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RP-146

25 JAN 1954

Reprinted from the "Proceedings of the Indian Academy of Sciences," Vol XXVIII, 1948



STUDIES IN COLLOID OPTICS

Part I. The Sol-Gel Transformation of Agar-Agar Gels

BY

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CATALOG

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Part I. The Sol-Gel Transformation of Agar-Agar Gels

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Received July 24, 1948

(Communicated by Dr. L. A. Ramdas, F.A.Sc.)

1. INTRODUCTION

THE transformation sol-gel or *vice versa* can be investigated by the methods of colloidal optics. The recent development in the field of Colloid-Optics, particularly in the hands of R. S. Krishnan,¹ has provided a very elegant method of investigating these problems.

The light scattering technique developed by R. S. Krishnan,¹ with the aid of a pair of double image prisms and the resulting four Tyndall cones, has proved very useful indeed for studying the variations in the size and shape of colloid particles.

2. THE EXPERIMENTAL TECHNIQUE OF R. S. KRISHNAN

Unpolarised light from a uniformly illuminated slit S (Fig. 1) is condensed by a long focus lens L through the double image prism P₁, which is adjusted so as to give two tracks, one vertically below the other as at V and H. These two well separated beams are of equal intensity, the electrical vector in V being vertical and that in H being horizontal. At the focus of these two beams is placed the scattering medium under investigation, contained in a suitable rectangular cell. The two tracks are viewed at right angles to the incident direction (in the horizontal plane) through the second

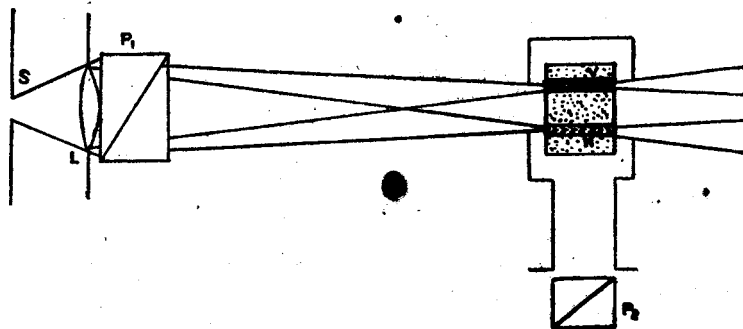


FIG. 1

double image prism P_2 which is crossed with respect to P_1 . We now see four beams as shown in Fig. 2. The incident beam V is split up into two beams V_v and H_v , the electric vector in them being *vertical* and *horizontal* respectively.

Similarly the incident beam H is split up into V_H and H_H (see Fig. 2) where the electric vectors are vertical and horizontal respectively.

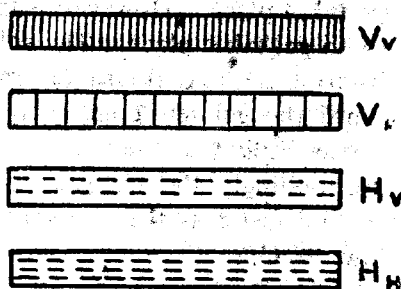


FIG. 2

As shown by R. S. Krishnan the principle of reciprocity is obeyed and the tracks V_H and H_v are always equal to each other in intensity.

The ratio $\frac{V_H}{H_H} = \rho_H$ is found to be a measure of the *size* of the scattering particles. Thus ρ_H is equal to 1 for particles of molecular dimensions and *decreases* in value as the size of the scattering particle increases.

Again, the ratio $\frac{H_v}{V_v} = \rho_v$ gives a measure of the anisotropy of the particle. It is *zero* for perfectly symmetrical particles and increases with the degree of asymmetry.

In the present investigation the optical method outlined above has been applied to the study of agar-agar dissolved in water in transformation from the 'Sol' to the 'gel' state and *vice versa*. Krishnamurthi² had investigated this problem earlier in regard to agar-agar but he had used the then known technique of one incident beam of unpolarised light. The present investigation is a re-survey of the field covered by Krishnamurthi, using the new technique of R. S. Krishnan. Before discussing the variations of ρ_H and ρ_v the corresponding variations in the total scattering by a centimeter layer of the gel-sol with incident light unpolarised are described.

3. PREPARATION OF THE SOLS

A weighed quantity of agar-agar in the form of white fibres is kept in running water for about an hour. It is then boiled with distilled water in

which it dissolves completely to form a turbid sol. The sol is then filtered hot several times to obtain a very clear transparent sol which is collected in a large test-tube. The test-tube is kept in a hot water-bath to prevent the sol from setting. This sol, when examined in a strong beam of convergent light shows no bright specks of light.

If the sol is maintained at a temperature above 40°C . for any length of time no appreciable change takes place. However, if the sol is below 40°C ., a slow conversion from sol to gel takes place. The sol becomes increasingly viscous and ultimately sets to a gel. At the same time the intensity of the scattered light increases until the gel sets completely. The time taken by the sol to set to a gel decreases rapidly as the temperature of transformation falls below 40°C . Correspondingly the changes in the intensity of the scattered light occur faster and faster as the temperature of the experiment falls more and more below 40°C .

During the gel-sol transformation the changes noticed are not exactly the reverse of the sol-gel transformation. The tendency is for the gel state to persist indefinitely even at temperatures as high as 60°C . As the temperature is increased still higher the gel gradually transforms itself into the sol state without indicating any distinct temperature of transformation.

4. METHOD OF MEASURING THE INTENSITY OF SCATTERED LIGHT

(a) *Experimental arrangements for measuring intensity in the transverse direction.*—Light from a pointlite lamp is concentrated on a square aperture at S (see Fig. 3). It then passes through the converging lens L and is concentrated at P. The sol to be examined, kept in a test-tube is immersed in a rectangular glass cell containing clean water. The temperature of the sol is given by the thermometer T_1 and that of the water-bath W by the thermometer T_2 . The whole of the test-tube is then painted dull black on the outside. There are just three rectangular apertures of suitable size, all in a horizontal plane, of which two serve for the entrance and exit of the beam of incident light, while the third aperture serves to observe the track of the incident beam at right angles to the direction of the incidence.

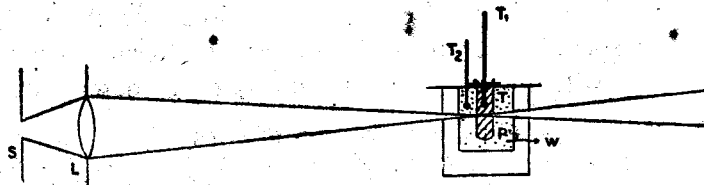


FIG. 3

The test-tube enters the water cell through an aperture in the cover and can be adjusted vertically. The water cell is 10 cm. by 10 cm. by 10 cm., made of good quality glass plates without optical strain. The glass sides are all painted dull black leaving only 3 transparent areas corresponding to those in the test-tube. The water cell is in turn surrounded by a wooden box the inner and outer sides of which are also painted dull black. This box also has three holes corresponding to the apertures in the test-tube and the glass cell. The transversely scattered light passes through the observational window and falls normally on a photo-cell which has a square aperture 1 cm. by 1 cm. and which is 10 cm. away from the track whose intensity is to be measured. As the photo-electric cell was most sensitive to red, a red filter was kept in front of the cell. The photocell and amplifier units are similar to those used by Ananthakrishnan,³ where the out of balance current in a wheatstone bridge system is a function of the intensity of the radiation incident on the cell. The bridge is adjusted so that the micro-ammeter reads zero with no illumination. The reading of the micro-ammeter increases with the intensity of the scattered light.

(b) *Range of the photometer for linear response.*—To see in what range the response of the above photometer is linear the following subsidiary experiment was performed. The light passing through the lens L is made to pass through two nicol prisms, one of which can rotate on a divided circle about a horizontal axis. The light transmitted by these two nicols is allowed to fall on the photocell of the photometer. The bridge is adjusted so that the micro-ammeter reads zero when the nicols are crossed. The deflections in the microammeter for different rotations θ of the adjustable nicol away from its crossed position are then noted. The intensity I corresponding to a rotation θ of the nicol is given by

$I = I_0 \sin^2 \theta$, where I_0 is the maximum intensity when the nicols are in the parallel position.

Fig. 4 shows that the response of the photometer is linear in the range of current from 0 to 90 micro-amperes, which covers the range of readings when the photometer is used to measure the intensity of scattered light in our experiments.

(c) *Measurement of intensity I_0 of the incident beam.*—To express the scattered light as a fraction of the incident light, a knowledge of the intensity of the incident light I_0 is necessary. This, however, cannot be measured directly by the photo-cell unit as it is too intense. Therefore, the intensity of the incident beam was cut down to the working range by reducing the aperture of the pointolite to its minimum and introducing a strong filter

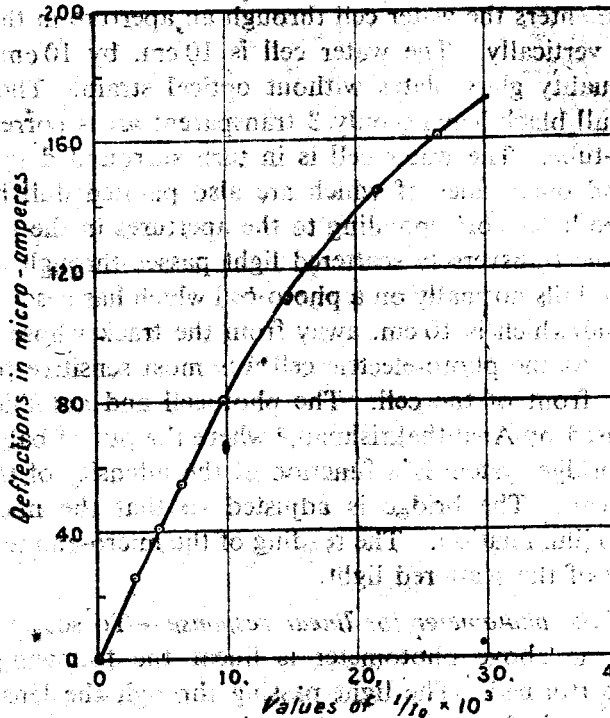


Fig. 4

of prussian blue, which has a strong absorption band in the red in front of the photo-cell. Various values of I transmitted through the filter for different concentrations of filter solution were measured in the range of the instrument. I , the intensity transmitted through the filter is given by

$$I = I_0 10^{-KC}$$

where I_0 is the initial intensity and C is the concentration and K a constant so that $\log I = \log I_0 - KC$.

Fig. 5 is the linear graph of $\log I$ against C from which by extrapolation $\log I_0$ is obtained by putting $C = 0$.

From Fig. 5 we have $\log I_0 = 3.76$ or $I_0 = 5754$ with minimum aperture width in terms of micro-ammeter readings in the range where the response is linear.

(d) Estimation of the total scattering (over a solid angle 4π) by a thin layer of the medium. In Fig. 6 let the horizontal track of the incident beam be AB along the x axis.

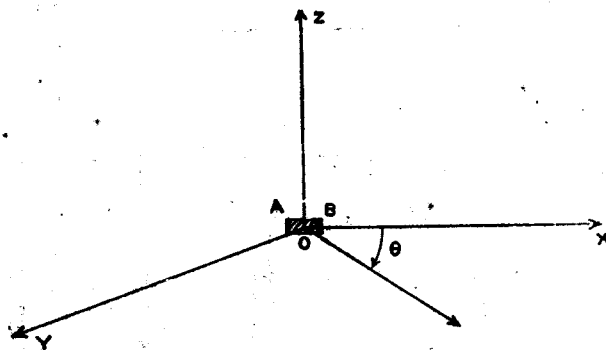
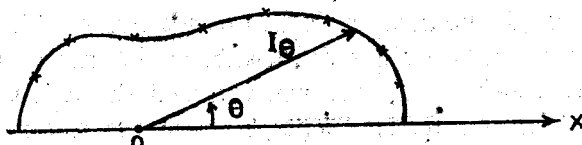


FIG. 6



Intensity distribution in the horizontal plane
For gel set at 25 °C.

FIG. 7

Using the values of intensity given in Fig. 7 it is easy to compute the total intensity of the scattered light passing out of the whole sphere as

$$I_T = \Sigma I_\theta (\text{zone}) = 2\pi \times 10^3 \times \Sigma I_\theta \sin \theta d\theta$$

$$= 20\pi \Sigma I_\theta \sin \theta$$

putting $d\theta = \frac{1}{10}$ radian, equal to the solid angle subtended by the aperture of the photo-cell.

This total intensity I_T can be expressed as a fraction $\frac{I_T}{I_0}$ of the incident beam. In Tables II to XI this value is given in the last column.

(e) *Measurement of the transmission by a layer of gel or sol.*—A beam of light passing through a diffusing medium undergoes depletion by scattering. Using the photometer it is easy to measure the transmission by a known thickness of the gel or sol. It is possible, therefore, to compare the total loss by scattering as estimated in (d) above with the loss in transmission.

An example of such comparison is given below:—

TABLE I
Light scattered by gel set at 25° C.

| Concentration of agar-agar in the gel | Total light scattered by one centimetre layer over solid angle 4π | Loss in transmission per cm. |
|---------------------------------------|---|------------------------------|
| % 0.2 | % 0.4 | % 0.38 |
| 0.5 | 1.0 | 1.1 |
| 1.0 | 2.4 | 2.3 |
| 1.5 | 4.8 | 5.0 |

From the above it is seen that most of the loss of intensity during the passage through the medium is due to scattering. This loss, in the case of agar, for the concentrations dealt with, is also not so large as to affect the intensity of the Tyndall cone when viewed through the thin layer of the medium which usually intervenes between the track and the boundary of the containing vessel nearest to the observer.

5. MEASUREMENT OF DEPOLARISATION

The method of obtaining the four Krishnan components has already been described in para 2. The outside of the test-tube containing the gel under investigation is covered dull black excepting for three small square holes in a horizontal plane, two of which are in a straight line to allow the incident beam to pass through the gel while the third one is at right angles to the line joining the first two, for observations. The test-tube is held vertical so that only the upper one of the two incident beams (Fig. 1), namely V, passes through the gel, the lower H being cut off. Correspondingly, only the first and the third beams (Fig. 2) are obtained when viewed through the double image prism P₁. They are then observed through a nicol to which a divided circle is attached. The two successive positions of the nicol θ_1 and θ_2 at which the two beams appear to be of equal intensity are found. The ratio ρ_v of the third to the first beam is calculated by

$$\rho_v = \tan^2 \frac{(\theta_2 - \theta_1)}{2}$$

The test-tube is then lowered slightly so that the upper beam V is cut off and the lower beam H only passes through the gel. The ratio ρ_H of the resulting two beams, namely (Fig. 2) the second to the fourth, is obtained in the same way as ρ_v . If the double image prism in the path of the incident beam is

removed then we have a beam of unpolarised light passing through the scattering medium. If we now determine the ratio of the two components, seen through the double image prism on the observation side we get the value of ρ_u which is related to ρ_v and ρ_H as

$$\rho_u = \frac{1 + \frac{1}{\rho_H}}{1 + \frac{1}{\rho_v}}$$

We are therefore in a position to compare the observed values of ρ_u with those calculated from the value of ρ_H and ρ_v measured independently.

6. MEASUREMENTS OF ρ_v , ρ_H , ρ_u AND I IN AGAR-AGAR GELS OF VARIOUS CONCENTRATIONS

Measurements of ρ_v , ρ_H , ρ_u and I the intensity of transversely scattered light during the sol-gel transitions of agar-agar gels of concentrations 0.06%, 0.2%, 0.5%, 1% and 1.5% were recorded in a series of experiments.

(a) *Sol-gel transition.*—The sol prepared for the investigation is first maintained at 90° C. for sometime by keeping the test-tube containing the sol in a hot water-bath and is then cooled to 80° C. by decreasing the temperature of the bath. It is then maintained at the constant temperature 80° C. for about 12 hours. At intervals of an hour the test-tube containing the sol is transferred to the water cell (Fig. 2) containing water at 80° C. for a short time and the intensity of the track viewed at right angles to the incident beam is measured with the photoelectric photometer. No change in intensity is noticed even after 12 hours. After measuring the intensity, the photo-cell unit is removed, and the double image prisms are inserted as in Fig. 1, to measure the values of depolarisation ρ_v , ρ_H , ρ_u . The above observations of intensity and polarisation are repeated at temperatures of 70° C., 60° C., 50° C., and 40° C., each time bringing the sol from 90° C. to the respective temperature and recording the variation of intensity with time keeping the particular temperature constant until no further change of intensity is noticed and finally the depolarisation values are also measured. No appreciable changes in intensity and depolarisation with time are noticed even after 12 hours at these temperatures above 40° C. Below 40° C., however, the observations are taken at each temperature down to 30° C. For each of these observations the sol is first cooled from 90° C. to 40° C. in the outside bath and the test-tube containing the sol at 40° C., is then transferred to the outer cell containing water at 40° C. The sol, along with the water in the outer cell is cooled at the uniform rate of one degree for

every three minutes, to the particular temperature required which is maintained constant and observations of variations in intensity with time are taken until the intensities reach their maximum constant values and finally the depolarisations ρ_v , ρ_H , ρ_u are also recorded.

The sol first becomes highly viscous and then sets to a gel as time passes. It will be seen from Tables III to VII that there is no sensible change in the intensity from 90°C . to 38°C . For 37°C . and below the intensity increases with time until it reaches a maximum constant value at the particular temperature, time being measured from the instant the sol reaches the particular temperature. At lower temperatures the increase in intensity is more and more rapid and the limiting or saturation values of intensity are also reached earlier. Figs. 8 to 12 show the variation in intensity with time at different temperatures for gel concentrations of 0.06%, 0.2%, 0.5%, 1%, and 1.5%. The final values of intensity for a typical concentration are also shown by the portions OAB of the diagram in Fig. 13.

Gel-sol transformation.—The gel set at 25°C . is slowly heated to different temperatures and maintained at these temperatures until the values of intensity and depolarisation become steady. The curve CD in Fig. 13 shows the final steady value of intensity at each temperature during the gel-sol transition. Similar curves were obtained for other concentrations

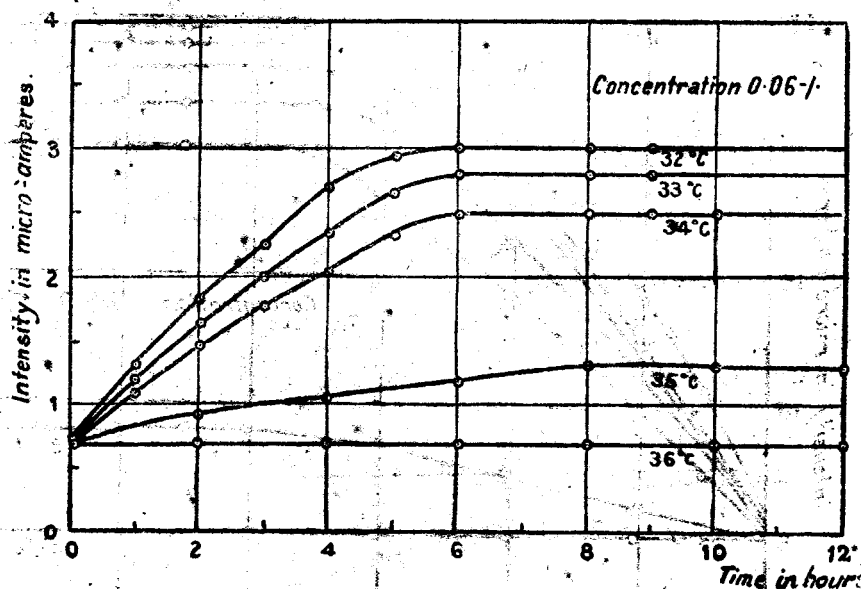


FIG. 8

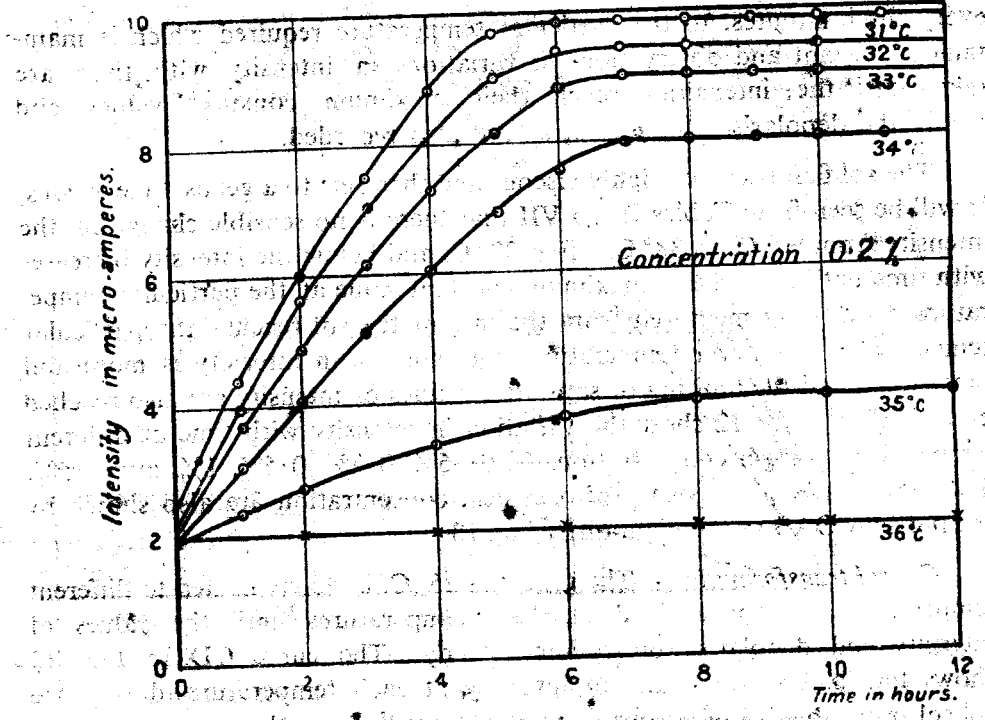


FIG. 9

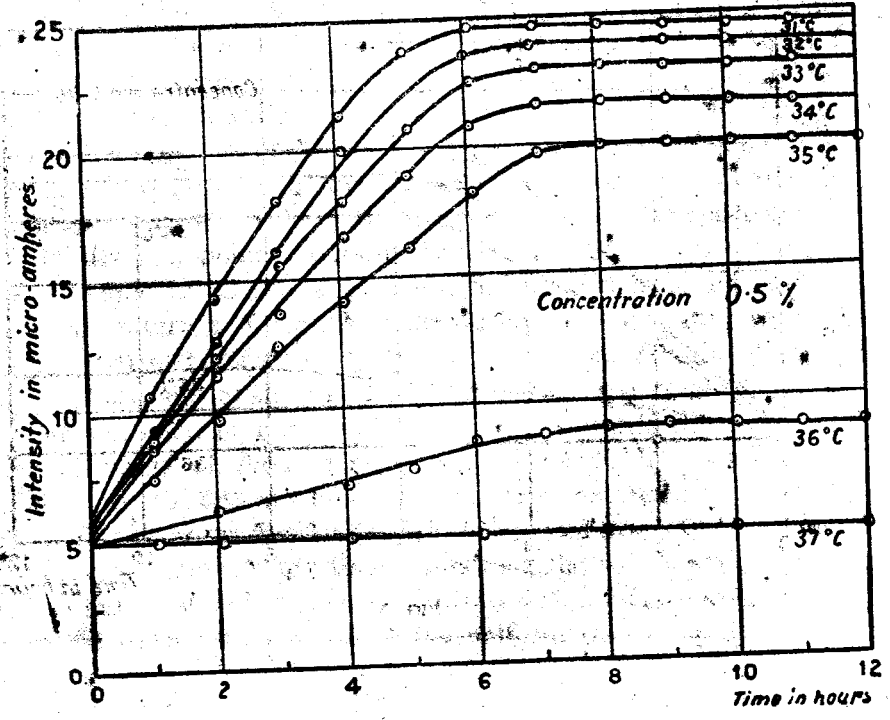


FIG. 10

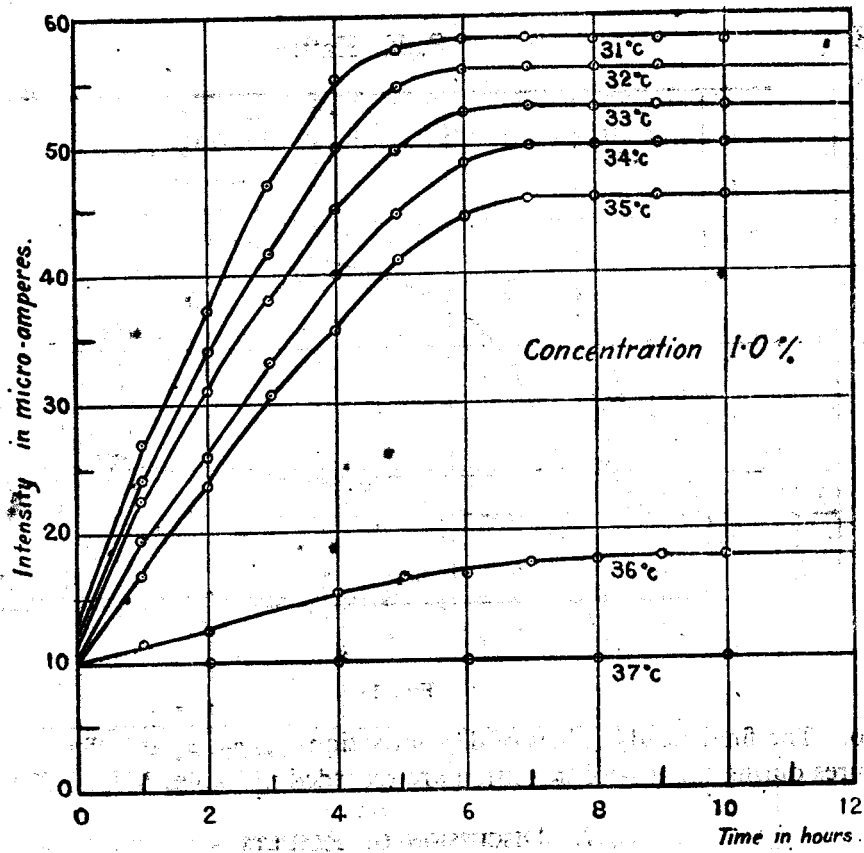


FIG. 11

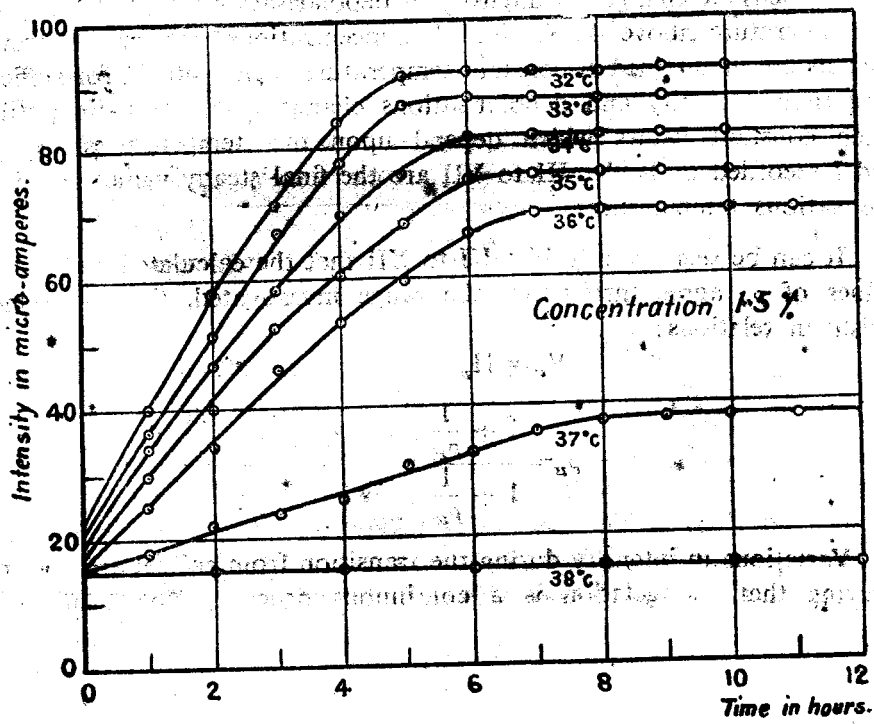


FIG. 12

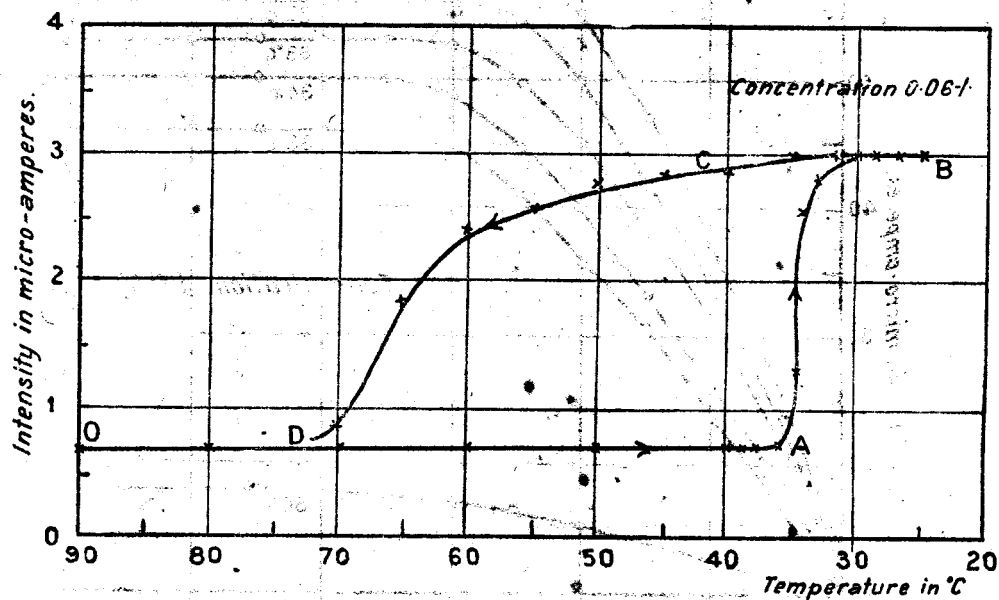


FIG. 13

also. The final steady values of depolarisations ρ_v , ρ_H , ρ_M at different temperatures during the gel-sol transition are recorded in Tables VIII to XII.

7. DISCUSSION OF RESULTS

No sensible changes of intensity or depolarisation with time are noticed at temperature above 40°C. for all concentrations studied. However, if the sols are maintained at constant temperatures below 40°C. for sufficiently long time, intensity and depolarisations change over gradually until they reach constant values which depend upon the temperatures. ρ_H , ρ_v , ρ_M and I recorded in Tables III to VII are the final steady values obtained at the various temperatures.

It can be seen from Tables III to XII that the calculated and observed values of ρ_M agree throughout the range investigated, thus verifying the Krishnan relations:

$$V_H = H_v \quad (1)$$

$$\rho_M = \frac{1 + \frac{1}{\rho_H}}{1 + \frac{1}{\rho_v}} \quad (2)$$

Variations in intensity during the transition from sol-gel are continuous showing that the gelation is a continuous process. The cooling curve

(Fig. 14) between 40° C. and 29° C. does not show any kink associated with sudden evolution or absorption of heat during the transition from sol to gel.

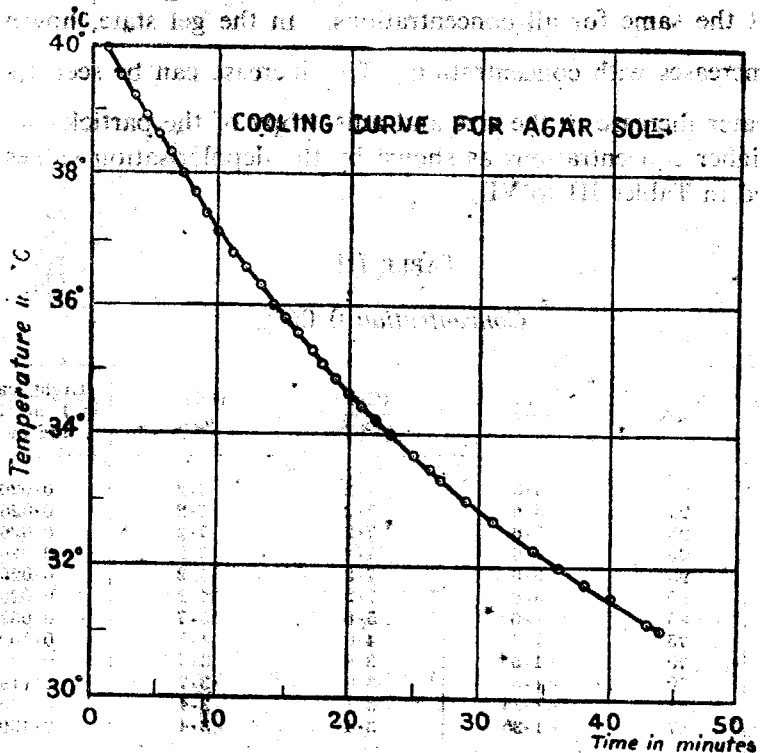


FIG. 14

TABLE II

Percentage of total light scattered (in all directions) in the sol and gel state, by agar-agar gels of different concentrations

| Concentration | Light scattered in the sol state (temp. above 40° C.) I _s | Light scattered by gel set at 25° C. I _g |
|---------------|--|---|
| 0.06 | 0.026 | 0.13 |
| 0.2 | 0.08 | 0.4 |
| 0.5 | 0.2 | 1.0 |
| 1.0 | 0.4 | 2.4 |
| 1.5 | 0.6 | 4.0 |

Variations in Intensity.—At temperatures above 40° C. the total amount of the light scattered in the sol state is directly proportional to the concentration (Table II), showing that size and shape distribution of the suspended

TABLE VIII.
Concentration 0.06%

| Temperature in °C. | $\rho_n\%$ | $\rho_p\%$ | (Cal.) $\rho_u\%$ | (Obs.) $\rho_u\%$ | Light scattered by 1 cm. layer of the medium |
|--------------------|------------|------------|----------------------|----------------------|--|
| 25 | 67 | 1.35 | 3.4 | 3.4 | 0.13×10^{-2} |
| 30 | 67 | 1.35 | 3.4 | 3.4 | 0.13 " |
| 35 | 67 | 1.35 | 3.4 | 3.4 | 0.13 " |
| 40 | 70 | 1.5 | 3.8 | 3.7 | 0.11 " |
| 45 | 70 | 1.5 | 3.8 | 3.7 | 0.11 " |
| 50 | 70 | 1.5 | 3.8 | 3.7 | 0.108 " |
| 55 | 75 | 1.7 | 4.0 | 4.1 | 0.104 " |
| 60 | 75 | 1.7 | 4.0 | 4.2 | 0.096 " |
| 65 | 85 | 3.0 | 6.5 | 6.6 | 0.072 " |
| 70 | 90 | 3.4 | 7.0 | 4.9 | 0.032 " |

TABLE IX
Concentration 0.2%

| Temperature in °C. | $\rho_n\%$ | $\rho_p\%$ | (Cal.) $\rho_u\%$ | (Obs.) $\rho_u\%$ | Light scattered by 1 cm. layer of the medium |
|--------------------|------------|------------|----------------------|----------------------|--|
| 25 | 65 | 1.60 | 4.0 | 4.0 | 0.40×10^{-2} |
| 30 | 65 | 1.60 | 4.0 | 4.0 | 0.40 " |
| 35 | 65 | 1.60 | 4.0 | 4.0 | 0.40 " |
| 40 | 65 | 1.60 | 4.0 | 4.0 | 0.39 " |
| 45 | 65 | 1.60 | 4.0 | 4.0 | 0.39 " |
| 50 | 70 | 1.9 | 4.5 | 4.6 | 0.37 " |
| 55 | 70 | 1.9 | 4.5 | 4.6 | 0.36 " |
| 60 | 75 | 2.2 | 5.0 | 5.2 | 0.34 " |
| 65 | 80 | 2.8 | 6.2 | 6.1 | 0.24 " |
| 70 | 95 | 3.6 | 7.2 | 7.1 | 0.10 " |

TABLE X AT
Concentration 0.5%

| Temperature in °C. | $\rho_n\%$ | $\rho_p\%$ | (Cal.) $\rho_u\%$ | (Cal.) $\rho_u\%$ | Light scattered by 1 cm. layer of the medium |
|--------------------|------------|------------|----------------------|----------------------|--|
| 25 | 65 | 1.6 | 4.2 | 4.2 | 1.0×10^{-2} |
| 30 | 65 | 1.6 | 4.2 | 4.2 | 1.0 " |
| 35 | 65 | 1.6 | 4.2 | 4.2 | 1.0 " |
| 40 | 65 | 1.6 | 4.2 | 4.2 | 0.98 " |
| 45 | 65 | 1.6 | 4.2 | 4.2 | 0.98 " |
| 50 | 67 | 1.7 | 4.2 | 4.2 | 0.96 " |
| 55 | 70 | 1.9 | 4.5 | 4.6 | 0.92 " |
| 60 | 70 | 1.9 | 4.5 | 4.5 | 0.90 " |
| 65 | 90 | 2.4 | 7.0 | 7.0 | 0.46 " |
| 70 | 95 | 2.8 | 7.2 | 7.2 | 0.24 " |

TABLE XI
Concentration 1.0%

| Temperature in °C. | ρ_H % | ρ_v % | (Calc.) ρ_H % | (Obs.) ρ_H % | Light scattered by 1 cm. layer of the medium |
|--------------------|------------|------------|--------------------|-------------------|--|
| 25 | 60 | 3.0 | 7.3 | 7.4 | 2.4×10^{-2} |
| 30 | 58 | 3.0 | 7.3 | 7.4 | 2.4 " |
| 35 | 60 | 3.0 | 7.3 | 7.4 | 2.4 " |
| 40 | 60 | 3.0 | 7.3 | 7.3 | 2.35 " |
| 45 | 60 | 3.0 | 7.3 | 7.3 | 2.35 " |
| 50 | 65 | 3.0 | 7.3 | 7.3 | 2.30 " |
| 55 | 65 | 3.0 | 7.3 | 7.3 | 2.30 " |
| 60 | 65 | 2.8 | 6.8 | 6.7 | 2.15 " |
| 65 | 75 | 3.1 | 7.0 | 7.2 | 1.04 " |
| 70 | 90 | 3.4 | 7.2 | 7.2 | 0.55 " |

TABLE XII
Concentration 1.5%

| Temperature in °C. | ρ_H % | ρ_v % | ρ_H % | ρ_H % | Light scattered by 1 cm. layer of the medium |
|--------------------|------------|------------|------------|------------|--|
| 25 | 55 | 8.4 | 22.0 | 22.0 | 3.8×10^2 |
| 30 | 55 | 8.4 | 22.0 | 22.0 | 3.8 " |
| 35 | 60 | 8.0 | 20.0 | 20.0 | 3.7 " |
| 40 | 60 | 8.0 | 20.0 | 20.0 | 3.65 " |
| 45 | 60 | 8.0 | 20.0 | 20.0 | 3.65 " |
| 50 | 60 | 8.0 | 20.0 | 20.0 | 3.60 " |
| 55 | 65 | 5.8 | 13.8 | 14.0 | 3.4 " |
| 60 | 65 | 5.8 | 13.8 | 13.8 | 2.8 " |
| 65 | 75 | 5.4 | 12.0 | 11.7 | 2.0 " |
| 70 | 90 | 4.2 | 8.5 | 8.8 | 0.8 " |

Variations in ρ_H .—The second columns in Tables III to VII show that ρ_H decreases with decrease in temperature suggesting that the size of the particles for gels set at lower temperatures is bigger. It can also be seen that ρ_H decreases with concentration at low temperatures showing that for gels set at the same low temperature the size of the particles increases with concentration.

Variations in ρ_v .—The changes occurring in ρ_v with decrease in temperature are very interesting and occur as follows for different ranges of concentration.

- (1) For sols of concentration 0.06%, 0.2% and 0.05%, ρ_v gradually decreases with temperature.
- (2) For 1% sol ρ_v first decreases and then increases as the temperature is decreased.
- (3) For 1.5% sol ρ_v gradually increases with temperature.

This suggests that in the first case the particles tend to become more symmetrical with decrease in temperature while in the second they tend to grow more symmetrical with decrease in temperature at first but asymmetry sets in with further lowering in the temperature. At 1.5% concentration the increase in asymmetry of the particle starts right from the beginning.

Mechanism of sol-gel transformation in agar-agar as suggested by the above results.—A consideration of the above mentioned variations in intensity, ρ_H and the three types of variations of ρ_v at the different ranges of concentration suggests that we have to deal here with two distinct phenomena. First of all we have the solute particles of small size initially, but tending to increase in size rapidly with the fall of temperature, as one approaches the range of temperatures associated with the sol-gel transformation. This process may occur in two stages. In the first stage (a) the particle tends to develop a cloud of H_2O molecules which surround it without however actually entering its structure. During this initial stage the intensity of the scattered light does not start increasing though the medium increases in viscosity. In the next stage of swelling; (b) we have also the actual physical swelling of the solute particle itself, somewhat analogous to what happens in osmosis. During this second stage, the intensity of scattering increases rapidly. In any case it is clear that during the stages (a) to (b) the individual particle undergoes an increase of size which tends to make it more symmetrical as is shown by the decrease in ρ_v . The later increase in ρ_v with further fall of temperature must be associated with aggregation or coagulation of swelled particles with a tendency to form increasingly unsymmetrical groups. This tendency becomes more pronounced at the higher concentration and indeed starts there at the very outset, as is the case with the highest concentration of 1.5%. Work on similar lines with gelatin gels is in progress.

SUMMARY

The transmission from sol to gel and *vice versa* in agar-agar gels has been investigated by means of the new experimental technique of R. S. Krishnan. Intensity distribution of the light scattered by a one centimeter layer of the scattering medium in the horizontal plane with incident unpolarised light has been measured by means of an electronic photometer during the various stages of the transition, and hence the total light scattered in all directions by a centimetre layer of the scattering medium during the transition has been calculated and expressed as a fraction of the incident intensity.

The depletion of the light transmitted through various thickness of the gels has also been measured and shown to be equal to the total light scattered by the gel. The detailed study of the variations in intensity and depolarisations during the various transitions from sol to gel and gel to sol suggest,

(1) The R. S. Krishnan relation :

$$\rho_{\mu} = \frac{1 + \frac{1}{\rho_H}}{1 + \frac{1}{\rho_v}}$$

holds throughout the range investigated.

(2) The transition from sol to gel is characterised by distinct stages.

(a) The solute particle first appears to develop a cloud of H₂O molecules round itself which results in an increase in the viscosity without any sensible change in the light scattering capacity of the gel.

(3) The actual particle size then increases as is indicated by the great increase in the light scattering capacity as well as the decrease in the value of ρ_H as compared with the sol. The values of ρ_v indicate that:

(a) The particles grow more symmetrical as their size increases for concentrations of 0.06%, 0.2%, and 0.5%.

(b) The particles first tend towards symmetry and later grow more and more unsymmetrical with increase in size for the concentrations of 1% while,

(c) The tendency for the particles to grow more unsymmetrical with increase in size exists from the very start for concentrations of 1.5% or more.

ACKNOWLEDGEMENT

In conclusion, I wish to express my deep gratitude to Dr. L. A. Ramdas, Director of Agricultural Meteorology, for suggesting me the problem, for guidance and encouragement during the course of the investigation and for providing the necessary facilities for the work. My thanks are also due to Mr. R. S. Kale for building the photo-electric unit used in the present investigation.

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