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*Acta Cryst.* (1963). **16**, 1107

## A Second Comparison of Various Commercially Available X-ray Films\*

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Various properties of commercially available X-ray films were tested for the second time, with an improved technique. (For the first tests, see *Acta Cryst.* (1956), **9**, 520.) The tested films were of forty-three types, made by eighteen manufacturers. The tested properties were: speeds for Cu and Mo radiation; fog density; granularity; absorption of Ag, Mo, Br, Cu, Fe and Cr  $K\alpha$  radiation;  $D-E$  characteristics; film homogeneity; sensitivity for safety lights; aging effect, etc. The results are tabulated and the properties of the films are briefly discussed.

### 1. Introduction

Some years ago, a comparison of 41 commercially available X-ray films was carried out under the sponsorship of the Commission on Crystallographic Apparatus of the International Union of Crystallography (1956) at the University of Groningen by Dr D. W. Smits and Prof. E. H. Wiebenga. The aim of the investigation was to obtain reliable information about the relative merits of various types of X-ray film. As since the time of this comparison new types of X-ray film had appeared and the properties of the investigated types might have been changed, the Commission decided at its Conference in Stockholm (1959) that the investigation be repeated. In addition it was felt desirable that some improvements in the experimental technique be made and some additional properties be included in the new comparison.

Two of the more important improvements made here were the following:

1. Fluorescent Cu and Mo radiations, which are almost pure  $K\alpha$  lines, were used instead of filtered radiations from Cu and Mo targets.

2. The film speeds were measured in an absolute scale (in  $\mu^2/\text{photon}$ ) instead of a relative one.

Forty-three different types of X-ray film made by eighteen manufacturers in ten countries were collected.‡ All the films were received between September 1960 and May 1961, and were tested during the period March 1961 to February 1962.

### 2. Experimental

#### (a) Experimental arrangement

The experimental arrangement is schematically shown in Fig. 1. It was constructed by the Rigaku-Denki Co. Ltd.§ to a special order.

The input to the X-ray generator was first stabilized by a 3 kVA electronic stabilizer *St*. The high voltage was of full-wave rectified and smoothed by a condenser *C* of 0.2  $\mu\text{F}$ , the tube current usually being

\* Reprints of this article can be obtained from the authors, or from the General Secretary of the International Union of Crystallography, Dr D. W. Smits, Mathematisch Instituut, University of Groningen, Reitdiepskade 4, Groningen, The Netherlands.

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‡ Films made in the U.S.S.R. were not available in the present investigation. Some of their data are given by БОГОМОЛОВ *et al.* (1961).

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20 mA. The input of the high-voltage rectifier *HT* and that of the filament transformer *FT* were stabilized by the feedback of the tube voltage and of the tube current respectively. The fluctuation of X-ray intensity in an hour was less than 1 or 2%, as measured with the monitor scintillation counter *MC*.

The radiation from the tungsten anode *A* excited the fluorescent radiation at the source *S*. The tube

whereas those in the back plate were covered with a black paper. The rotating sector\* of the shape shown in Fig. 2 was set in front of the film holder. The opening angles of the sector were designed to be  $15^\circ$ ,  $30^\circ$ ,  $45^\circ \dots 300^\circ$ , and they were checked by interrupting an X-ray beam of constant intensity. The error was found to be less than 0.5% even at small angles.

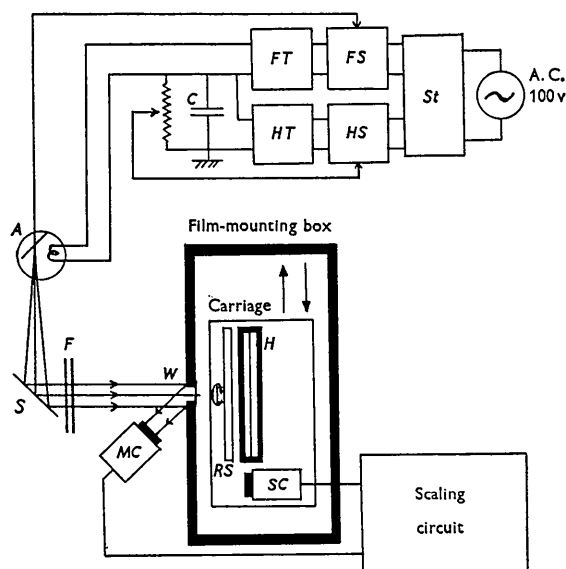


Fig. 1. Schematic diagram of the experimental arrangement.

voltage was 25 kV for the Cu source and 40 kV for the Mo source. The radiation from the Cu source was filtered by 0.02 mm nickel, and that from the Mo source by 0.1 mm zirconium and 0.27 mm aluminum. In both cases, the count for the continuous spectrum was less than 5% of that of the characteristic  $K\alpha$  radiation (*cf.* Appendix I).

The radiation from the source was transmitted by the filter *F* to a dark box through a window *W* (10 mm  $\times$  30 mm) which was covered with 0.01 mm aluminum foil. In the box, a film holder *H*, a rotating sector *RS* and a standard scintillation counter *SC* were mounted on a carriage. The distance between the source *S* and the film holder *H* was 40 cm. The absolute intensity of the radiation was measured by placing the carriage in such a position so that the incoming radiation fell directly on the standard counter. Since this measurement was possible only at the beginning and the end of the exposure, the constancy of the intensity was always checked by the monitor counter.

The film holder consisted of two brass plates between which a film of about 18 cm  $\times$  13 cm was mounted. Both plates were provided with circular and band openings (Fig. 2). All the openings in the front plate were kept uncovered during the exposures,

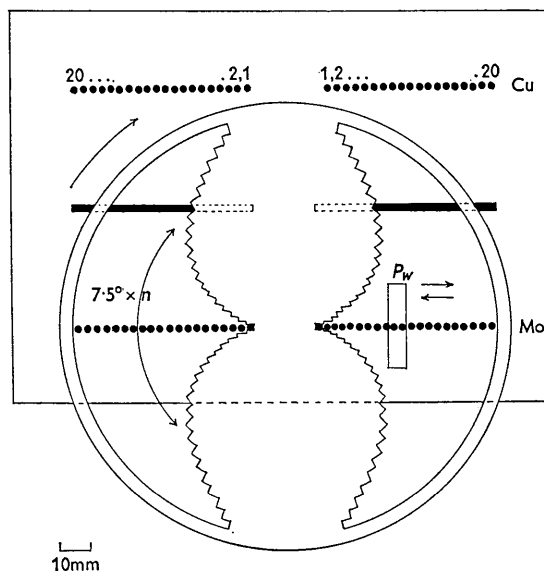


Fig. 2. Rotating sector and film holder in the position for the exposure of Mo scales. *P<sub>w</sub>* indicates the projection of the window in the dark box on the film.

### (b) Intensity scales

On each film, the following intensity scales were printed:

1. A set of two identical intensity scales made with Cu radiation, each scale consisting of 20 circular spots with a diameter of 2 mm, of which the exposures formed an arithmetical series.
2. A similar set of two intensity scales made with Mo radiation.
3. Two uniformly exposed bands made with Cu radiation, each of 2 mm width and 60 mm length.

To obtain uniform exposure, the sector was set at the largest opening and the carriage was repeatedly moved with a uniform speed of  $1.33 \text{ mm}\cdot\text{sec}^{-1}$  by a synchronous motor as indicated in Fig. 1. Thus the film and the sector were moved together. For the intensity scales the carriage was moved in a similar way, and in addition the sector was rotated at 600 rpm. The number of runs of the carriage and the intensity of the X-rays were adjusted in such a way

\* Made by Mr S. Takahashi in the workshop of the Faculty of Science, Nagoya University, Nagoya, Japan.

Table 1(a). *Details of films used*

No.	Manufacturer	Country	Type of film*	Developer	Period A† (months)	Period B† (months)	$w_{Ag}$ (mg.cm <sup>-2</sup> )
1	Adox	Germany	Neotest	Adox Röntgen-Rapid-	11		2.8
2			Mikrotest	Entwickler	11		2.8
3	Agfa-Leverkusen	Germany	Röntgen-Sino	Agfa-Leverkusen Röntgen-Rapid-Entwickler			3.2
4	Agfa-Wolfen	Germany	Laue-film	Agfa-Wolfen Röntgen-Rapid-Entwickler	10		1.4
5	Anseo	U.S.A.	Non Screen	§		8	3.1
6	Ceaverken	Sweden	Type-XH		8	7	2.0
7			Type-XM	§	14	7	1.7
8			Type-XL		17	9	1.5
9	Dupont	U.S.A.	†Type 504	Kodak Rapid X-ray		7	1.3
10			†Type 506	Developer		7	1.7
11			†Type 510			8	1.9
12	Eastman Kodak	U.S.A.	Type AA	Kodak Rapid X-ray		8	2.1
13			†Type M	Developer		6	1.9
14			†No Screen			7	4.0
15	Ferrania	Italy	Tipo I			11	1.3
16			Tipo IC	Ferrania RX-41		10	1.8
17			Tipo ID			7	3.3
18	Fotochem.- Berlin	Germany	Diasinex	§	7		2.6
19	Fuji	Japan	†Type 200			10	3.0
20			Type 100	Fuji Rendol		7	2.5
21			Type 80			5	2.6
22			Type FG (single-coated)			7	0.8
23	Gevaert	Belgium	†Structurix D10				3.2
24			†Structurix D7			7	2.1
25			Structurix D4	Gevaert G209A		7	1.3
26			Structurix D2			7	1.4
27			†Structurix S			7	1.2
28	Iford	U.K.	†Industrial B		10		2.6
29			†Industrial C	Iford Phen-X	10		2.7
30			Industrial CX		16		3.0
31			†Industrial G		16		2.9
32	Kodak	Australia	Industrial	Kodak Rapid X-ray Developer			3.0
33	Kodak	France	Kodirex	Radio Kodalk		1	2.6
34	Kodak	U.K.	†Crystallex			6	2.5
35			†Industrex D	Kodak DA-19b		6	2.6
36			†Kodirex			2	2.6
37	Konishiroku	Japan	Sakura N			10	3.5
38			Sakura RR	Sakura Konidol X		6	2.6
39			Sakura R			7	3.0
40	Perutz	Germany	†Type K	Perutz Röntgen-Entwickler			1.0
41	Typon	Switzerland	Osteo			7	2.1
42			†Progress	Typon E-7		12	1.3
43			Ferix			8	1.6

\* Base: Nos. 1, 2, 4, 22 and 40 are colourless; all the others are blue. — Package: Nos. 1, 2, 3, 6, 7, 8, 15, 16, 17, 18, 33, 34, 35, 36 and 41, each sheet kept in an envelope; all the others packed with interleaving paper. — Recommended developing time: No. 18, 8 min.; Nos. 41, 42 and 43, 6 min.; all the others, 5 min. — Recommended developing temperature: Nos. 15, 16, 17 and 18, 18 °C; all the others, 20 °C.

† Period A indicates the period from the time of film manufacturing to that of the speed measurement; B, from the time of the speed measurement to the deadline for development.

‡ The same type as tested in the previous investigation (*Acta Cryst.* (1956), 9, 520).

§ Kodak D-19b was used, since no special developer was recommended.

Table 1(b). *Properties of films*

No.	Speed for X-rays			Ageing† (%)	$G_s(\mu)$	Fog†	Film factor $F$
	$S (\mu^2/\text{photon})$						
	Cu	Cu*	Mo				
1	1.77	(1.84)	0.93	0	4.3	0.10+0.00	2.83
2	0.16	(0.14)	0.09	5	1.0	0.10+0.05	2.89
3	1.84	(2.08)	0.89	15	4.3	0.25+0.00	3.20
4	0.38	(0.40)	0.16	15	2.4	0.10+0.00	1.95
5	1.44	(1.38)	0.72	5	3.4	0.20+0.10	2.90
6	0.90	(0.90)	0.40	10	2.4	0.15+0.05	2.29
7	0.64	(0.64)	0.30	5	2.2	0.15+0.05	2.18
8	0.26	(0.26)	0.12	15	1.6	0.20+0.05	2.11
9	0.50	(0.52)	0.21	0	2.0	0.15+0.05	1.82
10	0.32	(0.25)	0.14	0	1.7	0.05+0.00	2.02
11	0.11	(0.08)	0.05	10	0.83	0.05+0.00	2.21
12	0.26	(0.24)	0.13	5	1.1	0.05+0.05	2.32
13	0.10	(0.07)	0.05	15	0.96	0.00+0.00	2.14
14	1.59	(1.36)	0.82	10	2.7	0.20+0.05	3.89
15	0.11	(0.07)	0.05	20	1.2	0.05+0.00	1.94
16	0.46	(0.44)	0.20	10	1.8	0.20+0.05	2.18
17	1.84	(1.46)	0.91	20	3.8	0.35+0.10	3.25
18	1.22	(1.07)	0.60	0	2.9	0.40+ > 0.3	2.71
19	1.68	(1.54)	0.86	15	2.8	0.20+0.15	2.96
20	0.25	(0.22)	0.13	15	1.3	0.05+0.00	2.63
21	0.16	(0.13)	0.08	15	1.1	0.00+0.00	2.64
22	0.04	(0.03)	0.02	15	0.86	0.00+0.00	1.43
23	1.44	(1.52)	0.81	0	3.0	0.30+0.10	3.33
24	0.29	(0.31)	0.15	5	1.4	0.05+0.05	2.36
25	0.08	(0.07)	0.04	5	0.72	0.00+0.00	1.92
26	0.02	(0.02)	0.01	20	0.43	0.00+0.00	1.95
27	0.51	(0.60)	0.21	10	2.3	0.20+0.10	1.71
28	0.36	(0.36)	0.20	10	1.3	0.10+0.00	2.73
29	0.12	(0.11)	0.07	10	1.1	0.05+0.00	2.84
30	0.18	(0.16)	0.12	15	0.85	0.05+0.00	3.19
31	1.08	(1.14)	0.54	0	2.3	0.15+0.00	2.93
32	1.49	(1.29)	0.73	15	3.0	0.15+0.10	3.21
33	1.14	(1.02)	0.56	0	3.2	0.30+0.10	2.64
34	0.29	(0.28)	0.15	15	1.2	0.10+0.05	2.47
35	0.51	(0.49)	0.25	10	1.8	0.10+0.00	2.62
36	1.38	(1.38)	0.64	15	2.9	0.15+0.05	2.89
37	2.6	(2.34)	1.34	15	3.5	0.15+0.10	3.40
38	0.30	(0.25)	0.16	5	1.1	0.05+0.05	2.75
39	0.16	(0.13)	0.08	10	0.80	0.05+0.00	3.03
40	0.25	(0.19)	0.09	30	1.8	0.05+0.00	1.69
41	1.36	(1.17)	0.58	25	3.1	0.20+0.05	2.24
42	0.73	(0.64)	0.30	10	2.1	0.20+0.05	1.86
43	0.39	(0.32)	0.17	5	1.6	0.10+0.05	2.10

\* Developed with Kodak D-19b, 20 °C, 5 min.

† Decrease of speed after a half-year aging (§ 3(c)). Fog density and fog increase after a half-year aging (§ 3(e)).

that the photographic density amounted to  $1.0 \pm 0.1$  for the uniform exposures, and to more than 2.0 for the darkest spot of the intensity scales. The number of runs was two or three for the fastest films and several hundreds for the slowest ones.

### (c) *Absolute measurement of X-ray intensity*

The scintillator of the standard scintillation counter was Harshaw's 2 mm  $\times$  1'  $\varphi$  sodium iodide crystal covered with 0.1 mm beryllium foil; the photomultiplier was a DuMont 6291. Taking into account the absorption in the foil, the counting efficiency of

Table I(b) (cont.)

No.	Characteristic curve										$1/D_{\max}$
	Relative exposure required for $D_{\text{eff}}$										
	0.2	0.4	0.6	0.8	1.0	1.2	1.4	1.6	1.8	2.0	
1	19	39	59	79	100	122	146	170	195	219	0.15
2	20	40	59	80	100	121	142	164	187	211	0.09
3	18	37	57	78	100	124	148	174	202	230	0.21
4	22	43	62	81	100	120	140	162	190	221	0.12
5	17	36	56	78	100	124	149	176	205	237	0.24
6	17	36	55	77	100	125	153	184	218	255	0.30
7	18	37	57	78	100	123	148	176	207	240	0.24
8	18	37	57	78	100	124	148	173	200	227	0.20
9	16	34	54	76	100	127	158	193	232	280	0.35
10	20	40	60	80	100	122	145	170	195	222	0.16
11	20	40	60	80	100	121	141	164	187	211	0.09
12	19	38	58	79	100	122	146	170	197	224	0.17
13	20	40	60	80	100	121	141	163	186	210	0.08
14	19	38	59	79	100	122	144	166	190	214	0.12
15	20	40	60	80	100	121	144	168	193	219	0.14
16	18	37	57	78	100	124	150	177	205	236	0.23
17	20	40	59	80	100	121	143	166	192	218	0.13
18	17	36	56	78	100	124	149	175	203	232	0.22
19	18	37	57	78	100	122	146	170	196	222	0.17
20	20	40	60	80	100	120	140	160	180	200	0.00
21	19	39	59	80	100	121	142	163	185	207	0.06
22	19	38	58	78	100	123	147	174	203	236	0.22
23	19	38	58	78	100	122	146	171	198	226	0.18
24	19	38	58	78	100	123	146	172	200	230	0.20
25	20	40	60	80	100	121	142	164	187	211	0.09
26	20	39	59	79	100	121	144	166	190	214	0.11
27	17	35	55	76	100	127	158	193	233	272	0.35
28	20	40	59	80	100	121	143	166	190	215	0.12
29	20	40	60	80	100	121	143	165	189	214	0.11
30	19	38	58	79	100	122	145	169	194	220	0.15
31	19	38	58	79	100	122	145	170	194	218	0.15
32	19	39	59	79	100	122	144	167	191	216	0.13
33	19	39	59	79	100	122	145	169	194	220	0.15
34	19	38	58	79	100	122	144	168	193	218	0.14
35	19	38	58	79	100	122	144	168	193	218	0.14
36	20	40	60	80	100	121	142	166	189	214	0.11
37	18	37	57	78	100	124	150	178	210	246	0.26
38	20	40	59	79	100	121	143	166	189	212	0.10
39	20	40	60	80	100	121	141	163	185	209	0.07
40	19	39	59	79	100	123	148	176	208	244	0.25
41	19	38	58	78	100	122	146	172	200	229	0.20
42	15	32	52	74	100	129	162	200	246	297	0.39
43	20	39	59	79	100	122	146	171	198	228	0.19

the arrangement was assumed to be 97.5% for  $\text{Cu}K\alpha$  and 99.4% for  $\text{Mo}K\alpha$  radiation because the absorption in the sodium iodide crystal should be 100% for both radiations (Taylor & Parrish, 1955; Parrish & Kohler, 1956). The resolution time was limited to 4  $\mu\text{sec}$  by the scaling circuit. Therefore the observed number of counts was corrected at this rate, the correction always being less than 5%. The noise pulses caused in the photomultiplier and the circuit

were eliminated by a pulse height discriminator, provided that the scintillator crystal was not damaged. The intensity of the X-rays was of the order of 10,000 photons  $\text{mm}^{-2}\cdot\text{sec}^{-1}$  for both Cu and Mo radiation.

With the aim of checking the absolute measurement by the scintillation counter, the measurement by an ionization chamber was also carried out. The difference between both measurements was about 15% for Cu radiation and about 30% for Mo radiation.

Table 1(c). *Safety factors*

No.	Safety factor for filtered lights*			
	Y-50	O-56	R-60	R-64
1	<0.5	0.5	1.2	2
2	<0.5	1.5	3	6
3	<0.5	3	11	(60)
4	<0.5	8	23	(45)
5	<0.5	6	23 (20)	(45)
6	<0.5	4	14	24 (20)
7	<0.5	4.5	(50)	(100)
8	<0.5	2	5	9
9	<0.5	4.5	20	40 (26)
10	<0.5	(40)	(260)	(900)
11	0.5	(320)	(1000)	(>2000)
12	<0.5	8	(20)	(110)
13	<0.5	(50)	(180)	(1000)
14	<0.5	6	50	(100)
15	<0.5	20	(60)	(130)
16	<0.5	1.5	5	50 (30)
17	<0.5	3	20	(50)
18	<0.5	0.8	2	3
19	0.6	15	36 (36)	(120)
20	<0.5	17	(80)	(270)
21	0.5	(60)	(160)	(650)
22	<0.5	1.5	8	18
23	<0.5	15	(50)	(120)
24	0.5	(30)	(120)	(210)
25	<0.5	17	50 (50)	(100)
26	0.5	30	(90)	(230)
27	<0.5	1.5	6	12
28	<0.5	0.5	1	3
29	<0.5	1.8	3	8
30	<0.5	1.7	3	8
31	<0.5	0.5	0.8	3
32	<0.5	18	(30)	(130)
33	<0.5	0.6	3	8
34	0.6	20 (18)	(60)	(200)
35	<0.5	8	50 (20)	(50)
36	<0.5	11	50	(30)
37	0.9	18	(80)	(200)
38	<0.5	3	12	24
39	<0.5	9	20	30 (26)
40	<0.5	3	9	18
41	<0.5	5	18	40 (26)
42	<0.5	2.7	9	20
43	<0.5	2.3	8	14

\* Values in parentheses are obtained with lamp B, the other values with lamp A (§ 3(h)).

However, these values depend very much on the assumed values of the absorption coefficient of air. For further details, see Appendix II.

#### (d) Processing

Processing was carried out in a thermostat tank, the films being hung vertically so as to make the rows of spots horizontal. The temperature of the

developer was adjusted to the prescribed one within  $\pm 0.2$  °C. During development the films were moved up and down gently. After development the films were rinsed in distilled water for about 30 sec, fixed in an acid fixing bath for 10 min, rinsed in running tap water for an hour, and dried in the air at room temperature.

The developers used were those recommended for each film by its manufacturer (Table 1(a)). Kodak D-19b was also used for all films for the measurement of the speed for Cu and Mo radiation. The developing temperature and time for each recommended developer were those recommended by the manufacturer (see footnotes of Table 1 (a)); and those for D-19b were 20 °C and 5 min.

#### (e) Density measurement

The density was measured relative to the reference strip of each type. The strip was made from an unexposed film by processing it under the same conditions but without development. The density is defined by  $D = \log_{10}(i_0/i)$ , where  $i_0$  and  $i$  are the intensities of a light beam transmitted by the reference strip and the test film respectively. The difference between the measured density and the fog density  $D_F$  is called the effective density  $D_{eff}$ .

The microdensitometer\* used is schematically shown

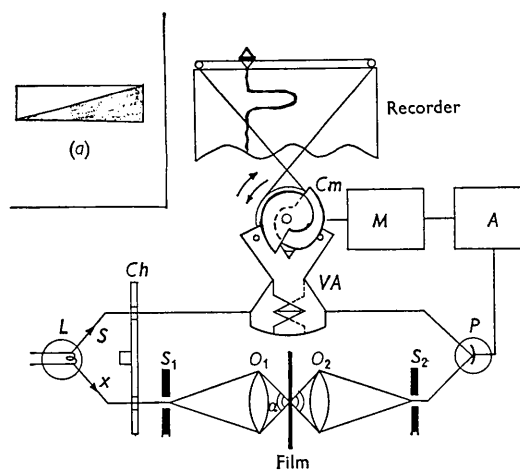


Fig. 3. Schematic illustration of the microdensitometer and the standard optical wedge (a).

in Fig. 3. The light from a lamp L was divided into two beams s and x which were alternately interrupted by a chopper Ch. The image of the first slit  $S_1$  was made by a microscopic objective lens  $O_1$  on the film to be measured. An identical objective lens  $O_2$  gave an enlarged image of the illuminated film at the

\* Made by Nalumi Co. Ltd., 4-23, Hongo, Bunkyo-ku, Tokyo, Japan.

second slit  $S_2$ . The area illuminated on the film was  $0.05 \times 0.05$  mm for the granularity measurements, and  $0.3 \times 0.3$  mm for the other measurements. The aperture angle of illumination  $\alpha$  was always  $30^\circ$ .

The area of the beam  $s$  was limited by a variable aperture  $VA$ , which was automatically controlled by a feedback loop consisting of a photomultiplier  $P$ , an amplifier  $A$ , a balancing motor  $M$  and a cam  $Cm$  so that the intensities of the two beams  $s$  and  $x$  as received by the photomultiplier were balanced. The cam had such a curvature that the rotating angle was directly proportional to the density. The rotating angle was recorded on a recorder.

The scale of the recorder was calibrated by means of a standard optical wedge, and it was found that the error was within 2%. The standard optical wedge consisted of two uniform triangular prisms (Fig. 3(a)), one transparent and the other neutral gray. The density was measured at two positions by a method which was based on the inverse-square law, and the linearity was assumed to exist between these two positions.

(f) *Chemical determination of the amount of silver in the films*

The determination of the amount of silver in the films was carried out by a chelatometric titration with ethylenediamine-tetra-acetic acid (EDTA) (Sjöstedt & Gringras, 1957).

A sample of the film ( $3 \times 3$  cm) was placed in a beaker containing 100 ml of diluted hydrochloric acid. After about 15 min, when the calcium present in the gelatine layer had been removed by the acid, the film sample was washed twice with distilled water. Then 10 ml of a buffer solution and 5 ml of concentrated aqueous ammonia were added to the beaker containing the film sample, followed by about 0.1 g potassium tetracyanonickelate. The buffer solution was made by dissolving 54 g ammonium chloride in water mixed with 350 ml of concentrated aqueous ammonia (sp.gr. 0.880), and diluting the solution with water up to 1 liter. When the silver halide had dissolved and the film sample had become transparent, the solution was diluted to 50 ml and was titrated with 0.01 molar EDTA solution with murexide as indicator. 1 ml of 0.01 molar EDTA solution corresponds to 2.16 mg of silver. The amount of silver per  $\text{cm}^2$  film,  $w_{Ag}$ , was calculated, and the results are listed in Table 1. To obtain the amount of silver per  $\text{cm}^2$  of the emulsion of the double coated films, the figures given in Table 1 should be divided by 2.

### 3. Results

The results have been summarized in Table 1. The period from the time of manufacturing of films to that of speed measurement and the period from the

time of speed measurement to the deadline for development are also listed, since the photographic properties of films depend on their age.

All the experiments except the absorption measurements were carried out between March 1961 and July 1961, before the deadline for development for each film. The fog and the speed for Cu radiation were measured again in November 1961 and December 1961 after the films had been kept for about half a year in a desiccator at room temperature (which rose to about  $35^\circ\text{C}$  during the summer). The fog increase and speed decrease due to aging were calculated by comparing the results of these two tests. It should be noted, however, that films are ordinarily stored under better conditions, e.g. at lower temperatures, sometimes even in a refrigerator, and that in these circumstances the aging effects may be expected to be less serious. This was not checked in the present investigation.

(a) *Coating, thickness and weight*

All films tested were double-coated except Fuji FG. The double-coated films had a total thickness of  $210\text{--}280 \mu$  ( $30\text{--}40 \text{ mg.cm}^{-2}$ ), consisting of  $15\text{--}30 \mu$  ( $2\text{--}6 \text{ mg.cm}^{-2}$ ) emulsion on each side and  $170\text{--}230 \mu$  ( $22\text{--}28 \text{ mg.cm}^{-2}$ ) film base. Fuji FG, the single-coated film, had a total thickness of  $165 \mu$  ( $21 \text{ mg.cm}^{-2}$ ), consisting of  $15 \mu$  emulsion,  $140 \mu$  ( $18 \text{ mg.cm}^{-2}$ ) base and  $10 \mu$  halation-preventive coating. The amount of silver  $w_{Ag}$  in the films was  $0.8\text{--}4.0 \text{ mg.cm}^{-2}$ .

(b) *Homogeneity*

The film homogeneity was determined from microdensitometer traces of the uniform bands as follows. Each of the two bands on a film was divided into ten regions, a total of 20 regions. The mean density was determined in 10 regions taken alternately from the 20 regions. The values of the mean density fluctuated from one region to another because of the inhomogeneity of the films. The root-mean-square deviation was normally less than 2%, but in some cases it amounted to 6%. The observed maximum deviation was nearly four times the root-mean-square deviation. The present result is almost the same as that of the previous comparison (Commission on Crystallographic Apparatus of the International Union of Crystallography, 1956). Since the deviations observed were probably not characteristic of the type of film, they have not been included in Table 1.

(c) *Speeds for X-rays*

The film speed  $S$  has been defined as the reciprocal of the number of X-ray photons per  $\mu^2$  required to produce an effective density  $D_{\text{eff}}=1.0$ . The film speed data listed in Table 1(b) are the averages of two or more experimental results obtained with the

recommended developing. The fluctuation in the experimental results was normally less than 5%. This fluctuation is not very large because the root-mean-square deviation from homogeneity was of the order of 2%.

The speed of most films decreased by 10–20% after the half-year aging. For some films, however, the decrease was hardly perceptible, but for others it amounted to 30%. The difference may be due not only to the type of film but also to the age of the film, *i.e.*, the period from the time of manufacturing to that of testing. It may be worth while to note that the decrease of speed before the deadline for development may be larger than the decrease observed here, although on the other hand the effect might be less if the films were stored under better conditions. In any case it is advisable to use films as fresh as possible.

The speed obtained with the D-19b developer was considerably different from that with the recommended developer. The speed differences were distributed between *ca.* -40% and +20%. The differences for Cu and Mo radiation agreed within about 5%. Therefore only the results for Cu radiation are listed in Table 1.

#### (d) Granularity

The film granularity was determined in the following way. The density was measured at 100 points, 0.06 mm apart, of the uniform bands. The measured values fluctuated from one point to another owing to the graininess of the film and also because of the film inhomogeneity. To eliminate the latter effect, the 100 points were divided into 10 groups of 10 successive points, and for each point the deviation from the mean value of its group was taken. The root-mean-square of the 100 deviations was assumed to represent the granularity  $G$  of the film.

Selwyn granularity  $G_s$  is defined as (Selwyn, 1935)

$$G_s = G/(2s) \quad (1)$$

where  $s$  represents the illuminating area, which was  $0.05 \times 0.05$  mm in the present experiment. Values of  $G_s$  calculated by this equation are listed in Table 1(b) for all types of film. It is known that  $G_s$  is almost independent of  $s$ , provided that  $s$  is not smaller than  $(40\mu)^2$  (Mees, 1954).

The value of  $G_s$  depends on the density. Since the density of the uniform band was not exactly constant for the different films ( $D_{\text{eff}} = 0.9 \sim 1.1$ ), the values of  $G_s$  listed in Table 1(b) may include relative errors of about 10%. It is evident from Table 1 that in general the granularity increases with the film speed. For further details, see § 4(d).

#### (e) Fog density

The fog density measured between March 1961 and July 1961, and the fog increase due to the half-year

aging, are listed in Table 1(b). The figures are rounded off at 0.05 as this is about the error in these measurements. Since the fog density depends on the age of the film and on the conditions under which the film has been stored, the values listed give only a semiquantitative indication. It is generally observed that the fog density and its increase due to aging are remarkably high for fast films.

#### (f) Absorption of X-rays

The film factor,  $F = I_0/I_t$ , where  $I_0$  and  $I_t$  are the incident and the transmitted intensity, respectively, was measured for crystal-reflected Cu  $K\alpha$  radiation. Since the absorption is caused by scattering absorption as well as by true absorption, the measurement was carried out at two positions as shown in Fig. 4. In Table 1(b), values measured at position 1 are listed. The standard deviation of these data is approximately 2%. Within this error, no difference was found between position 1 and 2.

The absorption was measured at position 1 also for Ag, Mo, Br, Fe and Cr  $K\alpha$  radiation. The results obtained for all types of film are plotted in Fig. 5

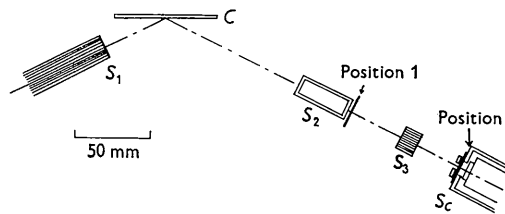


Fig. 4. Arrangement for the absorption measurements.

$C$ : LiF monochromatic crystal.

$S_1$ ,  $S_2$  and  $S_3$ : Soller slits; plates of  $S_2$  and  $S_3$  are normal to each other.

$Sc$ : Scintillation counter.

against the amount of silver in the films. The latter figure is useful for the estimation of the speed for various radiations (§ 4(c)). The data in Table 1(b) and Fig. 5 refer to films without envelopes or interleaving papers; the absorption of a sheet of paper amounted to 15–40% for Cr, 10–25% for Fe, 3–15% for Cu, 1–4% for Br and less than 1% for Mo and Ag  $K\alpha$  radiation.

#### (g) Characteristic curves

The characteristic curves were obtained from the intensity scales. The relative exposures  $E$  required to obtain an effective density  $D_{\text{eff}}$  of 0.2, 0.4 ... 2.0 are listed in Table 1(b). For each film the relative exposure is normalized to 100 at  $D_{\text{eff}} = 1$ . The data listed refer to Cu radiation; the data for Mo radiation are similar to these.

The characteristic curves could be represented by an empirical formula

$$D_{\text{eff}} = D_{\text{max}} \{1 - \exp(-\alpha E)\} \quad (2)$$



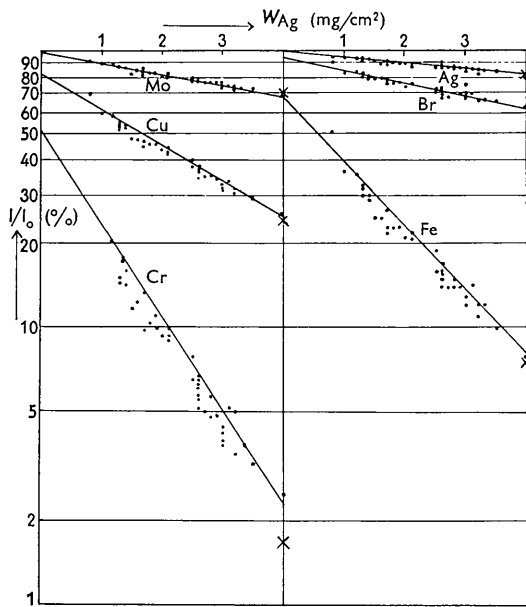


Fig. 5. Transparency of X-rays plotted against the amount of silver. Crosses indicate the data by Taylor & Parrish (1956). Straight lines are calculated, by assuming that silver is contained as silver bromide; bases and gelatine about  $25 \text{ mg.cm}^{-2}$ , consisting of 40% C, 10% N, 40% O and 10% H.

where  $D_{\max}$  is the only parameter, because  $\kappa$  is determined by the normalization

$$D_{\max}\{1 - \exp(-\kappa \cdot 100)\} = 1.$$

Equation (2) is a very good approximation, provided that the value of  $1/D_{\max}$  is properly determined. The root-mean-square deviation of the observed values from those calculated by (2) was less than 3% for all types of film with only one exception (Laue-Film from Agfa-Wolfen). Therefore, values of  $1/D_{\max}$  are given in Table 1(b). Within the limit of experimental errors, equation (2) could be applied also to Mo radiation with the same values for  $1/D_{\max}$ . It may be worth adding that the deviation from the linearity of the  $D$ - $E$  curve is about  $1/(2D_{\max})$  at  $D_{\text{eff}}=1$ .

#### (h) Safety factor for filtered lights

When an X-ray film is exposed to a filtered light, the exposure time which produces a density  $D_{\text{eff}}=0.1$  may be given as

$$t_{0.1} = k_x(r^2/l \cdot T_x) \quad (3)$$

where

- $r$  is the distance between the light source and the film;
- $l$  is the luminosity of the light source;
- $T_x$  is the mean transparency of the filter for wavelengths longer than the cut-off wavelength;
- $x$  indicates the type of filter;
- $k_x$  is the characteristic constant of the film.

In the present report  $k_x$  is called the safety factor for filtered light.

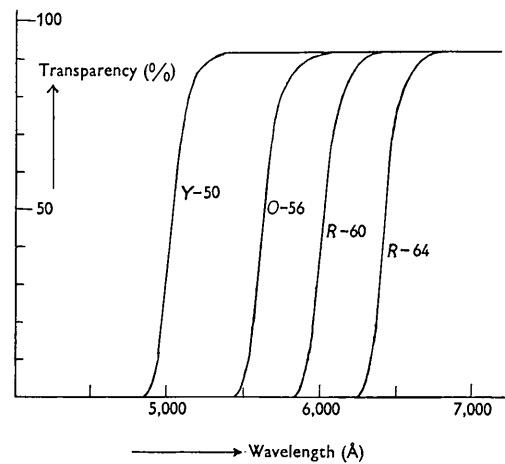


Fig. 6. Transparency of filters.

Four filters Y-50, O-56, R-60 and R-64, made by the Hoya Glass Works,\* were used. The transparency of these filters is shown in Fig. 6. A lamp  $A$  of colour temperature  $2500^\circ\text{K}$  and luminosity  $6 \text{ cd}$  and a lamp  $B$  of  $2800^\circ\text{K}$  and  $260 \text{ cd}$  were used as light sources. The distance  $r$  was  $60 \text{ cm}$ . The exposure time was varied from 3 to 300 sec and  $t_{0.1}$  was determined by interpolation. The safety factors for the four types of filters are given in Table 1(c). The values obtained with the lamps  $A$  and  $B$  are not the same because of the difference in colour temperature and the failure of the reciprocity law.

The measurements were made by using strong illumination to save time; in practice much weaker lights are normally used in dark rooms. When the failure of the reciprocity law is taken into consideration, the values of the safety factor expected for weak illuminations may be larger than those given in Table 1.

In ordinary laboratory dark rooms the distance  $r$  is about  $100 \text{ cm}$  and luminosity  $l$  is about  $10 \text{ cd}$ .† For example, if a red filter of 20% transparency is used for film No. 3, the value  $k_{R-60}=11$  may be taken (Table 1(c)), and  $t_{0.1}$  can be calculated to be 550 sec. Therefore, the fog produced is less than  $D_{\text{eff}}=0.1$  if the exposure time to the safety light in the dark room is less than 550 sec.

## 4. Discussion

### (a) Comparison with the previous investigation

Seventeen types of film marked by an asterisk in Table 1(a) were also tested in the investigation

\* 2-1 Kyobashi, Chuo-ku, Tokyo, Japan.

† According to Japanese Industrial Standard C-7501, 10 and 20 W lamps are  $5 \sim 7$  and  $12 \sim 15 \text{ cd}$ , respectively. The colour temperature is about  $2500^\circ\text{K}$  for both.

Table 2. Comparison of results of previous (I) and present (II) investigations

Film no.	Normalized ratios II/I				$1/D_{\max}$			Fog density	
	Speed		$G_s$	$F$	I	II	II-I	I	II
	Cu	Mo							
9	0.86	1.06	1.2	1.02	0.35	0.35	0.00	Medium	0.15
10	1.30	1.50	1.3	—	0.12	0.16	+0.04	Very low	0.05
11	0.99	0.86	1.0	0.96	0.00	0.09	+0.09	Very low	0.05
13	0.90	1.07	(1.5)	0.99	0.00	0.08	+0.08	Very low	0.00
14	1.43	1.28	1.1	1.00	0.05	0.12	+0.07	Low	0.20
19	0.75	0.74	1.1	0.93	0.23	0.17	-0.06	High	0.20
23	1.35	1.45	1.0	0.97	0.24	0.18	-0.06	Medium	0.30
24	1.00	1.07	0.8	1.00	0.14	0.20	+0.06	Low	0.05
27	0.76	0.69	0.9	0.86	0.34	0.35	+0.01	Medium	0.20
28	0.85	0.78	0.8	1.02	0.08	0.12	+0.04	Medium	0.10
29	0.67	0.75	0.9	1.20	0.00	0.11	+0.11	Very low	0.05
31	0.81	0.84	0.9	1.06	0.14	0.15	+0.01	Medium	0.15
34	0.93	0.92	0.9	0.93	0.16	0.14	-0.02	Very low	0.10
35	0.64	0.67	0.8	0.96	0.10	0.14	+0.04	Medium	0.10
36	1.63	1.71	1.2	1.09	0.21	0.11	-0.10	Medium	0.15
40	0.80	0.77	1.1	—	0.39	0.25	-0.14	Medium	0.05
42	1.17	1.17	1.0	1.04	0.34	0.39	+0.05	Medium	0.20

reported in 1956. The results obtained for these films in the previous (I) and present (II) investigation are compared in Table 2. For the comparison of speeds, the present speeds of 2.2 for Cu and 1.2 for Mo radiation were normalized to 100 because the above two values seem to correspond to the previous value of 100 on the average. Ratios (II/I) are listed in Table 2 for the film speeds for Cu and Mo radiation, the granularity and the film factor. In the next three columns the values for  $1/D_{\max}$  are given which were derived from the  $D-E$  characteristic curves found in the two investigations; the differences between these values are also given. In the last two columns the results found for the fog density are compared.

In Table 2, differences between the results of the two investigations seem to be appreciable. However, before interpreting the results, it should be emphasized that photographic properties of a film depend not only on the film properties but also on the developer, the developing conditions and the nature of the illuminating X-rays. The age and history of a film also change the photographic properties. Therefore any comparison of films should be made under strictly controlled conditions. The present investigation and the previous one were both carried out under such conditions. However, in the two investigations the conditions were not same. Therefore, the difference in results due to different conditions must be studied before any conclusion is drawn.

For example, the variation of speed due to different developers is described in § 3(c). A variation of the order of 30% takes place for different developers. Therefore the variation of this order seen in Table 2 may be possible when the developer is changed. It is known that the value of  $1/D_{\max}$  depends on the developer and developing conditions. Takagi, Kitamura & Morimoto (1961) observed in the case

of electron irradiation that the value of  $1/D_{\max}$  of the same plate was 0.0 for developer D-72 while about 0.3 for D-131. The similar change may be expected also for X-ray films. The value of  $G_s$  depends not only on the developer but also on the value of  $D_{\text{eff}}$  at which the measurement is done. It should be noted here that the values of  $D_{\text{eff}}$  were 0.6-1.0 in investigation I but 0.9-1.1 in II. It is clear that the fog density depends much on the effective age of films. The result for fog density in Table 2 seems to show rather the difference in the effective age. The film factor is the only quantity which is not influenced by the developer or developing conditions. In this case, crystal-reflected X-rays were used in both investigations. Thus the change of values may indicate the real change.

Finally, it seems highly probable that the film properties have been changed for some films. However, it may not be adequate to attribute the variation observed in Table 2 only to the change of film properties.

#### (b) Absolute speed

The absolute speed of X-ray films was measured by Seemann (1950) for No Screen and Type K of Eastman Kodak. His result for the former is almost one half of the present result. This disagreement may be due mainly to the different optical geometry used for the density measurements: Seemann measured the diffuse density whereas in the present investigation the nearly specular density was obtained. The diffuse density refers to true absorption of light in the film, the specular density to the total absorption, *i.e.* the true absorption plus the scattering absorption. The diffuse density is always smaller than the specular density.

## (c) Speed for various X-ray wavelengths

The speed  $S$  for X-rays may be given as the product of the absorption of X-rays in the film,  $A = (I_0 - I_t)/I_0$ , and the photographic efficiency  $P$  of the absorbed X-rays:

$$S(\lambda) = A(\lambda) \cdot P(\lambda) \quad (4)$$

where both factors  $A$  and  $P$  depend on the wavelength. The first factor, the absorption  $A$ , is expected to have large jumps at the Ag and Br absorption edges, and the ratio  $A_{Cr K\alpha}/A_{Ag K\alpha}$  even amounts to 10 (Fig. 5). The absorption for various wavelengths can be estimated from the amount of silver in the film ( $w_{Ag}$ ), or from the interpolation or extrapolation of the data in Fig. 5. The second factor, the efficiency  $P$ , varies only a little between Ag  $K\alpha$  and Cr  $K\alpha$  (Seemann, 1950); the ratio  $P_{Ag K\alpha}/P_{Cr K\alpha}$  was almost 2 and the jump at the Br absorption edge was small. In the present investigation,  $P_{Mo K\alpha}/P_{Cu K\alpha}$  was found to be only 1.2 ~ 2.0. Therefore, the efficiency  $P$  for various wavelengths may be obtained by interpolation or extrapolation of the values of  $P_{Cu K\alpha}$  and  $P_{Mo K\alpha}$  which can be obtained from the data of Table 1 and Fig. 5. As a rough estimation, the efficiency  $P$  may even be assumed as a constant, independent of the wavelength.

In the present investigation, the speed data were determined only for Cu and Mo radiation. However, both the absorption  $A(\lambda)$  and the efficiency  $P(\lambda)$  can be estimated as mentioned above. Therefore the speeds can be estimated for various wavelengths by using equation (4).

## (d) Relation between speed and granularity

As mentioned in § 3(d), the granularity increases

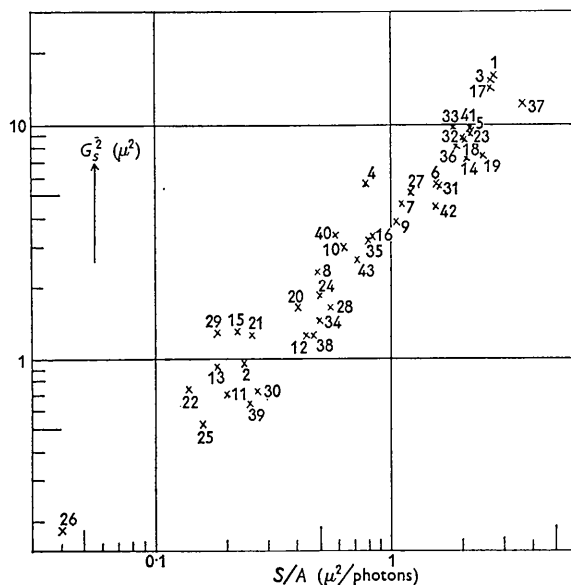


Fig. 7.  $G_s^2$  plotted against  $S/A$ .

with the film speed for X-rays. In Fig. 7 the square of the Selwyn granularity  $G_s$  is plotted against the speed  $S$  divided by the absorption  $A$  (*i.e.* against the efficiency  $P$ ). There is an empirical relation:

$$G_s^2 = 3.6S/A. \quad (5)$$

If this formula could be extrapolated beyond the region given in Fig. 7, it would be possible to prepare faster films by increasing  $G$  and/or  $A$ .

## (e) Grain yield per photon

According to Savelli (1959), the following relation exists between the Selwyn granularity  $G_s$  and the areal density  $d_g$  of the silver grains:

$$G_s^2 = 4.4/d_g \quad (6)$$

at  $D_{eff} = 1$ , under the assumption that the developed silver grains are spherical and are distributed at random (Picinbono, 1955). Substitution of equation (6) into equation (5) gives

$$d_g = 1.2A/S. \quad (7)$$

This implies that one absorbed photon produces 1.2 grains, which agrees with other experimental results (Mees, 1954). In the case of visible light, the grain yield per photon is much smaller than 1. This large difference in grain yield is known as the cause of various different photographic properties for X-rays and visible light (Mees, 1954).

## APPENDIX I

## Monochromaticity of the radiations

The spectral distribution of the filtered fluorescent radiations used in the present investigation was analysed with a crystal spectrometer. The radiation was collimated by a slit system of  $1^\circ$  divergence, analysed by a LiF crystal and counted by a scintillation counter. The angular region measured with the LiF crystal was  $8.5^\circ < 2\theta < 77^\circ$  ( $0.3 \text{ \AA} < \lambda < 2.5 \text{ \AA}$ ) for the Mo radiation and  $14^\circ < 2\theta < 77^\circ$  ( $0.5 \text{ \AA} < \lambda < 2.5 \text{ \AA}$ ) for the Cu radiation.

The Cu radiation used showed only the first order reflection of  $K\alpha$  in the angular width  $44^\circ < 2\theta < 46^\circ$ ; the Mo radiation showed the first, second and third order reflection of  $K\alpha$  in the angular widths  $19.5^\circ < 2\theta < 21.5^\circ$ ,  $40^\circ < 2\theta < 42^\circ$  and  $63^\circ < 2\theta < 65^\circ$ , respectively. The count integrated over the whole measured region except the reflection width or widths was less than 5% of the count integrated in the reflection width or widths. The count outside the reflection width or widths was partly caused by fluorescent and scattered radiations from the crystal holder, *etc.* Therefore, the count caused by the white radiation

was less than 5%. This figure did not change when the count was corrected to the intensity by taking into account the wavelength dependence of the reflecting efficiency of the crystal, etc.

## APPENDIX II

### Absolute measurement of the X-ray intensity by an ionization chamber

The construction of the ionization chamber is shown in Fig. 8. The nearly monochromatic X-rays from the fluorescent source were introduced into the chamber, and the ionization current  $J$  was measured by an electronic micro-microammeter. The homogeneity of the electric field in the chamber was checked by connecting the outer case  $C$  to the plus or minus potential. The saturation of the ionization current was obtained in a field of about  $10 \text{ V.cm}^{-1}$ .

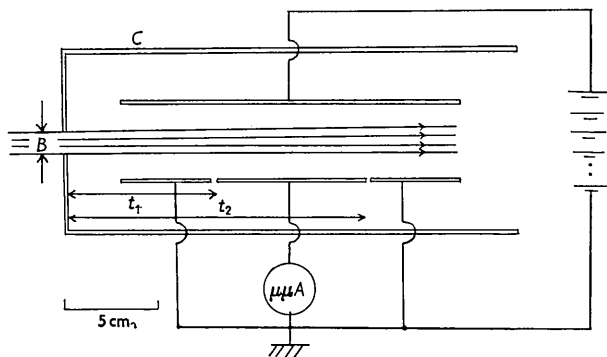


Fig. 8. Schematic diagram of the ionization chamber.

The absolute X-ray intensity  $n_i$ , i.e. number of photons/(time.area) in the front of the chamber, was estimated by the equation

$$J = e \cdot n_i \cdot B [\exp(-\mu t_1) - \exp(-\mu t_2)] v_p / v_i. \quad (\text{II } 1)$$

Here

$e$  is the charge of the electron;

$B$  is the size of the X-ray beam in the front of the chamber;

$t_1$  and  $t_2$  are geometrical parameters (Fig. 8);

$v_p$  is the energy of an X-ray photon;

$v_i$  is the energy required to produce a pair of ions;

$\mu$  is the absorption coefficient of X-rays in air, being equal to  $\sum_i (\mu_i / \rho_i) w_i$  where  $i$  indicates the constituent elements,  $(\mu_i / \rho_i)$  the mass absorption coefficient, and  $w_i$  the mass of the  $i$ th element per  $\text{cm}^3$ .

The geometrical parameters were:  $B = 10 \times 30 \text{ mm}$ ,  $t_1 = 8 \text{ cm}$  and  $t_2 = 16 \text{ cm}$ . The distribution of the X-ray

intensity was checked photographically and it was found to be not perfectly uniform. The deviation from the mean value was a few per cent and this was corrected in the effective value of  $B$ . The values of  $v_p$  were 8,010 eV for Cu radiation and 17,400 eV for Mo radiation;  $v_i$  was assumed to be 34 eV (Marton, 1959). The value of  $\mu$  was calculated with the mass absorption coefficients given by Bragg (1949) under the experimental conditions:  $16^\circ \text{C}$ , 760 mm Hg, 55% humidity. The calculated values are listed in Table 3.

Table 3. Calculated absorption coefficients

	Cu	Mo
(Bragg)	0.0132	0.00163
Present	0.85	0.70
K. & P.	0.96	0.46
(K. & P.)	0.010	0.00098
Present	1.11	1.17
K. & P.	1.26	0.76

The measured value of  $J$  was about  $20 \mu\mu\text{A}$  for Cu radiation and about  $6 \mu\mu\text{A}$  for Mo radiation. The value of  $n_i$  calculated by equation (II 1) was about 10,000 photons. $\text{sec}^{-1}\text{mm}^{-2}$ ) for both radiations. The intensity of X-rays at the position of the window of the ionization chamber was also measured by a scintillation counter with a window of 1 mm  $\phi$ . The number of photons thus measured,  $n_s$  photons/(time.area), was compared with the value  $n_i$ . The ratios  $n_i/n_s$  for Cu and Mo radiation are given in the second line of Table 3.

A similar experiment was previously carried out by Kohler & Parrish (1956). If their result is converted to the ratio  $n_i/n_s$ , it turns out as given in the third line of Table 3. It should be noted that the values of absorption coefficient used above do not seem to be very reliable. Kohler & Parrish used in their report the absorption coefficients as given in the fourth line of Table 3. If their values are used in the calculation of  $n_i$ , the ratios become as given in the fifth and sixth lines of Table 3. The cause of the difference between the present result and the result of Kohler & Parrish is not clear.

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## Self-Crystallizing Molecular Models

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It is shown that the structure of a molecular crystal corresponds to a minimum of electrostatic energy between the molecules when the molecules have simple shapes and sufficiently strong electric multipoles. In these cases the crystal structure can be reproduced with molecular models, which are made of magnets so as to simulate magnetically the electric forces between actual molecules. When their shapes and magnetization are adequately patterned after given molecules, the 'crystals', into which these models are assembled, show the crystal structures actually given by the molecules. Photographs of several types of such 'crystals' are shown.

The purpose of this paper is to show that the structure of a molecular crystal is governed by electrostatic forces between the molecules when the molecules have simple shapes and sufficiently strong electric multipoles. Molecular models devised by the author (Kihara, 1960) are used for this purpose.

The models are made of magnets so as to simulate magnetically the electric forces between actual molecules. When their shapes and magnetization are adequately patterned after given molecules, the 'crystals', into which these models are assembled,

show the crystal structures actually given by the molecules.

An approximate choice of such molecular models is the following (Fig. 1):

Type I. Fifteen octupolar spheres, each of which is composed of eight magnets in the symmetry  $\bar{4}3m$  ( $T_d$ ).

Type II. Fourteen quadrupolar spheres, each of which is composed of two hemispherical magnets.

Type III. Fourteen quadrupolar spindle-shaped models.