol in Fig. 1(b). But in the spectra of emulsions II to IV containing 1.6 wt.% gelatin, the absorbance rose gradually toward shorter wavelengths in the whole observed wavelength range [Fig. 1(a)]. This rise was due to scattering and suggested the coexistence of larger particles in the sols. Antoniades and Wey studied the effect of gelatin on the agglomeration of fine AgBr particles,[19,20] and showed that 50 g gelatin/mol AgBr was the lower limit of gelatin concentration to avoid the agglomeration of particles. The gelatin-to-AgX ratio of Emulsion IV was 171 g gelatin/mol AgBrI, i.e., more than three times as large as the limiting value. This disagreement probably arose from the presence of iodide ions and/or the ripening stage (15 min at 38°C) in

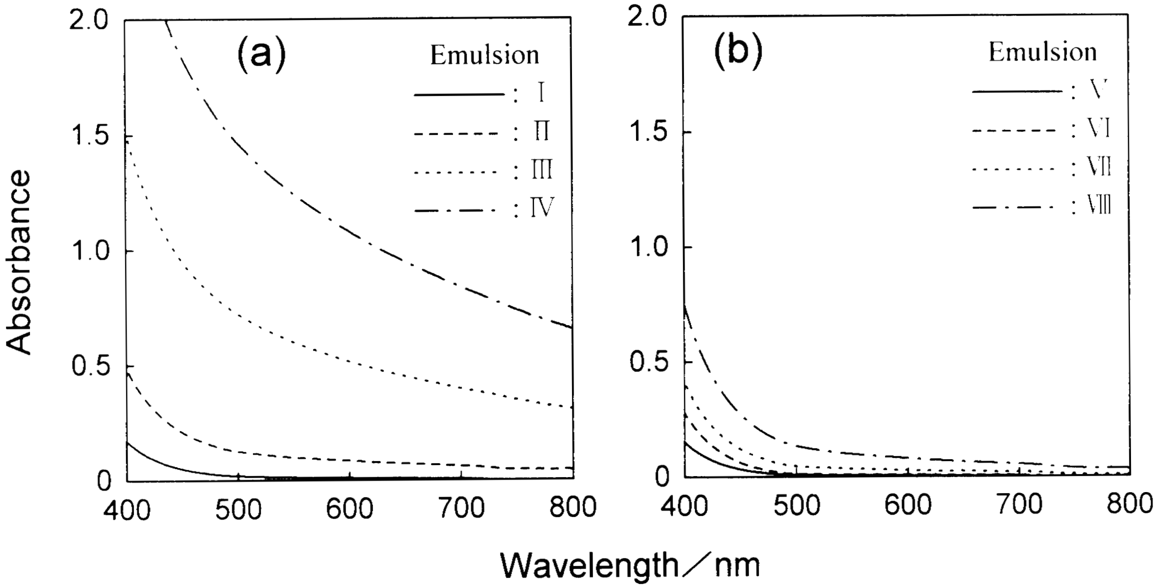


Figure 1. Absorption spectra of emulsion sols at room temperature. Optical path length: 1 mm. The spectra of emulsions II to IV were influenced by scattering of larger particles in sols.

TABLE II. Average Grain Sizes of Emulsions Ripened for 15 Min at 38°C

Emulsion	Diameter nm	Emulsion	Diameter nm
I	8	V	6
II	10	VI	8
III	12	VII	9
IV	15	VIII	10

TABLE III. Absorption Maximum Wavelengths λ_{max} of Dyes in Ethanol

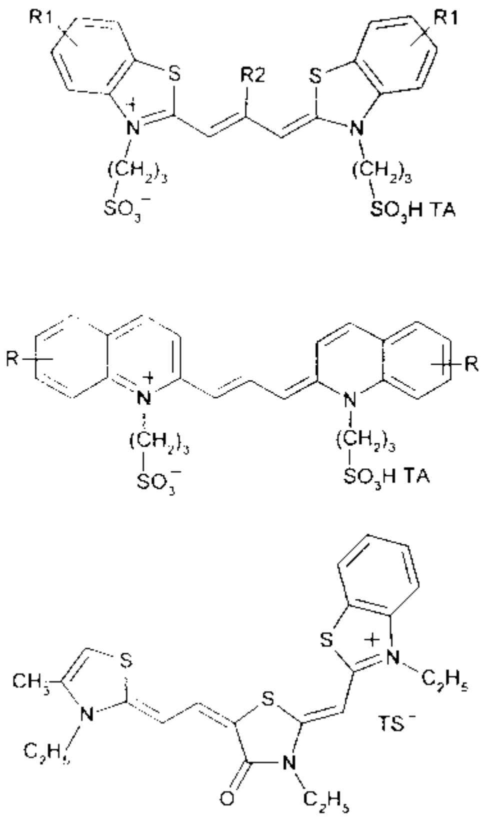
Dye	λ_{max}/nm	Dye	λ_{max}/nm
1	599	10	609
2	573	11	615
3	580	12	606
4	601	13	486
5	563	14	499
6	548	15	480
7	554	16	485
8	571	17	505
9	565		

our case. The average grain sizes of our emulsions were obtained from direct measurements on transmission electron micrographs (H-300, Hitachi, Tokyo, Japan). They were in the range of 6 to 15 nm, as listed in Table II.

Absorption Spectra of Sensitizing Dyes. Dyes 1 to 12 were used as spectral sensitizers in the red region, and dyes 13 to 17 in the blue and green regions. The structures of these cyanine dyes are shown in Figs. 2 and 3, respectively, and the absorption maximum wavelengths, λ_{max}, of their ethanol solutions are listed in Table III. The spectra of dyes for red sensitization in water (solid lines) in 0.5 mol dm^{-3} aqueous KBr solution (dotted lines) and in 50% aqueous ethanol solution (broken lines) are shown in Fig. 4. The dye concentration of each solution was 4×10^{-6} mol dm^{-3}. The optical path length was 10 mm. Symbols M, D, H, and J (also J′) in Figs. 4 to 6 indicate that the absorption bands are due to dye monomers, dimers, H-aggregates, and J-aggregates, respectively. Almost all molecules of these twelve dyes existed as monomers in 50% aqueous ethanol solutions. Addition of salts such as KBr promoted the aggregation of dyes in aqueous solutions by the effect of salting-out.[21] In aqueous KBr solutions, dyes 2 to 8, all thiacarbocyanine-type dyes, showed a large tendency toward J-aggregation, but dye 1 and dyes 9 to 12 did not. These five dyes tended to dimerize.[22,23] Especially dye 9 was apt to exist as dimers. Dyes 11 and 12 precipitated partly in aqueous salt solutions.

Absorption Spectra of Spectrally Sensitized Emulsion Sols. Absorption spectra of emulsion sols dyed with red-sensitizing dyes are shown in Fig. 5, and those dyed with green and blue in Fig. 6 (optical path length: 1 mm). The arrows in the figures indicate the wavelengths of laser beams: red, 633 nm; green, 515 nm; and blue, 488 nm. The monomer absorption band (M-band) is known to shift bathochromically by 20 to 30 nm[24] when dyes are adsorbed on AgBr crystal surface. As is seen, for example, the M-band of dye 1 red-shifted by as much as 30 nm in emulsion sols [compare Fig. 5(a) with Fig. 4(a)]. All red-sensitizing dyes showed bathochromic shifts in the range of 20 (dye 3) to 40 nm (dye 4). Blue- and green-sensitizing dyes in the present work, however, showed no shift of M-bands. Sharp J-bands were observed in aqueous dye 2, 5, and 6 solutions with salts, but J-bands were not observed in emulsion sols dyed with these dyes. A large part of dye molecules existed as dimers in these sols. Most of dyes 10 and 11 molecules existed as dimers or H-aggregates in emulsion sols.

Evaluation of Photographic and Holographic Characteristics of Laboratory-Made Plates. Holographic characterization was performed with both the red-sensitized and the blue/green-sensitized plates, but photographic characterization (sensitometry) was performed only with the red-sensitized plates. Because undyed silver iodobromide plates were sensitive up to the green region, we were interested in spectral red sensitization.



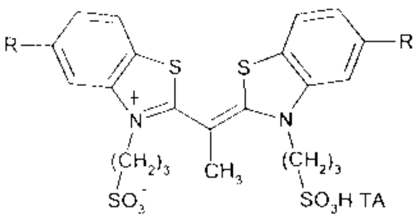
Dye 1 : 4,5,4',5'-dibenzo
Dye 2 : 4,5,4',5'-dibenzo, 9-methyl
Dye 3 : 4,5,4',5'-dibenzo, 9-ethyl
Dye 4 : 4,5,4',5'-dibenzo, 9-phenyl
Dye 5 : 5,5'-dichloro
Dye 6 : 5,5'-dichloro, 9-methyl
Dye 7 : 5,5'-dichloro, 9-ethyl
Dye 8 : 5,5'-dichloro, 9-phenyl
Dye 9 : 5,5'-dimethoxy, 9-ethyl

Dye 10
Dye 11 : 6,6'-dimethyl

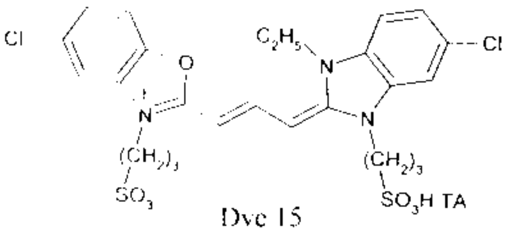
TA : Triethylamine
TS : p-Toluenesulfonic acid

Dye 12

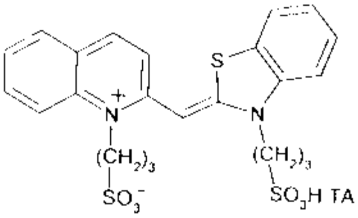
Figure 2. Structures of cyanine dyes for red sensitization.



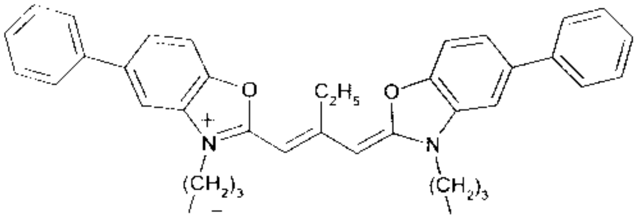
Dye 13 : 5,5'-dichloro
Dye 14 : 4,5,4',5'-dimethoxy



Dye 15



Dye 16



Dye 17

TA : Triethylamine

Figure 3. Structures of cyanine dyes for green and blue sensitization.

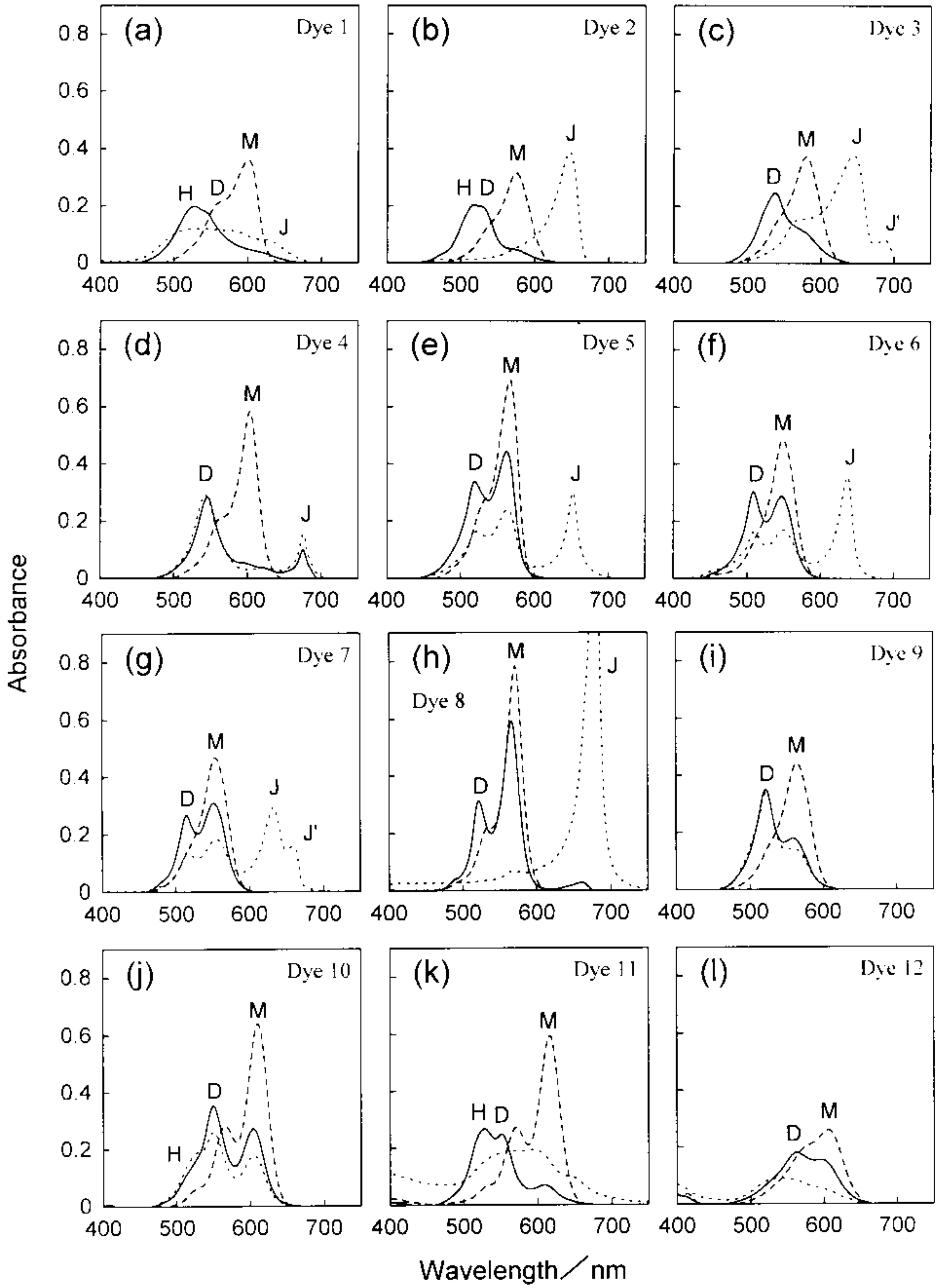


Figure 4. Absorption spectra of dye solutions at room temperature. Solid lines: aqueous solutions, dotted lines: 0.5 mol dm^{-3} aqueous KBr solutions, broken lines: 50% aqueous ethanol solutions. Dye concentration of each solution was 4×10^{-6} mol dm^{-3}. Symbols M, D, H, and J (also J′) indicate the absorption bands of monomers, dimers, H-aggregates, and J-aggregates, respectively. Optical path length: 10 mm.

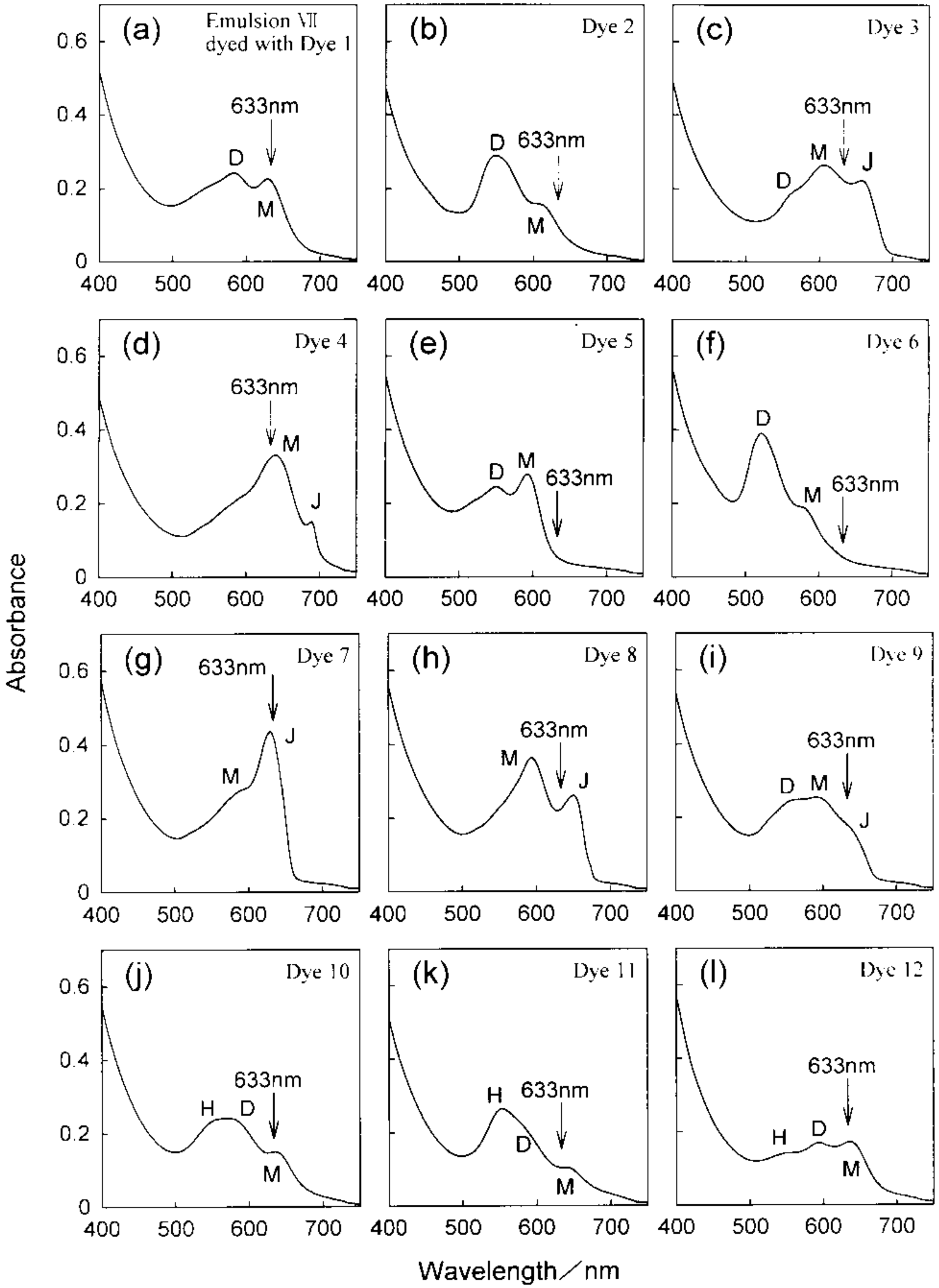


Figure 5. Absorption spectra of emulsion sols with red-sensitizing dyes at room temperature. Emulsion VII was dyed with dyes 1 to 12. Arrows indicate the wavelength of He-Ne laser beams. Symbols M, D, H, and J indicate the absorption bands of monomers, dimers, H-aggregates and J-aggregates, respectively. Optical path length: 1 mm.

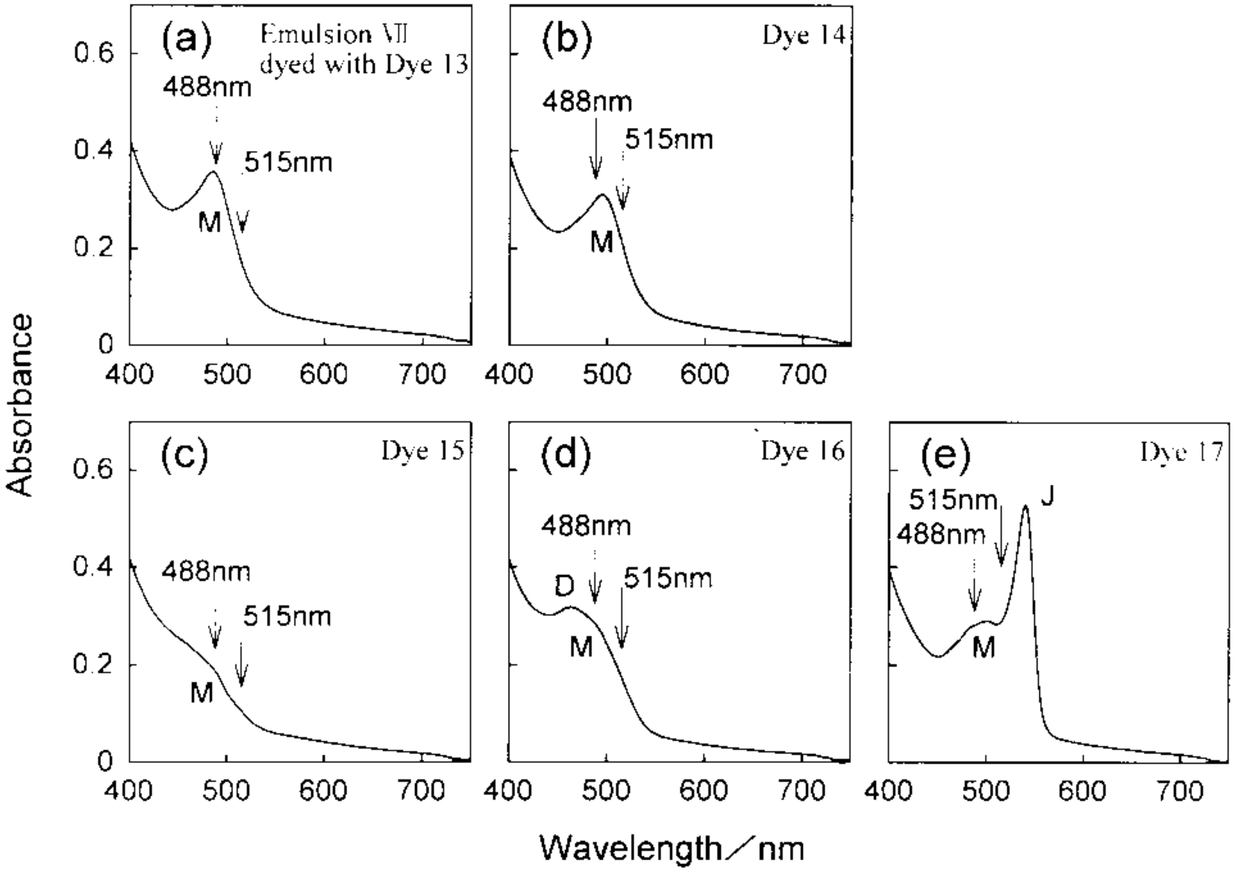


Figure 6. Absorption spectra of emulsion sols with green- and blue-sensitizing dyes at room temperature. Emulsion VII was dyed with dyes 13 to 17. Arrows indicate the wavelength of Ar-ion laser beams. Symbols M, D, and J indicate the absorption bands of monomers, dimers, and J-aggregates, respectively. Optical path length: 1 mm.

Photographic characteristic curves of red-sensitized emulsion plates are shown in Fig. 7. The solid line in Fig. 7(a) shows the result of the undyed emulsion plate, which was only slightly darkened at the exposure of 100 mJ cm^{-2}. In contrast with this, each dyed emulsion plate showed the maximum density of about 5. All the dyes (1 to 12) worked as spectral sensitizers, but their efficiencies were different. Dyes 1, 4, 7, and 12 worked most effectively. The absorption spectra of emulsion sols sensitized with dyes 1, 4, and 12 [Figs. 5(a), 5(d) and 5(l)] showed good agreement of the absorption maximum of the M-band with the wavelength of He-Ne laser beam (633 nm). In the case of dye 7, the J-band was just situated at 633 nm. Dyes 5 and 6 were less effective than the other red-sensitizing dyes. Because dyes 5 and 6 showed a tendency toward J-aggregation in aqueous KBr solutions, they were expected to J-aggregate on silver halide particles in sols and show J-bands around 633 nm, but in fact they did not form J-aggregates. Instead, they formed dimers and showed only the tail of M-band at 633 nm.

Figure 8 shows the diffraction efficiency–exposure curves of plates of emulsion VII dyed with dyes 1 to 12. They were exposed to 633 nm He-Ne laser beams. The values of diffraction efficiency plotted in this figure (and in Figs. 9 and 10) are the peak values of reflectance in such reflectance spectra, as in Fig. 11. The solid line in Fig. 8(a) shows the result of the undyed plate. The maximum efficiency was attained at the exposure of 10 to 30 mJ cm^{-2} in many of the dyes except dyes 2, 5, and 6. These observations agree well with the sensitometric results shown in Fig. 7. But in Fig. 8, appreciable efficiency decrease at higher exposure and lower values of maximum efficiency were observed with dyes 4, 7, and 12. These dyes were very good spectral sensitizers for our emulsions (see Fig. 7). Probably, the alternating developed parts of the fringes in the emulsion coatings with these dyes spread too much to make optimum holographic gratings. To discuss such undesirable results, an important factor coming from the difference between fixation and bleaching after development must be noted here. Silver halide particles remaining after development would be dissolved away from emulsion coatings after fixation, but would remain after bleaching. Phillips[25] showed various procedures of processing, discussed the reactions in emulsions while they were processed, and pointed out the modulation of recorded interference fringes via diffusion transfer of silver halide during bleaching. These remaining silver halide particles would influence the performance of the gratings significantly.

Diffraction efficiency–exposure curves of plates of emulsion VII dyed with dyes 13 to 17 exposed to 515- and 488-nm Ar-ion laser beams are shown in Figs. 9 and 10, respectively. The solid lines show the results of undyed plates as in Fig. 8. In Fig. 1(b), the undyed emulsion seemed to have no effective absorption at 515 nm but showed higher than 30% diffraction efficiency at the exposure of 100 mJ cm^{-2} in Fig. 9. Our emulsion VII without spectral sensitization was sensitive to 515 nm. At 488 nm, undyed emulsion VII showed 40% diffraction efficiency and, if dyed with dye 14, more than 60%. This means that the emulsion could record at least 6500 lines/mm, because

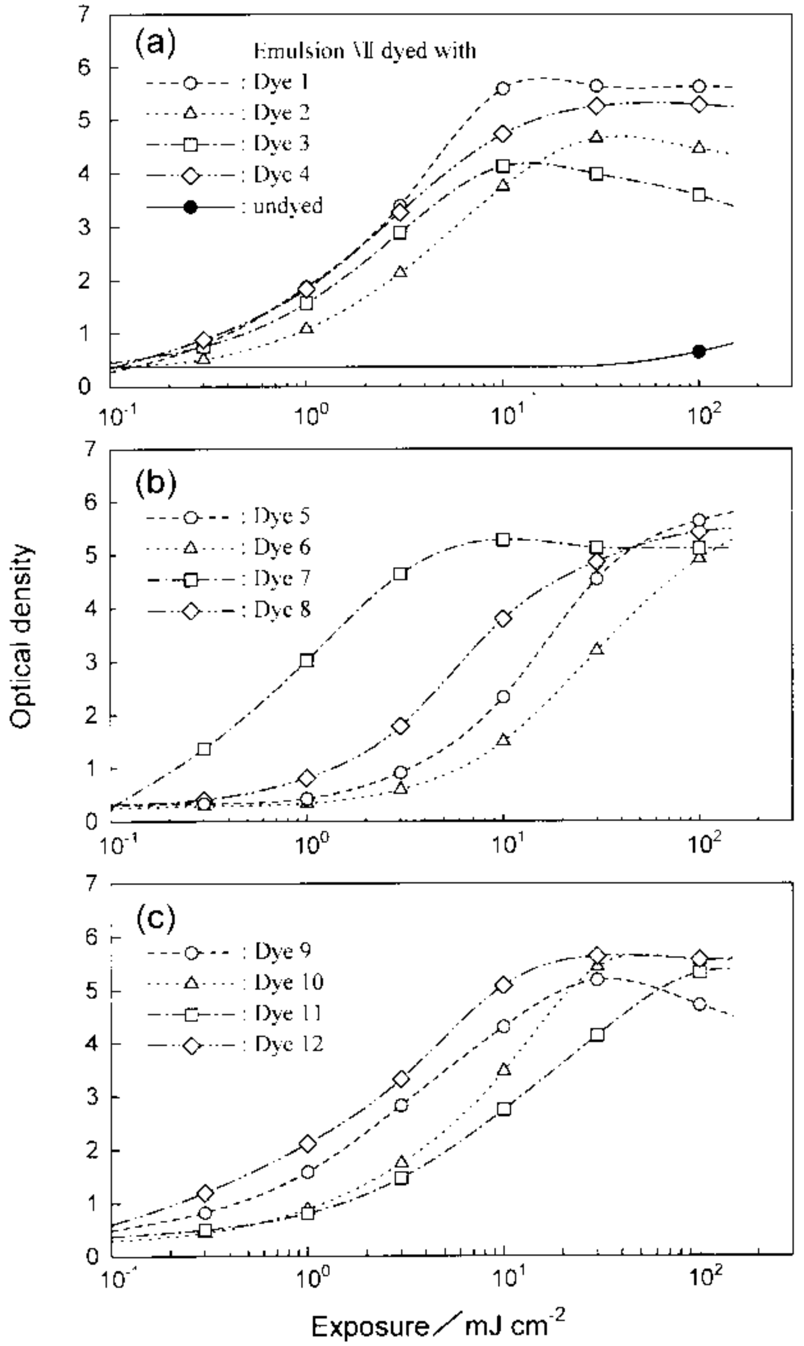


Figure 7. Characteristic curves of dyed emulsion plates exposed to 633-nm He-Ne laser beams (red). Emulsion VII plates dyed with dyes 1 to 12 were evaluated together with undyed emulsion plate [solid line in (a)]. They were developed with CW-C1 for 5 min and fixed with Fujifix for 5 min at 15°C.

the same optical arrangement gave 5090 lines/mm at 633 nm. Compared with dye 17, the dyes 13 to 16 were less effective as sensitizers. These dyes showed no shift of monomer band nor J-band in emulsions. On the contrary, dye 17 showed sharp J-band in emulsion and worked remarkably as a sensitizer. These results suggest that a large part of molecules of dyes 13 to 16 are situated in gelatin apart from silver halide particles, while dye 17 molecules adsorb to the particles and J-aggregate. Dye 17, however, could attain only a lower maximum efficiency than dyes 14 and 15. The same considerations as those given for Fig. 8 about dyes 4, 7, and 12 might be applied to dye 17.

Figure 11 shows an example of the wavelength selectivity of holographic gratings recorded on our laboratory-made plates. These gratings were prepared using the plates of emulsion VII dyed with dye 3 for red recording and dye 15 for green and blue recordings. Reconstructed wavelengths were 610, 504, and 478 nm for red, green, and blue recordings, respectively. The amounts of wavelength shift were 23, 11, and 10 nm. Russian emulsion PFG-03 Color was also tested by the same processes,[26] but at 18°C. The shift amounts of our emulsion were smaller than those of the Russian emulsion. The washing step in emulsion preparation as well as processing at

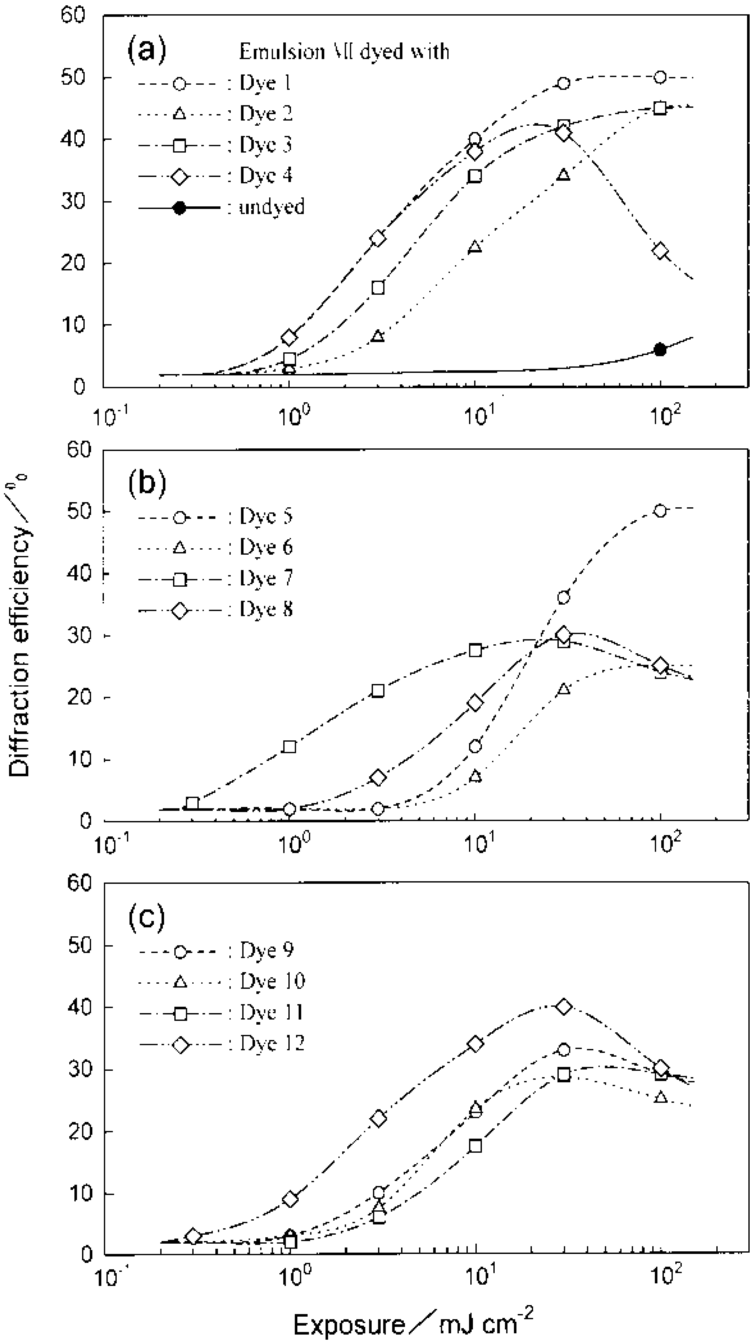


Figure 8. Dependence of diffraction efficiency on exposure to 633-nm He-Ne laser beam (red). Emulsion VII plates dyed with dyes 1 to 12 were evaluated together with undyed emulsion plate [solid line in (a)]. They were developed with CW-C1 for 5 min and bleached with ferric EDTA for 7 min at 15°C.

the relatively low temperature of 15°C were effective in reducing the shift toward a shorter wavelength. The washing procedure removed not only soluble salts but also low molecular weight gelatin and prevented the coatings from shrinking in the processing stage after exposure. Low-temperature processing also suppressed the dissolution of gelatin.

The solution-physical development GP-2 process,[16c] which needed no subsequent bleaching process, was suitable for Russian ultra-fine-grain pure silver bromide emulsions.[27] GP-2 processing was also tried with our laboratory-made emulsion plates. In this article, however, we discussed only the results of CW-C1 development followed by ferric EDTA bleaching, because the results of GP-2 processing were inferior to those of CW-C1 and ferric EDTA processing.

Summary

Silver halide emulsions for color reflection holograms were prepared in our laboratory. Ultra-fine-grain emulsions with average grain sizes of about 10 nm were spectrally sensitized with cyanine dyes. The absorption spectra of dyed emulsion sols could be measured directly because

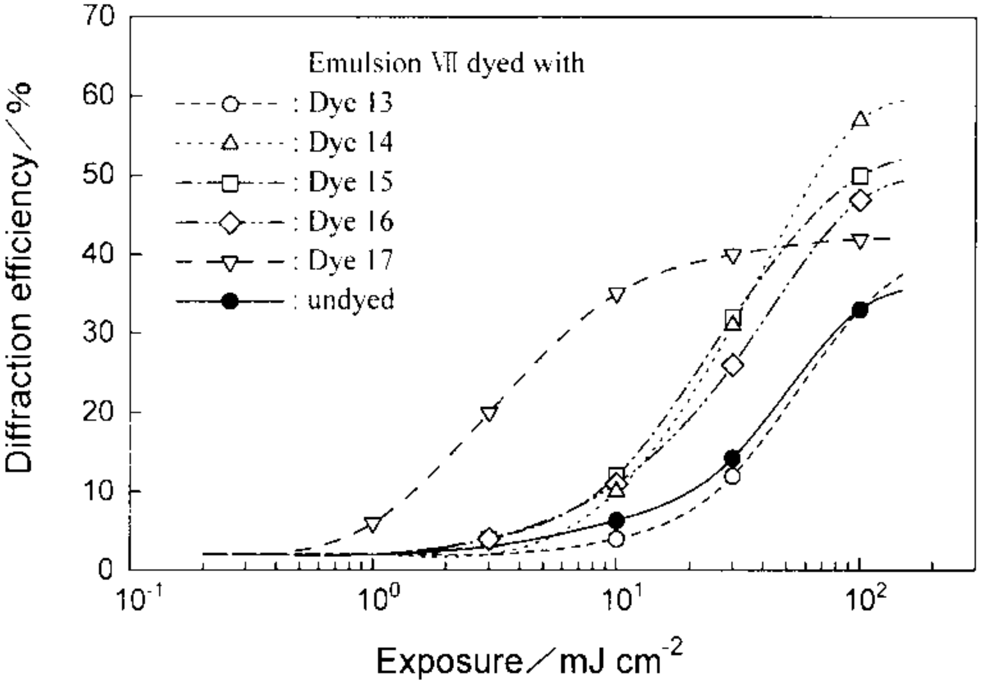


Figure 9. Dependence of diffraction efficiency on exposure to 515-nm Ar-ion laser beam (green). Emulsion VII plates dyed with dyes 13 to 17 were evaluated together with undyed emulsion plate (solid line). They were developed with CW-C1 for 5 min and bleached with ferric EDTA for 7 min at 15°C.

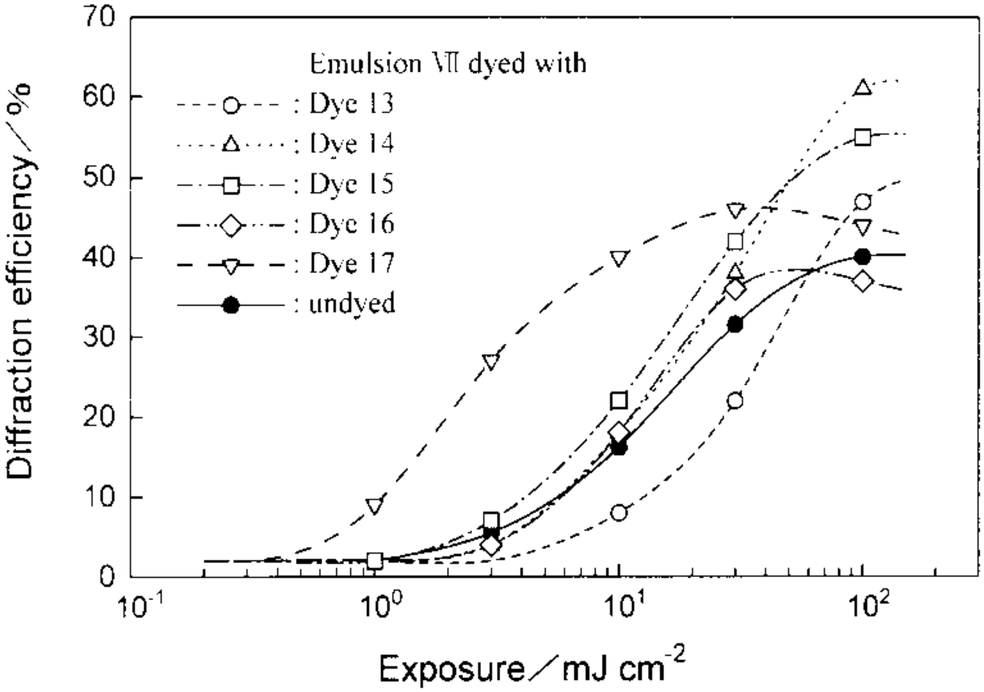


Figure 10. Dependence of diffraction efficiency on exposure to 488-nm Ar-ion laser beam (blue). Emulsion VII plates dyed with dyes 13 to 17 were evaluated together with undyed emulsion plate (solid line). They were developed with CW-C1 for 5 min and bleached with ferric EDTA for 7 min at 15°C.

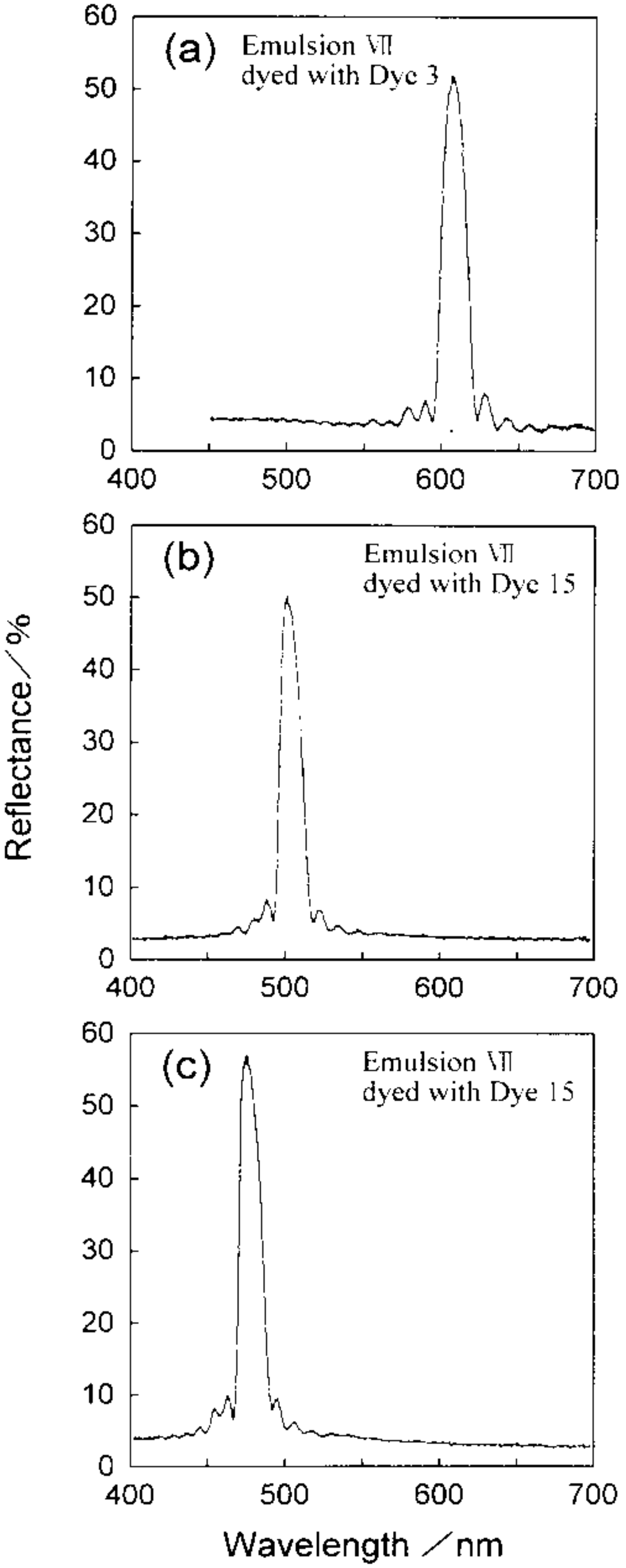


Figure 11. Reflectance spectra of holographic gratings recorded on laboratory-made emulsion plates: (a) red recording with 633-nm He-Ne laser beam, (b) green recording with 515-nm Ar-ion laser beam, (c) blue recording with 488-nm Ar-ion laser beam. The amount of each exposure was 100 mJ cm^{-2}. Reconstructed wavelengths are 610, 504, and 478 nm for red, green, and blue recordings, respectively.

the sols showed no observable scattering. The photographic and holographic characteristics of our laboratory-made emulsion plates were evaluated and discussed in connection with the absorption spectra of dye solutions and dyed emulsion sols. Adsorbed dyes on silver halide particles showed sharp J-bands or red-shifted M-bands in emulsion sols. The effective sensitizing dyes showed good agreement of the absorption maxima of J- or shifted M-bands in the spectra of dyed emulsion sols with the wavelengths of laser beams. Dyes showing no shifts of M-bands did not work as good sensitizers, which suggests that most of these dye molecules were situated away from the silver halide particles. Interference fringes were recorded with red (633 nm), green (515 nm), and blue (488 nm) laser beams on our laboratory-made emulsion plates. The reconstructed reflectance spectrum of each of the red-, green- and blue-recorded holographic gratings showed a higher than 50% diffraction efficiency.

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