Damage to High Speed Color Negative Films from Natural Background Radiation

Yasushi Nozawa* and Hideto Ikoma

Ashigara Research Laboratories, Fuji Photo Film Co., Ltd., Minami-Ashigara, Kanagawa, Japan

Masaharu Okano

Radiation Effects Association, Tokyo, Japan

The effect of natural background radiation is one of the most important determining factors for shelf life of high speed color negative films. Namely, high speed color negative films are inclined to suffer from increase in fog, decrease in speed and deterioration in graininess during their preservation. This damage is caused mostly by natural background radiation. We divided natural background radiation into three components: cosmic rays, environmental radiation, and self-contamination, and determined the fraction of the contribution of each component to this damage. As a model experiment of the above phenomena, we proved that the sensitivity of a silver halide photographic emulsion to gamma rays was in proportion to its intrinsic sensitivity to blue light and to the amount of gamma rays absorbed by the emulsion grains.

Journal of Imaging Science and Technology 46: 270–276 (2002)

Introduction

High speed color negative films offer many benefits: increasing depth of field; reducing the effect of motion blur; diminishing the number of pictures taken with underexposure; and improving background description. Recently, compact cameras with reduced size and extended zoom lenses have successfully come into the market. Downsizing of these cameras with zoom lenses demands the increase in sensitivity of color negative films.[1,2] Although nearly half of the compact cameras produced ten years ago had less than 1.5x zoom lenses, more than fifty percent of compact cameras produced two years ago had more than 2.5x zoom lenses. Although the average f-number of compact cameras produced ten years ago was about 8, the f-number of some of compact cameras now is 11. These trends made it necessary to increase the system speed of compact cameras. In addition to the increase in f-number, remarkable reduction in size of the electronic flash lamp in a compact camera resulted in a decrease in flash range. Thus the system speed of zoom-compact cameras is now more than one stop lower than that of compact cameras produced ten years ago, and the development of high speed films with sensitivity higher than ISO 400 was therefore highly required.

It is well known that a silver halide emulsion is sensitive to high energy radiation as well as to visible light.[3,4] The products on the basis of this character include x-ray films for industry, nuclear film plates, and film badges for dose measurement. However, the effect of natural background radiation on commonly used color negative films was thought to be negligible until the advent of very high speed color negative films with ISO sensitivity of four figures.[5] It is now believed that a color film with high sensitivity to visible light is inclined to be highly sensitive to natural background radiation, and therefore has a short shelf life. This article was undertaken to clarify the mechanism of the damage due to natural background radiation in order to get useful knowledge for the improvement of the shelf life of high speed color negative films.

For the analysis of the damage, we divided the background radiation into following three components: cosmic rays, environmental radiation; and self-contamination. Then, we analyzed the results of the following three experiments: (1) experiment to prove that the damage of the high speed color negative films was caused by natural background radiation, (2) experiment to determine the fraction of contribution of the above stated components to the damage of films, and (3) experiment to detect the interaction between a photographic emulsion and gamma rays, which bring about the highest contribution to the damage of films by the natural background radiation.

Experiments

Shield of Films from Natural Background Radiation

It was necessary to secure a place where the influence of natural background radiation except for self-contamination could be reduced to the smallest possible level. Fortunately, we were allowed to use the Low-Level Radioactivity Laboratory (LLRL) in Kanazawa University. The LLRL is placed in a tunnel at the depth of 240

Original manuscript received November 15, 2001

* yasushi_nozawa@fujifilm.co.jp

TABLE I. Locations for Aging Experimental

1	LLRL1	Inside of a lead block in the LLRL
2	LLRL2	Outside of a lead block in the LLRL
3	Osaka1	The first floor of a 6-storied building in Osaka
4	Minamiashigara1	The second floor of a 5-storied building in Minamiashigara
5	Minamiashigara2	The first floor of a 5-storied building in Minamiashigara
6	Minamiashigara3	Inside of a refrigerator in Minamiashigara1
7	Odawara1	The second floor of a usual 2-storied house in Odawara

m below the surface in order to prevent cosmic rays from reaching films, since the transmission power of cosmic rays is so high as to need such thick ground to absorb them. On the other hand, a thick block of lead could absorb environmental radiation. Because lead itself includes a radioactive isotope, it was necessary to use lead in which this isotope had been allowed to decay for many years after initial refinement. We stored color negative films in a box made of lead, 10 cm thick, which contained only a very small amount of ^{210}Pb (about 0.12 pCi/g). This box had an inside wall made of oxygen-free copper 1.4 cm thick. Furthermore, this box was air conditioned to filter out radioactive ^{222}Rn.

Radiation Measurement

The amounts of background radiation were measured at several places by a portable 7.5 cm spherical NaI (Tl) scintillation counter.[6,7] The detected radiation with energies higher than 3 MeV and lower than 3 MeV were ascribed to cosmic rays and environmental radiation, i.e., gamma rays with various energies, respectively. A thermoluminescence detector (TLD) was also used, depending on circumstances.

Preparation of Samples

The samples used in this study were layers of the AgBrI emulsions as models for color negative films, as well as commercial color negative films on the market when the experiments in this study were carried out.

By changing the temperature and time of emulsion preparation, we obtained thick twinned tabular grains with average sizes of 0.8, 1.2 and 2.1 μm. Moreover, using emulsions with an average grain size of 2.1 μm, we obtained a series of emulsions with various levels of added dye, such as 0, 0.5, 1 and 2 times the optimal amount. Thin layers of the above-stated emulsions were prepared by coating and drying them on cellulose triacetate (TAC) film bases with 2.0 g/m^2 as Ag and 0.1 mol of magenta coupler/mol of Ag. Using the emulsion containing grains with average size of 2.1 μm, we obtained a series of samples with various coated amounts of silver halide, e.g., 1, 2 and 4 g/m^2 Ag. In this series, the amount of coupler coated was fixed at 0.1 mol of magenta coupler/mol of silver halide when the coated amount of silver was 2 Ag/m^2.

Conditions of Aging

We stored several films under conditions with various amounts and energy spectrum of natural background radiation; in boxes made of lead, stainless steel, or aluminum, with 2 and 10 mm thick, at places in Japan, including the LLRL, as in the places 3 ~ 7 in Table I, we stored the films in the boxes as listed in Table II.

TABLE II. Conditions of Aging (Shielding)

1	Outside of a box
2	In a box made of lead 2 mm thick
3	In a box made of lead 10 mm thick
4	In a box made of stainless steel 2mm thick
5	In a box made of stainless steel 10 mm thick
6	In a box made of aluminum 10 mm thick

Sensitometry

The sensitometry and the measurement of R.M.S. granularity were carried out under the following conditions. Exposure time was 1/100 second. For measurement of the intrinsic sensitivity to blue light, model films were exposed to light through a Fuji Photo Film band path filter BPN42. Instead of natural background radiation, we used 120 kVp x-rays filtered by H_2O with 5 cm thick for the study of the interaction between an emulsion and high energy radiation. Development was performed using CN-16 chemistry at 38°C with the development time of 3 min and 15 sec for commercial films and the development time of 2 min and 45 sec for model films. The sensitivities of these films were given by the reciprocal of the exposure to give an optical density of 0.20 above fog density.

Results and Discussions

Damage to High Speed Color Negative Films

Photosensitive materials suffer from the deterioration of performance owing to their exposure to natural background radiation. Since processes to detect visible light and high energy radiation are fundamentally the same, it is natural that films with high sensitivity to visible light are also highly sensitive to natural background radiation. Natural background radiation usually causes not only increase in fog, but also decrease in speed and increase in graininess.

Figure 1 shows the change in characteristic curves of a color negative film (Super HR-1600, Fuji Photo Film, 1986), which was stored for 2 years at Osaka1, Hiroshima, and Fukuoka Offices of our company, and at the LLRL1 in Kanazawa University. Compared with the minimum density at LLRL1, the minimum densities were changing significantly at Osaka1, Hiroshima, and Fukuoka. Compared with the change in minimum density, the change in gradation was negligible. Thus, the effect of natural background radiation on characteristic curves of films looked like the effect of uniform light exposure. However, as shown in Fig. 2, natural background radiation dramatically increased R.M.S granularity contrary to the effect of uniform light exposure. Figure 3 shows the relationship of the increase in fog density of a red sensitive layer after preservation for 6 months at Osaka1 with ISO sensitivity of several color negative films, that were commercially available on the market when this experiment was carried out. As seen here, the higher the sensitivity of the film was, the larger the increase in fog density. This result strongly indicates that the mechanism of fog formation by natural background radiation was similar to the mechanism of latent image formation by light.

Table III shows the shielding effect of the LLRL, as measured by a scintillation counter. The amount of cosmic rays in the tunnel was 0.25%~0.50% of that of the rays on the ground. Figure 4 shows the shielding effect of the box made of lead. The abscissa indicates the relative wavelengths of gamma rays. The amount of the environmental gamma rays inside the box was between 1% and

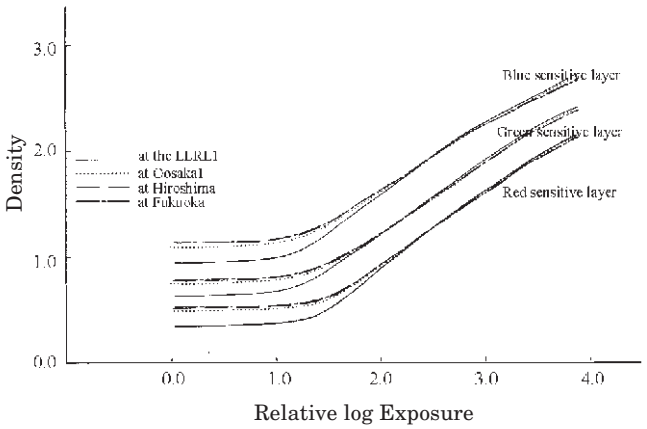

Figure 1. The change in characteristic curve of blue sensitive (upper curves), green sensitive (middle curves), and red sensitive (lower curves) layers of Fuji Color Super HR1600 films which were stored for 2 years at Osaka1 (dotted lines), Hiroshima (broken lines), Fukuoka Offices (dotted and broken lines) of our company, and at the LLRL1 (solid lines).

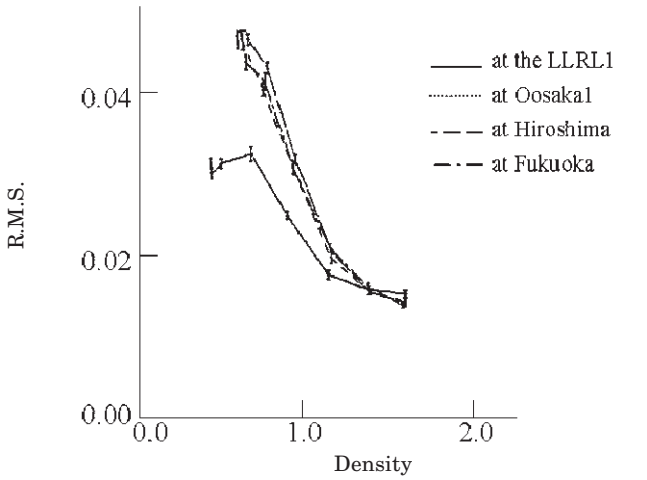

Figure 2. The change in R.M.S. granularity of a red sensitive layer of Fuji Color Super HR1600 films that were stored for 2years at Osaka1 (dotted lines), Hiroshima (broken lines), Fukuoka Offices (dotted and broken lines) of our company, and the LLRL1 (solid lines).

0.1% of that of the rays out of the box. Namely, natural background radiation had practically no influence on the film in the box placed at the LLRL1. Figure 5 shows the increase in fog density of a red sensitive layer in a color negative film (HR1600, Fuji Photo Film, 1984) as a function of the amount of natural background radiation accumulated during its storage for six months at various places including LLRL1, LLRL2, Osaka1, Minamiashigara1, Minamiashigara2, and Minamiashigara3. The result with the film stored at LLRL1 is indicated by the closed square with the smallest amount of radiation in this figure. The results are summarized as follows.

1. The damage was very small at LLRL1.
2. The increase in fog during storage was in proportion to the amount of radiation.
3. The increase in fog was in proportion to the sensitivity of the film.

TABLE III. Amount of Cosmic Rays at Various Places

	Amount of cosmic rays (relative)
At the surface 1	4.2×10^2
At the surface 2	2.8×10^2
In the LLRL	1.2×10^0

The surface 1: right above the LLRL
The surface 2: at the beginning of the tunnel

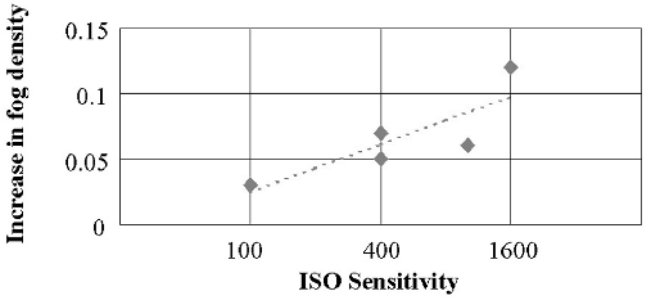

Figure 3. Increase in fog density of red sensitive layers in commercial color films owing to their storage for 6 months at Osaka1 as a function of their ISO sensitivity.

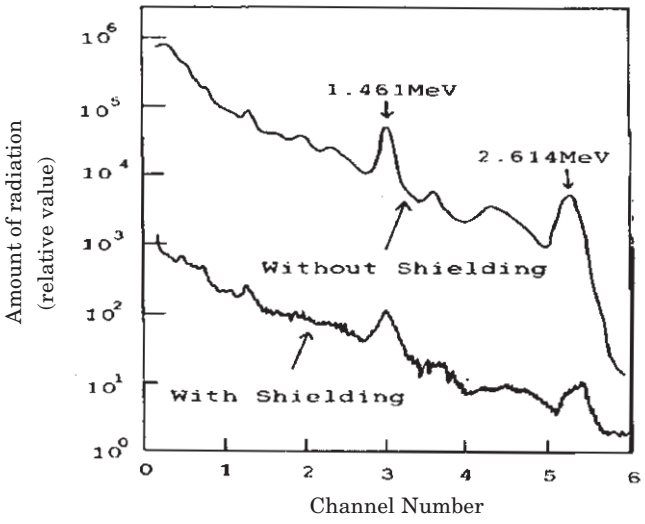

Figure 4. Energy distribution of the detected natural background radiation with and without shielding by lead 10 cm thick and filtering out radioactive substances in the air.

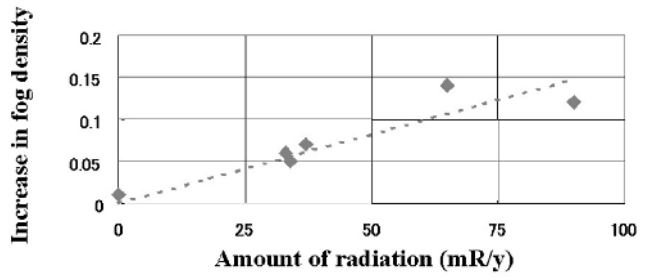

Figure 5. Increase in fog density as a function of amount of the environmental radiation of a red sensitive layer of Fuji Color HR1600 films owing to their storage for 6 months at Osaka1.

On the basis of the above results, it is concluded that the damage was caused by natural background radiation.

Components of Natural Background Radiation

As described in the **Introduction**, we divided natural background radiation into three components, cosmic rays, environmental radiation, and self-contamination.

Cosmic rays are high energy radiations incident on the earth from the universe, and are classified into primary and secondary cosmic rays. The majority of primary cosmic rays consist of protons and alpha particles. Secondary cosmic rays are generated by the interaction of primary cosmic rays with the atmosphere. Most doses on the surface of the earth are secondary cosmic rays. It is known that the dose of cosmic rays increases with altitude. The dose at the altitude of 1500 m is about double of that at sea level. Moreover, the dose increases with increasing latitudes. Because the dose is influenced by solar activity, it changes with time.

The origin of the environmental radiation is radioactive nuclei with long lifetimes that exist in nature. From the time of the birth of the earth, these nuclei existed in the crust and are called primitive radioactive nuclei. The doses, which originated from the following three primitive radioactive nuclei, are predominant. They are ^{40}K, whose half-life is about 1,250 million years, Thorium series with ^{232}Th as a parent nuclide, whose half-life is about 14,000 million years, and Uranium series with ^{238}U as a parent nuclide, whose half-life is about 4,500 million years. Because these nuclei are contained in almost all substances in various concentrations, the amount of the environmental radiation varies from place to place. The penetrating power of beta rays is so weak that substances surrounding films, such as packaging materials, absorb almost all of them. Therefore, photosensitive materials are seldom influenced by beta rays. It is gamma rays whose penetrating power is strong enough to actually influence films. As a result of interaction between gamma rays and surrounding substances, gamma rays have a broad energy distribution extending to the low energy side.

The third component of natural background radiation is called "self-contamination". The origins are radioactive nuclei with long half-life, which actually exist in films themselves. The dose of beta rays, which originated from ^{40}K in films, is predominant in the self-contamination. Beta rays are fluxes of electrons. The interaction of high energy electrons with substances is strong. The high energy electrons generated outside films do not reach the films. On the other hand, the electrons, generated inside films, cause heavy damage to them. (There are two kinds of high speed electrons that appear within a film. One of them is a beta ray from radioactive materials in films, which is the origin of the self-contamination. The other is a secondary electron that is generated in the film when the film absorbs gamma rays.)

We experimentally determined the fraction of the contribution of each component to the damage according to the following way. First of all, the LLRL1 was used to get the storage condition under which a sample was free from external radiation. Secondly, we stored films under conditions with varying amounts of natural background radiation and various energy spectra in many kinds of metal boxes at many places in Japan, as already explained in the **Conditions of Aging** section. Thirdly, the amounts of natural background radiation at several places were measured by a portable 7.5 cm spherical NaI (Tl) scintillation counter, and were analyzed.

TABLE IV. Dose of Cosmic Rays and Gamma Rays at Various Places

		Dose of cosmic rays μR/h	Dose of γ rays μR/h	F/D*
At Odawara1	Without shielding	2.91	3.29	1.00
At Osaka1	Without shielding	1.47	6.94	0.85
	Shielding by stainless steel with 10 mm thick	1.29	5.55	0.77
	Shielding by lead with 10 mm thick	1.38	3.48	0.52

*F/D: flux/dose (number of photons/1 μR)

Table IV shows an example of the measurement of natural background radiation in a normal house in Odawara (Odawara1) and the Osaka branch office of our company (Osaka1). The dose of gamma rays at Odawara1 was smaller than that at Osaka1, since Odawara area is covered with Kanto loam layer, which contains little radioactive material. The Osaka level was nearly average for Japan and the world. The doses of cosmic rays at the two places were different, depending on the filter effect. In Odawara1, the measurement was made at the second floor of an usual 2-storied house. On the other hand, in Osaka1, the measurement was made at the first floor of a 6-storied building. The interception by metals is more effective for the lower energy component than for the higher energy gamma rays, and more effective for gamma rays than for cosmic rays. The flux per dose is the number of photons per 1 μR, and the smaller the flux per dose, the higher is the energy of gamma rays.

We used the fog density of a red sensitive layer in HR1600 films in this experiment, since we knew that the fog density of a red sensitive layer in a HR1600 film changed little by heat. We accordingly made the following three assumptions: in the first place, the contribution of cosmic rays to film damage was proportional to the amount of cosmic rays; second, the contribution of the environmental radiation was proportional to the amount of gamma rays absorbed by a silver halide emulsion that was evaluated on the basis of the absorbance as a function of gamma rays energy; and third, the contribution of the self-contamination was equal to the damage observed during its preservation at the LLRL1.

The increase in fog density of films as a function of the amount of the background radiation during their storage under various conditions was analyzed under the following equation,

$$\Delta\mathrm{Dmin} = a\,\mathrm{X} + b\,\mathrm{Y} + c \qquad (1)$$

where ΔDmin is the increase in fog density of a red sensitive layer of a high speed color negative film (HR1600) as a result of its preservation for 19 months under various conditions, X is the amount of cosmic rays, Y is the amount of gamma rays absorbed by silver halide grains, a and b are constants, and c is a constant that is equal to the damage detected as a result of storage for 19 months at LLRL1.

Figure 6 shows the increase in fog density of the sample stored at LLRL1. The increase in fog density of 0.055 was ascribed to the contribution of the self-contamination for 19 months. The constant c in Eq. (1) was therefore 0.055. The constants a and b in Eq. (1) were determined by the regression analysis. The constant a was 3.8×10^{-2}, and b was 1.9×10^{-2}. In Fig. 7, the abscissa indicates the increase in the fog density as a result of the application of the measured values of the radiation in each preservation place to Eq. (1), and

TABLE V. Fraction of Each Component in the Background Radiation in Osaka1 as Estimated by Use of Eq. (1)

Component	The Fraction	
Cosmic rays	*a* X = 0.056	about 20%
Environment gamma rays	*b* Y = 0.201	about 60%
Self-contamination	*c* = 0.055	about 20%

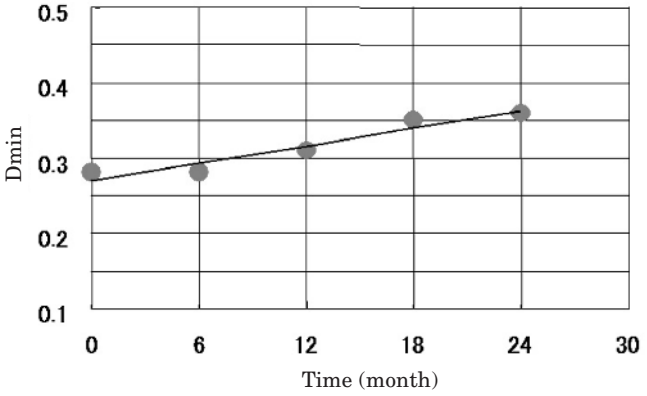

Figure 6. Increase in fog density of a red sensitive layer in Fuji Color HR1600 films at the LLRL1

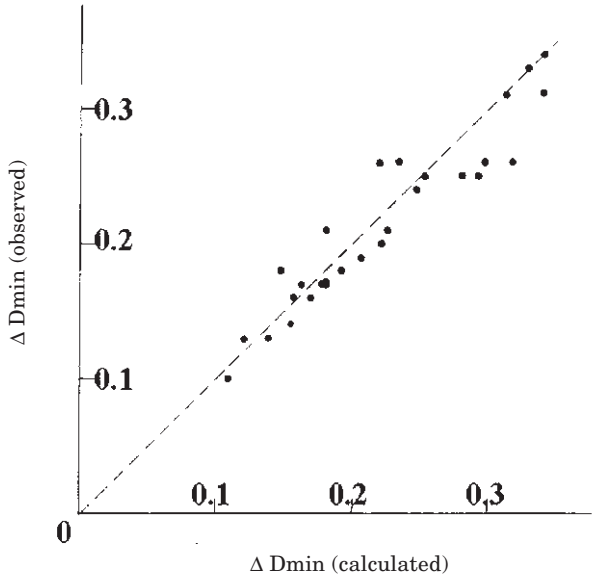

Figure 7. Relation between observed and calculated values of the increase in fog density (ΔDmin) of red sensitive layers of Fuji Color HR1600 films owing to their storage for 19 months at several places. The calculated values were given by Eq. (1) (see text).

the ordinate indicates the actually observed increase in the fog density after 19 months preservation. The agreement between them was satisfactory, and proved our assumption.

The obtained equation was available to predict the increase in fog density caused by each of the three components of natural background radiation under conditions with variation of X and Y. For instance, the fraction of each component in Osaka1 in Japan was shown in Table V. Although the damages significantly depended on the storage places and kind of sensitized materials, we think that the fraction of each component in natural background radiation at most places in the world might not differ too much from that shown in Table V.

Sensitivity of a Photographic Emulsion to Gamma Rays

In this section, we will show you our experimental result on the interaction between gamma rays and silver halide emulsion grains. The contribution of the environmental gamma rays to film damage is the highest among three components of natural background radiation. It is known that the interaction of a gamma ray quantum with a silver halide emulsion grain first produces a high energy secondary electron, and then this high energy electron interacts with grains to produce many more electrons that take part in the formation of silver clusters, i.e., fog centers caused by natural background radiation. This consideration led to the following equation,

$$S_\gamma = k_1 A_\gamma S_e \tag{2}$$

where A_γ is the amount of the gamma rays absorbed by silver halide grains, S_γ and S_e are the sensitivities to the gamma rays and to high energy electrons, respectively, and k_1 is an empirical constant. Equation (2) is based on the assumption that the sensitivity of a color film to high energy electrons is proportional to its intrinsic sensitivity to blue light, which depends on the efficiency with which electrons in the conduction band of silver halide are used for latent image formation, as expressed by the following equations.

$$\begin{aligned} S_e &= k_2 S_I \\ S_\gamma &= k_1 k_2 A_\gamma S_i \end{aligned} \tag{3}$$

where S_i is the intrinsic sensitivity to blue light, and k_2 is a constant.

An examination was made on the proportionality of S_γ to S_i for various emulsions. In order to simulate the effect of the environmental gamma rays, we used 120 kVp x-rays to get x-ray sensitivity, which should be proportional to S_γ, and we regard blue sensitivity as S_i. Figure 8 shows the result of sensitometry of three emulsions of the same type with variation in average grain size. Figure 9 shows the result of the same experiment for four emulsions with variation in added amount of dye. The abscissa indicates their relative blue speed, and the ordinate indicates relative x-ray speed. These figures clearly indicate that the relative x-ray speed was proportional to the relative blue speed. This result supports Eq. (3) as proposed in this article. Figure 10 shows the relation between x-ray and blue sensitivities of the monolayer coatings of an emulsion with large variation of silver halide coating weight. On the contrary to the results in Figs. 8 and 9, blue sensitivity increased only slightly with increasing the amount of coated silver halide grains, while x-ray sensitivity increased significantly. Figure 11 shows the relation between x-ray sensitivity and the silver halide coating weight, and Fig. 12 shows the relation between the blue sensitivity and the silver halide coating weight. The x-ray sensitivity increased significantly with increasing the coated amount of silver halide grains, while the blue sensitivity did not.

We analyzed the results on the basis of the following considerations. First of all, the x-ray sensitivity was

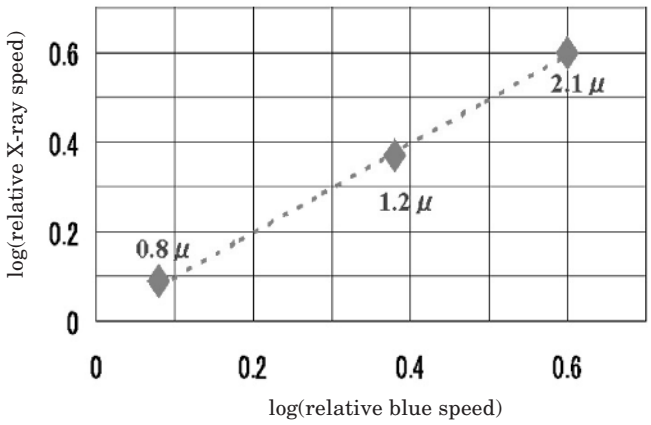

Figure 8. X-ray speed as a function of blue speed of AgBrI emulsions with grain size as described in the figure.

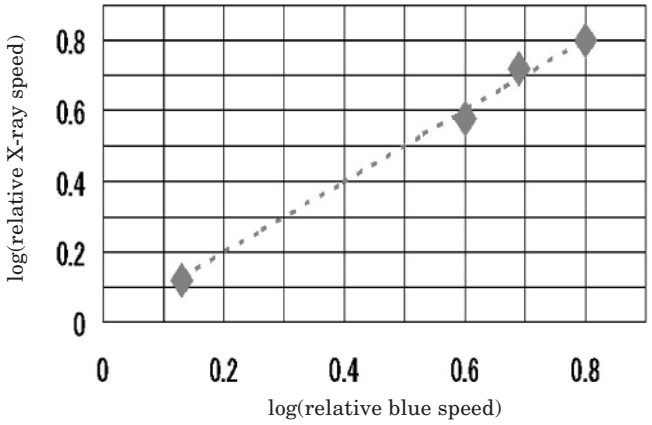

Figure 9. X-ray speed as a function of blue speed of AgBrI emulsions with variation in added amount of dye.

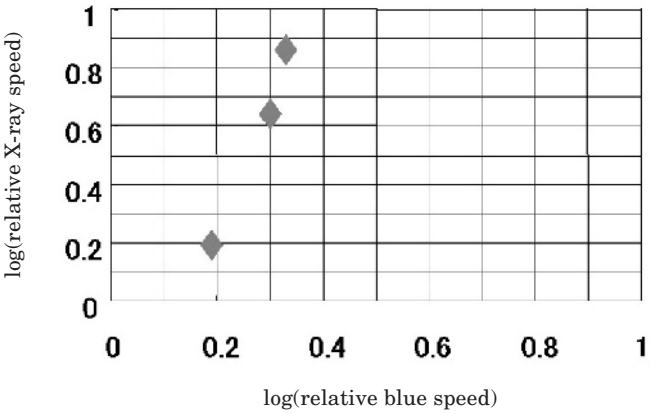

Figure 10. X-ray speed as a function of blue speed of AgBrI emulsions with variation of C_{Ag}.

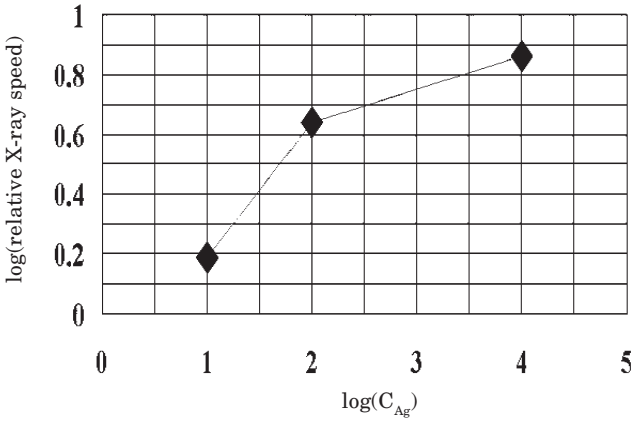

Figure 11. X-ray speed as a function of C_{Ag} of the emulsions described in Fig. 10.

brought about by the absorption of an x-ray quantum by a silver halide grain. The absorbed x-ray quantum gives a high energy secondary electron, which then creates many electrons in many surrounding grains. This is the reason why the x-ray sensitivity was more dependent on coated amount of silver halide than on grain size and added amount of dye. The absorbance of 120 kVp x-rays by an element is decided by the efficiency of the external photoelectric effect, which is proportional to the 5th power of an atomic number of the element. It was therefore considered that the absorbance of x-rays by an emulsion layer should be approximately proportional to the silver coating weight, C_{Ag}. This model led to the following equation,

$$S_\gamma = k_3 \, C_{Ag} \, S_i \qquad (4)$$

where k_3 is an empirical. Figure 13 was reproduced from the data shown in Figs, 10, 11, and 12. The abscissa indicates the sum of the logarithm of S_i and the logarithm of C_{Ag}, and the ordinate indicates logarithm of the x-ray sensitivity. This figure shows that the x-ray sensitivity of emulsion layers was proportional to C_{Ag} and to S_i in good accord with the model expressed by Eq. (4).

Then, we propose a second consideration as an explanation from another angle for the difference between the effects of C_{Ag} on x-ray and blue sensitivities, as shown in

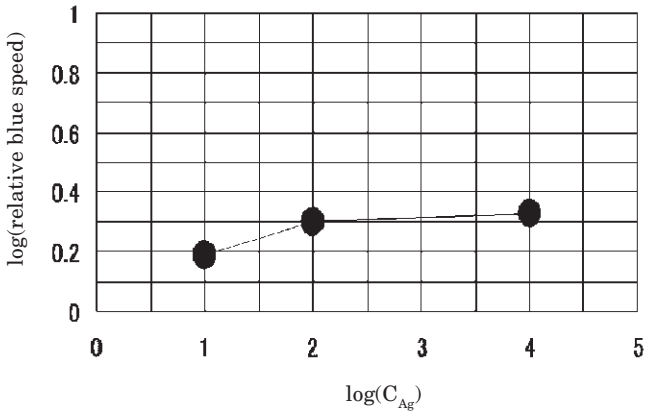

Figure 12. Blue speed as a function of C_{Ag} of the emulsions described in Fig. 10.

Figs. 11 and 12. In the case of blue light, the absorption of a photon by a silver halide grain brings about photochemical processes only within the grain that absorbed it. Therefore, the increase in C_{Ag} causes an increase in gradation, while it causes only a slight increase in sensitivity. In the case of high energy radiation, the absorp-

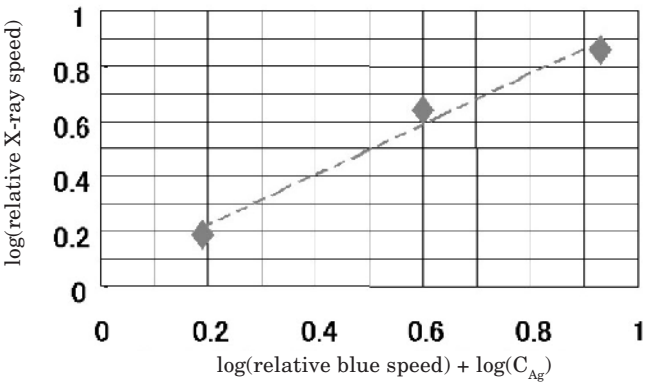

Figure 13. X-ray speed as a function of blue sensitivity and C_{Ag} of the emulsions described in Fig. 10.

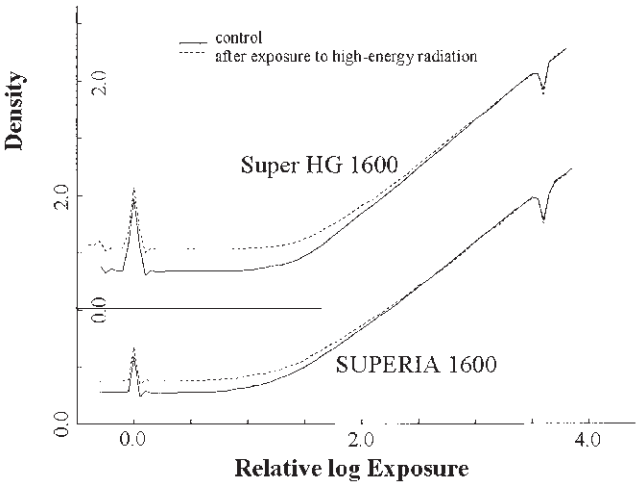

Figure 14. Changes in characteristic curves of red sensitive layers of a Fuji Color Super HG1600 film and a Fuji Color SUPERIA 1600 film caused by high energy radiation. Solid curves are control, and broken curves are given after their exposure to high energy radiation.

tion of a quantum by a grain brings about chemical processes not only in the grain that absorbs it, but also many surrounding grains. If the distance to which the interaction of the quantum extended was longer than the thickness of the emulsion layer, the number of grains that were made developable by a single quantum should increase in proportion to C_{Ag}. The x-ray sensitivity would therefore increase steeply with C_{Ag}. From calculation on the basis of the energy loss rate and/or the observed range for an electron to interact with an emulsion layer,[3,9] the distance to which the interaction of a high energy quantum extended was longer than the thickness of typical emulsion layer in our experiments.

Conclusions

High speed color negative films are inclined to suffer from increase in fog density during their storage. This damage is mostly caused by natural background radiation. We divided natural background radiation into three components: cosmic rays, environmental radiation, and self-contamination. We determined the fraction of the contribution of each component to the damage. The sensitivity of a silver halide photographic emulsion to gamma rays was in proportion to its intrinsic sensitivity to blue light and the amount of gamma rays absorbed by the emulsion grains.

Some Comments for the Future

We have developed a high speed color negative film, which is hardly influenced by natural background radiation on the basis of the knowledge obtained in this study. Specifically, our film was developed on the basis of the following design considerations.

1. The ratio of the intrinsic sensitivity to blue light to total sensitivity of our film was designed to be low by increasing the efficiency of spectral sensitization
2. The silver halide coating weight in our film was designed to be low in order to lower the rate of the absorption of radiation.
3. The self-contamination was designed to be small by reducing the amount of radioactive materials in our film.

Figure 14 shows the characteristic curves of Fuji Color SUPERIA 1600 produced in 2000 in comparison with those of Fuji Color SUPER HG1600 produced in 1990. This figure shows the comparison between them in terms of the increase in fog density of the red sensitive layer caused by natural background radiation for two years, as estimated by extrapolation of measured values. The technical progresses achieved for about ten years decreased the level of deterioration due to radiation. The damage to Fuji Color SUPERIA 1600 by natural background radiation is very small, as compared with traditional high speed films.

As for photographic materials, high sensitivity will continue to be valuable, making it possible to provide consumers with attractive new products. Therefore, we think that the technology to reduce the influence of natural background radiation on films is one of the most important basic technologies for photographic materials in the future.

Acknowledgments. The authors thank Prof. M. Sakanoue of Kanazawa University for permission to use the low-level radioactivity laboratory (LLRL) and for useful advice. And we thank S. Takada, S. Aida and S. Shibayama of Ashigara Research Laboratories, Fuji Photo Film Co., Ltd., for useful discussions.

References

1. Y. Suga, S. Aida, Y. Ishii, Y. Kume, H. Ikoma, and K. Ando, *Fuji Film Research and Development,* **46** (2001).
2. K. Yokota, H. Tamaoki, Y. Suga, Y. Nozawa, and Y. Kume, *J. Soc. Photogr. Sci. Tech. Japan,* **64,** 298 (2001).
3. R. H. Hertz, *The Photographic Action of Ionizing Radiations*, Wiley-Interscience, London, 1969.
4. D. P. Jones and R. H. Barber, *Photogr. Sci. Eng.* **8,** 212 (1964).
5. A. F. Sowinski and P. J. Wightman, *J. Imaging Sci.* **31,** 162 (1987).
6. M. Okano, K. Izumo, H. Kumagai, T. Katou, M. Nishida, T. Hamada, and M. Kodama, *Natural Radiation Environment III,* Symposium Series D0E51 (CONF-780422), 896 (1980).
7. M. Okano, K. Izumo, H. Kumagai, T. Katou, M. Nishida, and T. Hamada, in *Natural Radiation Environment,* by K. G. Vohra, Ed., Wiley Eastern Ltd., New Delhi, 1982, pp. 188–193.
8. S. Shibayama, S. Aida, T. Aizu, and S. Takada, US Patent No. 5,047,317 (1991).
9. P. Broadhead, in *The Theory of the Photographic Process,* 4th ed., T. H. James, Ed., Macmillan, New York, 1977, p. 646.