estimation, where peak densities are compared. For a single layer of photosensitive grains, the total photographic action is proportional to sec ψ, but is now spread over an area that increases in the same ratio, and therefore the action per unit area is independent of ψ, so that $D_2 \equiv 1$. For a multiplicity of layers, the screening of the lower layers must be taken into account, and

$$(D_2)_{\text{visual}} = \left(\frac{\text{Action}}{\text{Area}}\right)_0 \Big/ \left(\frac{\text{Action}}{\text{Area}}\right)_\psi > 1\ ,$$

in contrast to the factor for integrated intensities. The thickness of the emulsion will be negligible compared with the linear dimensions of the spots, and if we assume that for the film base, $t \tan \psi$ is very much less than d, the spot diameter, it follows that the area of the spot is simply proportional to sec ψ, and from equation (1),

$$\begin{aligned}(D_2)^{t=0}_{\text{visual}} &= \frac{\sec\psi(1-\exp[-A])}{1-\exp[-A\sec\psi]} \times \frac{1+\exp[-(A+B)]}{1+\exp[-(A+B)\sec\psi]} \qquad (5)\\ &= 1+(\sec\psi-1)[(\tfrac{1}{2}A+\varphi)+(\sec\psi-1)\varphi(\varphi-\tfrac{1}{2}B)-\\ &\qquad \tfrac{1}{12}A^2(2-\sec\psi)+\ldots]\ ,\end{aligned}$$

where

$$\begin{aligned}\varphi = \frac{(A+B)\exp[-(A+B)]}{1+\exp[-(A+B)]} &= 0{\cdot}35(A+B)\pm 0{\cdot}02 \quad \text{for} \quad A+B<\tfrac{2}{3}\ ,\\ &= 0{\cdot}25\pm 0{\cdot}03 \quad \text{for} \quad \tfrac{2}{3}<A+B<2{\cdot}3\ .\end{aligned}$$

The second and third terms inside the square brackets together produce an error of less than 0·02 in D_2. To this accuracy, we obtain

$$(D_2)^{t=0}_{\text{visual}} = 1+(\sec\psi-1)(\tfrac{1}{2}A+\varphi)\ . \qquad (5a)$$

We can also estimate the change in formula (5) when the condition $t\tan\psi \ll d$ does not hold. Clearly, the image in the back layer of emulsion is displaced through a distance $t \tan \psi$ with respect to the stronger image in the front layer. As a reasonable estimate of the density distribution for a spot, we take the Gaussian form, $p \exp[-10r^2/d^2]$, so that the density falls to $\frac{1}{12}$ of the peak value at a distance $\frac{1}{2}d$ from the peak. It is not difficult to show that the resultant peak density for the composite spot is approximately

$$\begin{aligned}&p(1+\exp[-(A+B)\sec\psi])\\ &\times \exp\left[-\frac{10t^2\tan^2\psi}{d^2\sec^2\psi}\right.\\ &\qquad\left.\times\left\{\frac{\exp[-\frac{1}{2}(A+B)\sec\psi]}{1+\exp\left[-(A+B)\sec\psi\left(1-\frac{5t^2\tan^2\psi}{d^2\sec^2\psi}\right)\right]}\right\}^2\right]\\ &\simeq p(1+\exp[-(A+B)\sec\psi])\\ &\qquad\qquad \times \exp[-10(t^2/5d^2)\sin^2\psi] \quad (A+B<\tfrac{3}{2})\ ,\end{aligned}$$

so that

$$\begin{aligned}(D_2)^t_{\text{visual}} &\simeq \exp[10(t^2/5d^2)\sin^2\psi](D_2)^{t=0}_{\text{visual}} \qquad (6)\\ &\simeq \exp[10(t^2/5d^2)\sin^2\psi](1+(\sec\psi-1)(\tfrac{1}{2}A+\varphi))\ .\end{aligned}$$

With typical values of $t/d = \frac{1}{4}$, $A = \frac{1}{3}$, $\varphi = \frac{1}{4}$, the first factor is less important than the second. Typical curves for $(D_2)^t_{\text{visual}}$ and $(D_2)^{t=0}_{\text{visual}}$ are plotted in Fig. 1 for comparison with the other curves. The correction factors for visual work are seen to deviate very markedly from those for integrated densities. The effect of a non-linear density–exposure relationship will still be given by equation (4) if $(D_2)_{\text{corr.}}$ is replaced by $(D_2)_{\text{visual}}$; this will make the experimental $(D_2)_{\text{visual}}$ greater still.

Film-to-film factor for multiple-film exposures

Invariant over the foregoing mathematical manipulations is the following property of the absorption:

With the exponential law, the intensity factor from one film to the next in a multiple-film stack at normal incidence is

$$K_0 = \exp[2A+B]\ ,$$

and therefore, for incidence at an angle ψ, it is

$$K_\psi = \exp[(2A+B)\sec\psi] = (K_0)^{\sec\psi}\ , \qquad (7)$$

which is a useful result since K_0 is known accurately. The effect of the progressively decreasing radius for successive films is small and produces a negligible error in formula (7).

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The unit-cell dimensions and space groups of some tetrazole compounds.

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The investigation of tetrazole compounds in this laboratory has made available a large number of tetrazole derivatives. Some of these substances which formed good crystals have been investigated by X-ray single-crystal methods with the view of finding one or more suitable for a complete structure determination. The desirability of obtaining direct proof of the existence of the tetrazole ring in these compounds and of the dimensions of this ring is obvious. Table 1 lists the results from six compounds examined. The most satisfactory crystals were all derivatives of 5-aminotetrazole. These compounds were obtained from Drs Ronald A. Henry, William G. Finne-

Table 1.

Compound	Crystal system	a_0 (Å)	b_0 (Å)	c_0 (Å)	β	N	Space group	Density (g.cm.$^{-3}$) obs.	calc.
5-Aminotetrazole monohydrate	Monoclinic	6·39	7·28	9·80	90° 15′	4	$P2_1/c$	1·51	1·504
Guanidine salt of 5-aminotetrazole	Orthorhombic	11·94	7·05	7·05	—	4	Pba	—	1·201
5-(N-Methyl)aminotetrazole	Monoclinic	9·70	13·70	7·09	109° 45′	8	$C2/c$ or Cc	1·46	1·485
5-(N-Nitro)aminotetrazole	Monoclinic	9·40	5·56	9·36	105° 0′	4	$P2_1/c$	1·82	1·832
5-(N, N-Dimethyl)aminotetrazole	Orthorhombic	13·78	9·90	7·88	—	8	$Pbca$	—	1·398
1-Methyl-5-(N-methyl)aminotetrazole	Orthorhombic	14·24	10·01	7·77	—	8	$Pbca$	—	1·357

gan, and Eugene Lieber of the Chemistry Division, U.S. Naval Ordnance Test Station.

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Die Struktur der Harnstoff-Additionsverbindungen*.

Von W. Schlenk, *Badische Anilin und Sodafabrik, Ludwigshafen am Rhein, Deutschland*

(*Eingegangen am* 13. *Februar* 1953)

In der Mitteilung von A. E. Smith 'The Crystal Structure of the Urea–Hydrocarbon Complexes' findet sich in der Einleitung folgender Passus: 'A general investigation of the field of urea complexes was carried out at these and associated laboratories .... In the early stages of this work an investigation of the structure of these complexes was therefore undertaken .... Since the completion of most of this work several additional papers (Zimmerschied, Dinerstein, Weitkamp & Marschner, 1949*a*, *b*, 1950; Schlenk, 1949) on these complexes, including a brief account of the structure determination by C. Hermann (Schlenk, 1949), have appeared.' Diese Formulierung könnte Anlass zu einer irrigen Auffassung über den Zusammenhang und die Entstehungszeiten der massgeblichen Arbeiten geben, die auf diesem Feld geleistet worden sind.

Die Entdeckung der Harnstoffaddition geradkettiger Verbindungen (1939) wurde von M. F. Bengen in einer Patentanmeldung niedergelegt, die 1940 in den Besitz der Badischen Anilin- und Sodafabrik übergegangen ist. Die Arbeiten der BASF auf dem durch die Entdeckung Bengens erschlossenen Feld haben 1940 begonnen und sind bis heute ohne Unterbrechung fortgesetzt worden. Sie gelangten bereits 1941 bis zur Strukturaufklärung der Addukte. Über die in den Laboratorien der BASF erhaltenen Ergebnisse wurde 1949 zusammenfassend berichtet (Bengen & Schlenk, 1949; Schlenk, 1949); aber schon 1945 waren die bis dahin gewonnenen Resultate, auch das bereits 1941 gefundene, und später von Smith (1952) untersuchte und bestätigte Strukturbild der Harnstoffaddukte, den unsere Laboratorien informationshalber aufsuchenden wissenschaftlichen Beauftragten des Auslandes mitgeteilt worden.

Nach einer Auskunft von Herrn A. E. Smith, die uns durch den Herausgeber dieser Zeitschrift (P. P. Ewald) zur Kenntnis gebracht wurde, ist die Struktur der Harnstoffaddukte in den amerikanischen Laboratorien in durchaus selbstständiger Arbeit ermittelt worden. Dieser Sachverhalt soll nicht bezweifelt werden; es ist vielmehr im Interesse der Sache nur zu begrüssen, wenn ein neuartiger und wichtiger Befund durch voneinander unabhängige Untersuchungen von verschiedenen Seiten gesichert wird. Historisch jedoch kann es leicht irreführend wirken, wenn die Hermannschen Befunde und unsere übrigen grundlegenden Untersuchungen auf diesem Gebiet als 'additional papers' aufgeführt werden.

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* Ergänzende Bemerkung zu der Mitteilung von Smith (1952).