STUDY OF OPTIMAL CONDITIONS FOR CHEMICAL PROCESSING OF *AGFA PERSONAL MONITORING* PHOTOEMULSION WITH AN INCREASED FOGGING

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Claritatea imaginii la filmul Agfa personal monitoring, utilizat în monitorizarea personalului expus profesional la radiaţii ionizante, depinde de tipul emulsiei fotografice şi de calitatea prelucrării chimice a acesteia. În acest sens, în lucrarea de faţă s-au studiat condiţiile optime de prelucrare chimică a emulsiei fotografice Agfa pe patru loturi de filme cu următoarele densităţi optice de voal: 0.44; 1.06; 1.735; 1.84. S-a constatat că cele mai bune condiţii de desfăşurare a reacţiilor de reducere a ionului Ag^+ *din emulsiile cu voal crescut sunt temperatura* 19^0C *şi durata 2 minute.*

The image clarity of "Agfa personal monitoring" film utilised for monitoring the Romanian workers occupationally exposed to ionizing radiation depends on the type of photographic emulsion and quality of its chemical processing. The paper presents studies of optimal conditions for chemical processing of Agfa photoemulsion using four series of films with the following optical densities: 0.44; 1.06; 1.735; 1.84. It has been found that the best conditions for carrying out reduction reactions involving silver grains from emulsions with increased fogging are 19^0C *for bath temperature and 2 min for processing duration.*

Keywords: photo emulsion, film processing, developer, film fogging, irradiation dose

1. Introduction

The processing of photographic films subjected to light or ionizing radiations has been treated in numerous papers, the physical aspects of behaviour of different kinds of dosimetric films as a result of interaction between ionizing radiation and photoemulsion being extensively discussed [1-4]. Although the working principle for a photo dosemeter system as dosemeter probe involves the chemical effect of electromagnetic radiation incidence to photoemulsion [5], this chemical process was less treated in published papers.

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It is known that the photographic emulsion is generally a dispersion of silver halide (AgX) in colloidal medium (gelatine) in which centers of latent image are formed due to radiation incidence; as a consequence, in the bulk of emulsion AgX molecules convert in unstable species and Ag^+ and X^- ion pairs occur. Then, the latent image passes in a visible image by chemical reduction of Ag^+ ion to metallic Ag using a developer (solution containing a reductive agent). In practice, the degree of blackening is measured as optical density for the film exposed to radiation and subsequently developed by chemical or physical means. Obviously, the value of film optical density may be considered as a response of photoemulsion to certain amount and intensity of incident radiation.

The dimension of AgX grains dispersed in gelatine determines the sensibility of photographic film towards visible radiations and radiations having higher energy, including the penetrant radiations. However, due to the complexity of chemical composition of photographic emulsion, the existence of physical and/or chemical conditions, such as continuation of chemical maturation (so called "natural fogging" or "chemical fogging of emulsion"), storage conditions as well as further film processing, a series of other chemical processes are unavoidable.

By discussing the sensibility of photographic film we mention that it is involved in many chemical processes in a contradictory manner. For instance, the increase of silver halide concentration in emulsion leads to improvement of this parameter but also to increasing the film fogging by a natural ageing within a more short time. As a consequence, the standards of individual dosimetry recommend for dose meters that serve for monitoring the personal exposed to ionizing radiations to measure doses starting from 0.1 – 0.17 mSv [6]. Generally, the Agfa film used in our experiments allows measurements of doses starting from minimum level of 0.1 mSv, and sometimes even from minimum 0.05 mSv, obeying the condition of a film fogging between 0.2–0.43 units of optical density. The increase in time of film fogging leads to higher measuring errors as well as to a considerable decrease of photo dose meter sensitivity.

It is well known that the film sensibility depends in a large measure on its quality and on accurate chemical processing of image recorded after exposure. It is also important for this film to have a low level of fogging and also a very low rate of maturation during storage. Unfortunately, besides the centers of recorded image formed as a result of interaction between incident radiation and silver halide grains, other chemical processes that may have an influence to quantitative determination of absorbed doses take place. An example is the film fogging formed by continuation of the chemical maturation of emulsion, this meaning that a certain number of sensitivity centers may grow up to dimensions of developing centers. During film developing these centers may be recorded together to the centers of latent image and may generate a supplementary fogging, which is actually the chemical fogging of emulsion. For these reasons it is important to use a more selective developer in order to be able rather to chemically reducing the image centers formed by radiation-emulsion interaction and much less the fogging centers. Some chemical compounds such as KBr, methyl-oxy-triazo-indolisine, nitro-benzimidazole etc., added in developing solutions have an "anti-fogging" action for photographic film. However, the control or separation of simultaneous processes is very difficult. A study may be carried out by establishing optimum conditions in the film developing step, especially for time duration and temperature of developer containing solution. It is desired to have different rates of latent image developing and fogging grains developing [7] with a higher rate for the latent images occurred by irradiation. Generally, because the Ag grains formed by maturation have smaller dimensions comparing with grains resulted by radiation interaction, it is expected for these last grains to be first reduced.

The paper aims to analyse the conditions of developing processing of Agfa photographic films used for personal monitoring, which are films with increased values of fogging densities. We propose an evaluation of information obtained on increased fogging films by using the sensitometric curves (optical density *vs.* irradiation dose) of films with relatively low fogging. These sensitometric curves cover the irradiation dose interval from 0.1 mSv to 1000 mSv and were obtained experimentally for films with optical density within the range 0.37 - 0.78.

2. Experimental Part

The Agfa personal monitoring dosimetric film consists in two layers with different sensibilities of emulsion, separated by an absorbant paper strip. One layer (D10 film) has 1.5 μm grain size and a value of 0.2 for initial optical density, being very sensitive for irradiation dose range from 0.1 mSv to 70-100 mSv. The second layer (D2 film) has smaller granulation (0.2 μm grain size) and 0.17 value of initial optical density, being less sensitive and useful for greater irradiation doses, in the range from 50 mSv to 1000 mSv. The solutions used for film developing were G128–type developer bath and G328-type fixing bath provided by Agfa company. The recommended processing conditions were 5 minutes, pH=10.5 and 20 ^{0}C (for developing bath). In experiments, aqueous solutions of 2 w% acetic acid for developing stopper and 1 w% ethyl alcohol for pre-washing were also used. The final step of film processing consisted in continuous washing during 30 min.

For studying the influence of temperature and film immersion time in developing bath the densitometric method was applied; it consists in measuring the transmission optical density of 10 readings on each film. The equipment was the Gretag D200-II (Switzerland) densitometer with a measuring uncertainty from ±0.04 to ±0.10 depending on the optical density range.

3. Results and Discussion

For simplicity, we denote the optical density resulted by irreversible interaction between AgX grains with gelatine emulsion as “background fogging”, whereas the optical density due to maturation during storage will be denoted simply as “fogging”. For whatever chemical reaction in developer or fixing bath and temperature, the background fogging may not be removed; on contrary, the film fogging would be more diminished by choosing the optimal conditions.

We present here a study of six Agfa film series. The first two series were denoted L_A and L_B being stored in standard conditions and non-climatized rooms, respectively. The following four series (denoted as 1, 2, 3 and 4) have a gradual decrease of sensibility. For these last ones, the values of fogging optical densities, taken as an average of 5 films, were: 0.44 –for series 1, 1.06 –for series 2, 1.735 – for series 3, and 1.84 –for series 4. The standard deviations have values ± 0.006 for D10-type and ± 0.0001 for D2-type (with finer granulation), demonstrating a very good uniformity of silver grains dispersed in photographic emulsion. All the above values of optical densities will be considered as reference values, possible to be attained in the environmental conditions of intensely light, temperature higher than 20^0C or 3-5 months of carried dosemeter (although the change of dosemeter is recommended at every one month).

In order to study the fogging evolution during storage, L_A film series was kept at constant temperature of 20 ^{0}C and 40-45 % controlled humidity, whereas L_B film series was kept at increased temperature (up to 35 ^{0}C) and 32-35% humidity. The calibration of films was performed using Cs 137 radiation within the dose interval of 0.1 – 1000 mSv.

We noticed that films belonging to L_A series had a gradual increase in time of optical density, although they are stored in conditions indicated by Agfa company, *i.e.* 20 ^{0}C, 40-45 % atmospheric humidity and 90 nGy environmental radioactivity. Their fogging evolution in first 10 months of storage is presented in Table 1. The listed values were obtained by developing monthly 7 films (denoted from M1 to M7), with 10 readings of optical density for each film. We consider that a uniform dispersion of silver halide in gelatine as well as the quality of focused radiation determines both homogeneity and linearity of emulsion response as a consequence of interaction with electromagnetic radiation.

Table 1

Evolution of fogging optical density for Agfa films stored in standard conditions

Month	Film-type	Values of optical density for seven films stored at 20 ^{0}C and 40–45% humidity atmosphere							Average value ± standard deviation
		M1	M2	M3	M4	M5	M6	M7	
January	D10	0.26	0.25	0.26	0.26	0.26	0.27	0.26	0.26 ± 0.0058
	D2	0.16	0.16	0.16	0.16	0.16	0.16	0.16	0.16 ± 0
February	D10	0.25	0.27	0.28	0.28	0.28	0.29	0.27	0.27 ±0 .0127

	D2	0.15	0.16	0.16	0.16	0.16	0.16	0.16	0.16 ± 0.0038
Mars	D10	0.35	0.33	0.33	0.34	0.35	0.35	0.31	0.34 ± 0.015
	D2	0.17	0.16	0.16	0.16	0.16	0.17	0.16	0.16 ± 0.0049
April	D10	0.36	0.35	0.38	0.39	0.35	0.36	0.36	0.36 ± 0.0151
	D2	0.16	0.16	0.16	0.16	0.17	0.16	0.16	0.16 ± 0.0038
May	D10	0.38	0.37	0.37	0.37	0.41	0.42	0.40	0.39 ± 0.0212
	D2	0.16	0.17	0.16	0.16	0.16	0.17	0.17	0.16 ± 0.0054
June	D10	0.45	0.45	0.45	0.50	0.51	0.50	0.47	0.47 ± 0.0269
	D2	0.15	0.19	0.17	0.17	0.16	0.18	0.16	0.17 ± 0.0135
July	D10	0.46	0.46	0.47	0.47	0.49	0.46	0.46	0.47 ± 0.0111
	D2	0.16	0.16	0.16	0.16	0.16	0.17	0.17	0.16 ± 0.0049
August	D10	0.53	0.46	0.51	0.50	0.50	0.52	0.50	0.5 ± 0.0222
	D2	0.17	0.15	0.17	0.16	0.16	0.17	0.17	0.16 ± 0.0079
September	D10	0.53	0.50	0.53	0.54	0.54	0.56	0.56	0.54 ± 0.0205
	D2	0.16	0.17	0.17	0.18	0.16	0.16	0.16	0.17 ± 0.0079
October	D10	0.56	0.55	0.57	0.57	0.60	0.55	0.54	0.56 ± 0.0198
	D2	0.17	0.17	0.17	0.17	0.18	0.17	0.17	0.17 ± 0.0038

One can observe a decrease of film homogeneity for M1-M7 series during longer storage. The standard deviation has also an increasing tendency (up to ±0.027 for D10-type film and ±0.0135 for D2-type film), this fact being explained by a different growing rate of fogging centers in M1-M7 emulsions. In conditions of storage inside non-climatized rooms (case of L_B film series) this growing rate is enhanced, as Table 2 shows.

Table 2.

Evolution of fogging optical density for Agfa films stored in non-climatized conditions

Month	Film-type	Values of optical density for seven films stored at 26-35 ^{0}C and 32–35% humidity atmosphere							Average value ± standard deviation
		M1	M2	M3	M4	M5	M6	M7	
May	D10	0.48	0.48	0.53	0.52	0.53	0.59	0.59	0.53± 0.0453
	D2	0.16	0.17	0.17	0.17	0.17	0.17	0.17	0.17± 0.0038
June	D10	0.63	0.64	0.67	0.60	0.63	0.63	0.64	0.63±0.0207
	D2	0.17	0.16	0.17	0.17	0.17	0.17	0.16	0.17±0.0049
July	D10	0.82	0.83	0.83	0.80	0.78	0.83	0.86	0.82±0.0254
	D2	0.16	0.17	0.17	0.17	0.17	0.17	0.16	0.17±0.0049
September	D10	0.94	0.98	0.98	0.94	0.98	0.98	0.98	0.97±0.0195
	D2	0.16	0.16	0.17	0.17	0.17	0.17	0.17	0.17± 0.0049

Nevertheless, by analysing the data listed in Table 2 we did not observe a significant increase in evolution of fogging optical density during each month, suggesting that a temperature with 50 % over threshold of optimal conditions affects the fogging below 17 % during 30 days. This fact is in a good agreement with ISO standards [8] that limited this increase up to 20 % from corresponding point of sensitometric curve. However, by comparing with film fogging obtained for storage in optimal conditions the increase is significant, for instance it varies from 0.54 to 0.97 in Septembre. It worth also to mention the homogeneity of developing centers formed in a same extent for the whole film series with deviations for L_B films identical with corresponding values for L_A films.

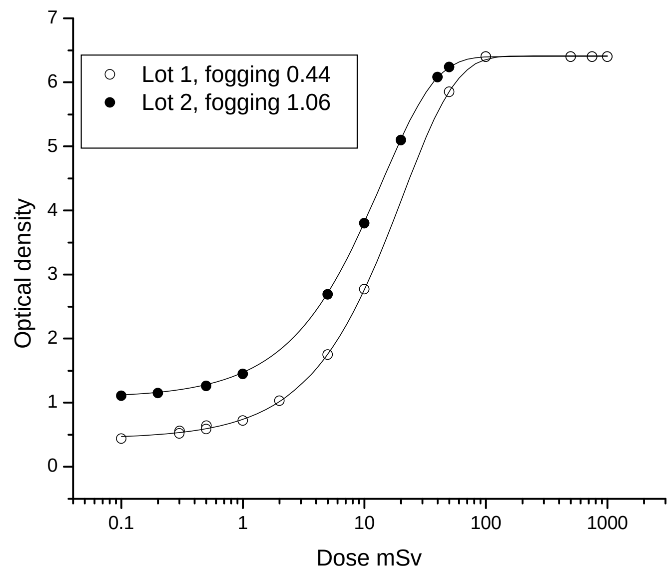

Fig 1. The densitometric curves showing the dependence of optical density vs. radiation dose for Agfa D10 and D2 films with 0.44 and 1.06 averaged fogging optical densities (batches 1 and 2).

Figure 1 shows the results of standard calibrations (sensitometric curve) for film lots 1 and 2, illustrating the importance of changing the film quality because the shape of sensitometric curve depends mainly on the fogging of entire film series. For instance, a deviation of *ca.* 300% for irradiation dose might be recorded if the optical density of value 2 measured with a film with 1.06 fogging is used for estimating the irradiation dose with another film, with 0.44 fogging.

During 10 months storage the film fogging increases significantly even by keeping the optimal conditions, examples being values from 0.25 to 0.56 for more sensitive film (D10-type film in Table 1). Thus, the increase of fogging optical density comparing to values determined during standard calibration measurements of films leads to significant errors in measuring the equivalent dose. Nevertheless, we determined irradiation doses of 0.1 mSv for a film fogging value of 0.56, with an error up to 15 %.

In cases of film series (lots) 3 and 4, we observed that the calibration step is not sufficient because the film behaviour to radiations changed considerably. In these cases the dependences optical density *vs.* irradiation dose cannot be useful for determination the absorbed dose employing films with such high fogging values. An example is shown in Figure 2 where the optical density increased in the dose range from 0.1 mSv to 0.5 mSv and then decreased.

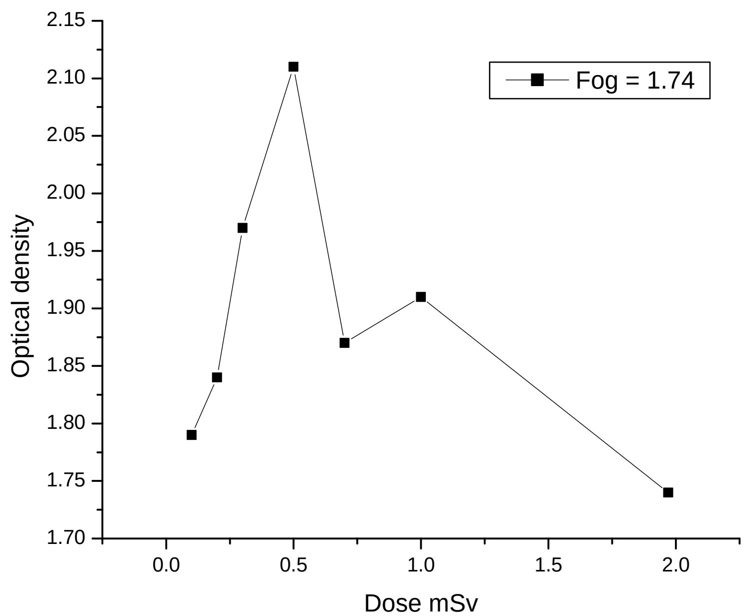

Fig.2 The densitometric curve for the film series having averaged fogging optical density of 1.74

The diminution of fogging optical densities was performed by lowering both temperature and developing time during film processing. Table 3 shows the experimental results illustrating the separation between fogging developing and background fogging.

Table 3

Variation of fogging optical density for Agfa D10-type films developed in various conditions of bath processing

Bath temperature [°C]	Developing time [min]	Fogging optical density			
		Film series 1	Film series 2	Film series 3	Film series 4
17	2	0.31 ± 0.0071	0.69 ± 0.01155	1.22 ± 0.01729	1.15 ± 0.0029
	5	0.35 ± 0.0048	0.84 ± 0.0115	1.40 ± 0.01647	1.38 ± 0.0185
19	2	0.33 ± 0.0048	0.63 ± 0.0058	1.07 ± 0.0183	1.08 ± 0.0067
	3	0.32 ± 0.0044	0.79 ± 0.0057	1.23 ± 0.0151	1.51 ± 0.0122
20	2	0.33 ± 0.0085	0.65 ± 0.0059	1.13 ± 0.0286	1.12 ± 0.0063
	3	0.35 ± 0.0096	0.72 ± 0.0058	1.21 ± 0.0070	1.24 ± 0.0135
	4	0.41 ± 0.0056	0.91 ± 0.01	1.57 ± 0.011	1.64 ± 0.007
	5	0.44 ± 0.00516	1.06 ± 0.01	1.73 ± 0.0017	1.84 ± 0.0216
	6	0.46 ± 0.0084	-	-	-

We noticed a smaller influence of developing temperature for reducing process of Ag grains inside developing centers formed by natural ageing of photoemulsion. In contrast, during the processing time a considerable diminution of film fogging was produced. On the basis on these observations, a temperature of 19 ^{0}C and 2 min processing time were selected as optimal conditions for developing films with increased fogging.

In case of film series 1 the fogging optical density remains constant even for temperatures below 20 ^{0}C and 5 min processing time. For longer processing times a linear increase of optical density was recorded.

In case of film series 2 we obtained a significant decrease of film fogging, namely, from a value of 1.06 in conditions considered optimally, until a minimum value of 0.63 at 19 ^{0}C and 2 min processing time.

In cases of film series 3 and 4, the fogging optical density decreases up to values of 1.07 and 1.08, respectively.

Other experimental observations reveal a uniform developing for the whole film surface, with a standard deviation below 0.01. As Figure 3 illustrates, by film processing with too short developing time a non-uniform developing was produced.

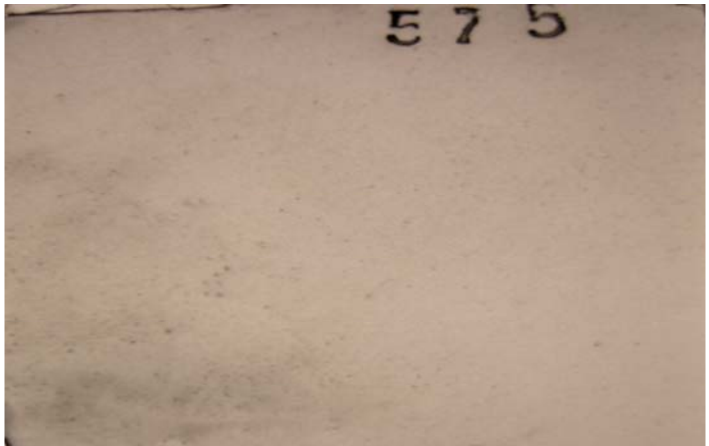

Fig. 3. The image of an Agfa D10 film developed for 1 min at 20 ^{0}C bath temperature

For the D2-type film having a finer granulation, the fogging is less influenced upon developing conditions, in comparison with less sensitive D10-type film. However, we consider that optimal conditions selected for D10-type film, regarding operating parameters like bath temperature and developing time, remain also valid for photoemulsion of D2-type film. During determination of irradiation doses using D2 film, a value of ± 0.01 deviation in optical density leads to a dose measuring error of 15.53 % for 20 mSv dose and 9.54% for 50 mSv dose, respectively. We notice that irradiation doses up to 50 mSv are usually detected using D10-type film.

Table 4

Variation of fogging optical density for Agfa D2-type films in various conditions of developing

Bath tempe-rature [°C]	Develo-ping time [min]	Fogging optical density			
		Film series 1	Film series 2	Film series 3	Film series 4
17	2	0.15 ± 0.00	0.15 ± 0.00	0.15 ± 0.00	0.15 ± 0.00
	5	0.16 ± 0.00	0.16 ± 0.00	0.16 ± 0.00	0.16 ± 0.00
19	2	0.16 ± 0.0048	0.16 ± 0.00	0.16 ± 0.00	0.16 ± 0.0042
	3	0.16 ± 0.0042	0.16 ± 0.00	0.16 ± 0.00	0.16 ± 0.00
20	2	0.165 ± 0.0048	0.16 ± 0.00	0.17 ± 0.00	0.17 ± 0.00
	3	0.16 ± 0.0032	0.16 ± 0.00	0.18 ± 0.00	0.18 ± 0.00
	4	-	0.17 ± 0.00	0.18 ± 0.00	0.18 ± 0.00
	5	0.16 ± 0.0042	0.17 ± 0.00	0.18 ± 0.00	0.18 ± 0.00
	6	0.16 ± 0.00	-	-	-

3. Conclusions

The experimental results show clearly that the photographic response to radiations of *Agfa personal monitoring film* is not affected by temperature during one month of utilisation in the range 20-35 ^{0}C. It is expected an increase of this influence by working only at temperatures higher than 50 ^{0}C and atmospheric humidity greater than 60%, such conditions being possible met in some industrial cases. The results obtained using the film series 1 are the most relevant and allowed us to determine irradiation doses with good accuracy. In case of film series 2 the fogging was decreased in a large extent so that an estimation of equivalent dose employing one of available sensitometric curves become possible. Whatever the developing duration and bath temperature, the own fogging of films belonging to series 3 and 4 does not decrease at a limit which may allow the estimation of absorbed irradiation dose using available densitometric curves determined in our laboratory by film calibration. It is also necessary a calibration for films with fogging value of 1.08 ± 20%, because using this quality of film it is possible to estimate low irradiation doses.

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