LII.—*The Viscosity of Solutions of Cellulose.*

By WILLIAM HOWIESON GIBSON.

PART I.

DURING the progress of work in connexion with the manufacture of propellant powders, some observations of general interest on the viscosity of solutions of cellulose have been made, and this paper gives a brief account of this portion of the work.

Very many determinations of viscosity have been made, so a selection only has been given.

A number of determinations of the viscosity of cuprammonium solutions of various forms of cellulose, such as cotton, wood-pulp, etc., were made by Ost (*Zeitsch. angew. Chem.*, 1911, **24**, 1892), who claimed that viscosity determinations carried out by his method gave useful information on the nature and technical value of celluloses.

As the determination of viscosity seemed to be a promising method of obtaining information of technical value, a considerable number of experiments were carried out by Ost's method, but the results were far from satisfactory, owing to the progressive changes which took place in the solution, which are referred to in Ost's paper.

In order to improve the method, elimination of the progressive change in the solutions was essential, and as air-oxidation seemed a probable cause, complete exclusion of air was tried.

The difficulties in obtaining trustworthy viscosity determinations were overcome in this way by the use of the hydrogen capillary viscosimeter, and, later, the falling sphere viscosimeter (see Part II).

Although the hydrogen capillary viscosimeter was somewhat troublesome to manipulate and could only be used for solutions of cellulose up to 1 per cent., it was found very useful in studying the effect of various treatments on cellulose.

The viscosity in C.G.S. units of 1 per cent. solutions in cuprammonium of twenty-eight samples of purified cotton-waste examined in the hydrogen capillary viscosimeter ranged from 4.53 to 0.14, and the viscosity of a trade sample of unpurified waste was found to be 7 units. These results indicated that considerable difference existed between different samples of purified cotton-waste, which could probably be ascribed to difference in treat-

ment. The high viscosity of unpurified cotton was also definitely established.

The effect of variation in the conditions of treatment of cellulose with sodium hydroxide solutions on the viscosity of the product was studied on the laboratory scale, using the hydrogen capillary viscosimeter.

The results obtained in these experiments are given in the following tables, and are plotted in Fig. 1.

TABLE I.

Conditions of boiling. Cotton 100 parts. NaOH 18 parts.	Number of curve in Fig. I.	Viscosity in 1 per cent. solution C.G.S. units. Time of boiling in hours.					
		0.	1½.	3.	6.	9.	12.
Pressure, 5.6 kilos. per sq. cm.							
4 per cent. NaOH solution.	I.	14.48	0.40	0.13	0.09		
2 " " "	II.	14.48	0.54	0.21	0.14		
Pressure, 2.8 kilos. per sq. cm.							
4 per cent. NaOH solution.	III.	14.48	2.51	1.46	0.84	0.45	0.42
2 " " "	IV.	14.48	3.61	2.40	1.87	0.75	0.53
Pressure, 1.4 kilos. per sq. cm.							
4 per cent. NaOH solution.	V.	14.48					2.65
2 " " "	VI.	14.48					3.90

TABLE II.

		Parts of NaOH solution to 1 part of cotton.			
		2.	4.	8.	16.
Pressure, 5.6 kilos. per sq. cm.					
Time 1½ hours.					
4 per cent. NaOH solution		0.43	0.40	0.29	0.32
2 " " "		1.27	0.72	0.54	0.53

Consideration of these results leads to the following conclusions:
 (1) Temperature is the main factor in reducing the viscosity of cotton during boiling with sodium hydroxide.

(2) The viscosity falls rapidly at the beginning of the experiment, and the rate of fall slackens as the boiling continues.

(3) The concentration of sodium hydroxide solution influences the viscosity. With a 2 per cent. solution, the viscosity is

approximately about 50 per cent. higher than with a 4 per cent. solution.

(4) At high temperatures, the viscosity falls so rapidly that the concentration of the solution is of little practical importance.

(5) With mild treatments, the actual difference between concentrated and dilute solutions is more pronounced.

(6) When the proportion of solution to cotton is increased, the

FIG. 1.

viscosity is reduced. This may be due to the concentration of the solution remaining more uniform during the boiling.

The experiments indicated quite clearly that the viscosity of purified cotton could be controlled by controlling the conditions of the boiling process. Examination of samples of bleached and unbleached cotton-waste were made, and led to the conclusion that the process of bleaching with weak bleaching powder solution

had little effect on viscosity. The effect of treating the cotton with sulphuric acid (0.25 per cent.) was found to be negligible; thus, the viscosity of the same sample of cotton before treatment was 0.42 unit, and afterwards 0.45 unit.

Many samples of cotton boiled on the manufacturing scale under various conditions have now been examined with the falling sphere viscosimeter, and several interesting relationships have been indicated. The large-scale work has proved conclusively that the conditions of the boiling process determine the viscosity of the purified cellulose, and it has been found possible by standardising these conditions to keep the viscosity of cotton-waste used for nitration within much narrower limits than heretofore. The results of the determination of the viscosity of one blend of cotton-waste, using different treatments, are given in table III.

TABLE III.

Solution.	Pressure of steam. Kilos. per sq. cm.	Time. Hours.	Viscosity of 2 per cent. solution.
2 per cent. NaOH			
2 per cent. Na ₂ CO ₃	Atmospheric.	10	317
2 per cent. NaOH			
2 per cent. Na ₂ CO ₃	„	20	256
4 per cent. NaOH	„	10	332
4 per cent. NaOH	„	20	193
2 per cent. NaOH	1.4	10	240
2 per cent. NaOH	„	20	94
1 per cent. NaOH			
3 per cent. Na ₂ CO ₃	2.8	10	172
2 per cent. NaOH			
2 per cent. Na ₂ CO ₃	„	10	129
2 per cent. NaOH			
2 per cent. Na ₂ CO ₃	„	20	19
2 per cent. NaOH	„	10	73
2 per cent. NaOH	„	20	24
4 per cent. NaOH	„	10	48
4 per cent. NaOH	„	20	4

These results again indicate the importance of temperature in reducing viscosity, and the influence of strength of solution is shown. It is also evident that sodium carbonate is ineffective compared with sodium hydroxide.

The experiments on different classes of waste given in table IV are of considerable interest. In these experiments, the conditions were the same in all cases, but the Eastern cottons give products the viscosity of which is about half that of American cotton of similar grade.

There is also a marked difference in the viscosity obtained with long-fibred cop-waste and short-fibred fly. It is hoped that further work in this direction will enable more definite correlations to be

made between climatic conditions of growth of cotton and the viscosity of solutions, and also between maturity of fibre and viscosity.

TABLE IV.

Treatment.—Ten hours under a pressure of 2·8 kilos. per sq. cm. with 4 parts of 3 per cent. sodium hydroxide solution to 1 part of cotton.

Material.	Viscosity of 2 per cent. solution.
American Fly	128
Eastern Fly	53
American Cops	57
Eastern Cops	26

A further relationship has been shown to exist between the viscosity of cellulose solutions and the viscosity of nitrocellulose solutions. The property inherent in cellulose to which viscosity is due is transmitted to the nitrated derivative. Table V indicates that high viscosity in the cellulose is accompanied by high viscosity in its derivative.

TABLE V.

Viscosity of cotton : 2 per cent. cuprammonium solution.	Viscosity of nitro-cotton : 5 per cent. solution in a mixture of ether and alcohol.
317	67
172	43
48	29
4	10

At the instance of Mr. F. R. Jackson, Director of the Flax Experimental Station at Selby, five samples of flax supplied by him were examined for viscosity by the falling sphere method. The results obtained are given in table VI.

TABLE VI.

	Viscosity of 1 per cent. solution.
A. Pond retted, hand scutched flax	22·3
B. As A, but twice scutched by machine.....	19·7
C. Twice retted in tanks under controlled conditions....	30·2
D. Russian dew-retted, low grade.....	11·3
E. Originally scutched without retting. fibre then retted and re-scutched	26·5

These preliminary results indicate that the viscosity bears a relationship to the character of the flax; thus, Russian dew-retted, low-grade flax differs widely in viscosity from sample C, Selby flax, retted in warm water, and of far superior quality.

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The viscosity-concentration curve for cellulose in cuprammonium has been studied, but Arrhenius's simple formula appears only to apply over short ranges of concentration.

A relationship between viscosity and solubility of cellulose in cuprammonium solution is indicated by observations made with a variety of cellulose.

The work has scarcely progressed far enough for wide generalisations to be made, but it seems clear that certain treatments can be applied to cellulose which alter its physical properties profoundly, but produce no decided change in chemical properties. This lends support to the view that the production of cellulose in the plant is a process of polymerisation, the older layers, presumably, being more highly polymerised than new growth; the process of boiling with sodium hydroxide may effect the reverse change and depolymerise the cellulose.

PART II.

[With LEO SPENCER and ROBERT MCCALL.]

The Hydrogen Capillary Viscosimeter.

The chief difficulty with Ost's method of determination of viscosity is the progressive alteration which the solutions of cellulose undergo, changes in viscosity occurring all through the process of solution and up to the time of making the determination. Investigation showed that this change was caused by the presence of air and light, so a viscosimeter was devised in which solution of the cellulose was effected in an atmosphere of hydrogen and the influence of light was avoided. With this apparatus, concordant results were obtained, and it was possible to repeat determinations of viscosity of cellulose with satisfactory results. The apparatus consisted of: (1) a Kipp's hydrogen apparatus; (2) a washing bottle filled with sodium hydroxide solution to free the hydrogen from acid; (3) a glass cylinder of about 100 c.c. capacity to contain the cotton and cuprammonium solution, and a three-holed rubber stopper to fit; (4) a capillary viscosimeter, bulb 3—4 c.c., capillary of internal diameter 0.7 mm., length 12—15 cm.; (5) a tap funnel of 10 c.c. capacity to introduce the solution into the containing cylinders; (6) a supply of large glass beads.

The arrangement of the apparatus is shown in Fig. 2.

Procedure.—Half a gram of dry cotton is cut into small pieces to facilitate solution. The cotton, together with six or eight glass

beads, is introduced into the cylinder. The rubber cork containing the viscosimeter and inlet tubes is then inserted, and the air is displaced by a current of hydrogen whilst the cylinder is in an inverted position. A period of less than twenty minutes is necessary for complete displacement of air, and this is most essential, as even minute traces of air lower the viscosity of the cuprammonium solution of cotton. When the displacement of air is complete, 50 c.c. of cuprammonium solution are added in the following manner. The tap funnel, *D*, is attached to the capillary outlet tube at *C*, and the tap is opened for a few seconds to displace any air in the outlet tube and the connexion. The current of hydrogen is then stopped, the clip at *B* unscrewed, and

FIG. 2.

the solution is allowed to run from *D* into the cylinder. The clips at *B* and *C* are then screwed up tightly, the hydrogen is again turned on, and the contents of the cylinder are shaken vigorously until solution is complete. To carry out the determination, the solution is forced up into the viscosimeter bulb by closing the clip at *A* and applying suction at *B*. The clip at *B* is then closed and that at *A* opened, allowing the solution to fall in the capillary tube. The time of fall between two marks is then measured by a stop-watch. The measurement is repeated, with intervening shaking, until the results are seen to be constant, as it is sometimes difficult to tell by inspection whether solution has been complete. Brown paper covers for the cylinder and viscosimeter serve to protect the solution from the action of light. The

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viscosimeter is standardised by means of a mixture of glycerol and water; the viscosity of this mixture is then compared with that of water in a viscosimeter of the Ostwald type of smaller bore.

Effect of Air on the Viscosity of Cellulose Solutions.

The following experiment indicates the effect of air on the viscosity of cuprammonium solutions of cellulose. A solution of cotton in cuprammonium was made in the presence of hydrogen, and the hydrogen was then displaced by air.

In one case the solution was allowed to remain without shaking, and in the second the solution was shaken continuously.

Time of contact with air in hours.	Viscosity : Water = 1	
	Standing only.	With shaking.
0	73	60
$\frac{1}{2}$	73	8
1	72	4
2	63	3
4	48	2.5
10	31	—
30	16	—
40	12	—

Effect of Light on Viscosity.

In this experiment, a solution of cotton in cuprammonium was made in the presence of hydrogen, and the viscosity was measured. The solution was then exposed to light. The following determinations were made at intervals:

Initial viscosity.	1376.	Water = 1.
After exposure to bright sunlight for 10 minutes	191	
After a further $4\frac{1}{2}$ hours protected from light	186	
After further exposure for 10 minutes	84	
" " " 15 " 	56	
" " " 30 " 	38	
" " " 75 " 	27	

Use of the Falling Sphere Viscosimeter for Cuprammonium Solutions.

It appeared from the work with the hydrogen capillary viscosimeter that the falling sphere viscosimeter (see preceding paper) could be adapted for use with cuprammonium solutions of cellulose. It was necessary to use a narrower tube, 1 cm. in diameter, owing to the deep blue colour of the solution, otherwise the standard apparatus previously described was used.

As more viscous solutions could be dealt with advantageously, 2 per cent. solutions of cellulose in cuprammonium were used.

It was necessary to avoid contact with air during the solution of the cellulose. This is effected by placing 100 c.c. of cuprammonium solution with eight or nine glass beads in a stout, brown glass bottle of 150 c.c. capacity, and adding the weighed quantity of cotton. The bottle is then closed at once with an indiarubber stopper, through which a short capillary tube passes, one end being flush with the bottom of the stopper, the other end being connected by a short length of indiarubber tubing provided with a screw clip, to a second and similar bottle. The latter is closed with a rubber stopper through which pass two short glass tubes, one provided with a glass tap connecting it to the first bottle, the other connecting it by means of a short piece of pressure-tubing provided with a screw clip to the water-pump.

The connexion between the first and second bottle is closed by means of the tap, and the second bottle is exhausted by the pump; the screw clip between the second bottle and the pump is then closed and the glass tap is suddenly opened, when the rapid suction of air from the first bottle into the second causes bubbles to rise from the cuprammonium solution and to displace the air. This operation is repeated three times. The use of the second bottle ensures efficient and standard exhaustion of air without undue loss of ammonia. The screw clip attached to the first bottle, hitherto open, is now closed as tightly as possible, the bottle is detached, and immediately shaken vigorously for five minutes. The bottle is then immersed in a covered bath of water at 20° for eighteen to twenty-four hours with occasional shaking. The viscosimeter tube is filled from the bottle without access of air in the following manner. When solution is complete, the stopper is removed from the bottle without disturbing the liquid, and is replaced by a rubber stopper carrying a short inlet tube reaching to 1 cm. from the bottom of the bottle, and an outlet tube, diameter 5 mm., flush with the bottom of the stopper and about 50 cm. long, the inner end of which is capped by a cover of fine-meshed copper gauze. The viscosimeter tube, exactly 1 cm. diameter, is slipped over the long outlet tube, and the bottle is then inverted without shaking the liquid. The solution flows down into the viscosimeter tube, and, as the latter fills, the outlet tube is raised, but not above the surface of the liquid, care being taken to avoid air bubbles. With highly viscous solutions the flow is accelerated by the application of compressed air to the inlet tube of the bottle. When the tube is filled to within 3 cm. of the top the bottle is removed. The filled viscosimeter tube is then fitted

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with a rubber stopper carrying a releasing tube for the steel balls, which should be exactly central. The viscosimeter tube is then placed in a vertical position in the water-bath at 20°, and the determination made by measuring the time of fall of a 0.15 cm. steel ball through 15 cm. With this method of filling, the measurement is made through solution which has not been exposed to air.

Calculation of Absolute Viscosity.

If the mean time of fall of the ball through 15 cm. is t , the absolute viscosity, η , is given by

$$\eta = 0.42t,$$

the constant 0.42 including the correction factors. These are determined by calibration with castor oil of known viscosity, thus: Density of cellulose solution, 0.95; viscosity of standard sample of castor oil, 9.888, D 0.96; time of fall of ball through 15 cm. in 1-cm. tube, 23.4 sec.

$$\text{Constant} = \frac{\eta}{t} = \frac{\eta_1(s - \sigma)}{t_1(s - \sigma_1)} = \frac{9.888 \times 6.7}{23.4 \times 6.69} = 0.423.$$

Study of the Cuprammonium Solution for Cellulose.

Although the main difficulties in the way of trustworthy determinations of the viscosity of solutions of cellulose in cuprammonium disappeared when the precautions used to avoid the action of light and air had been adopted, minor irregularities were still sometimes obtained, and as Ost's method for the preparation of the cuprammonium solution was being followed, this was eventually suspected as the cause of trouble. It was noticed that whilst duplicate experiments carried out on the same day gave results which were quite concordant, irregular results were obtained when an interval of some weeks was allowed to elapse before the duplicate experiment was carried out. A few extreme results may be given to indicate the nature of the discrepancies.

Cotton A.	Viscosity in Ost's solution	2.22
	Viscosity 3 weeks later.....	1.50
	Viscosity 6 weeks later.....	1.72
Cotton B.	Viscosity in Ost's solution	1.23
	Viscosity 6 weeks later.....	1.50

The analysis of various solutions showed that the variations in copper content were somewhat larger than would be expected from the examples given by Ost.

The extreme figures were:

Copper, grams per litre.	Ammonia, grams per litre.
15.1	196.4
11.6	225.2

When the copper and ammonia contents were intentionally varied widely, it was found that the viscosity of a sample of cotton was affected as follows:

Composition of solution, grams per litre.		Viscosity of cotton.
Cu.	NH ₃ .	
8.47	266.9	1.64
15.33	202.9	2.52
22.84	175.1	3.54
28.40	129.7	5.44

In the preparation of Ost's solution, besides the apple-green basic copper sulphate, two distinct precipitates are obtained, one pale blue and the other dark blue. Addition of water to the solutions containing these precipitates caused the dark blue needles gradually to dissolve, but left the pale blue precipitate undissolved.

The solutions were found to contain sulphate in quantities which increased as the concentration of the ammonia decreased. The dark blue needles, described by Ost as cuprammonium hydroxide, had the same crystalline form as tetrammine copper sulphate (Found: Cu=25.84; SO₄=37.7. Calc.: Cu=25.86; SO₄=39.1 per cent.). The pale blue precipitate was free from sulphate and was apparently copper hydroxide.

The precipitate of basic copper sulphate used by Ost to prepare his solution therefore appears to dissolve in ammonia as cuprammonium hydroxide and as tetrammine copper sulphate, any excess being left as tetrammine copper sulphate crystals and copper hydroxide.

To obtain further information, the solubilities of tetrammine copper sulphate in ammonia and copper hydroxide in ammonia were determined, with the following results.

Solubility of Tetrammine Copper Sulphate at 15° in Ammonia.

Cu, grams per litre.	NH ₃ , grams per litre.
0.43	234.1
1.13	222.1
2.37	192.9
3.46	175.8
5.16	153.7
7.66	145.3
11.11	134.0

The solubility of copper hydroxide in ammonia has been previously determined by Bonsdorff (*Zeitsch. anorg. Chem.*, 1904, **41**, 132) and Dawson (T., 1909, **95**, 370). In our work, the copper hydroxide was prepared by triturating basic copper sulphate with sodium hydroxide, as described by Dawson.

Solubility of Copper Hydroxide in Ammonia.

Solution	Cu, grams per litre.	NH ₃ , grams per litre.
<i>A.</i>	11.8	195.3
1st dilution	11.6	150.5
2nd " 	11.0	132.5
3rd " 	10.4	119.6
4th " 	8.9	99.1
5th " 	7.4	85.1
6th " 	6.4	72.8
7th " 	5.2	63.7
8th " 	4.6	56.4
9th " 	2.9	38.0
10th " 	1.3	22.5
<i>B.</i>	12.2	205.2
1st dilution	12.1	176.4
2nd " 	11.6	147.2
3rd " 	9.6	112.8
4th " 	7.8	95.9
5th " 	6.4	77.7
6th " 	5.2	60.8
7th " 	4.0	46.2

The solubility of tetrammine copper sulphate in ammonia is a definite figure, since this substance is a definite crystalline compound, but the solubility of copper hydroxide seems to depend on the method of preparation adopted, and unless great precautions are taken, slight variations may be obtained even when one method of preparation is used, giving slightly variable solubility figures.

If the basic copper sulphate obtained by Ost's method behaves, on treatment with ammonia, as tetrammine copper sulphate and copper hydroxide, there will be a definite solubility of the tetrammine copper sulphate in ammonia of any particular strength and a solubility of copper hydroxide, which may vary owing to variations in the conditions of preparation of the basic copper sulphate. The use of an excess of basic copper sulphate should, therefore, give a solution in ammonia saturated with respect to tetrammine copper sulphate and with respect to copper hydroxide.

This was tested quantitatively in the following manner. A large excess of basic copper sulphate obtained by Ost's method was added to ammonia (D 0.880), and the whole kept in a thermostat at 15° in a tightly stoppered bottle for twenty-four hours and well shaken at intervals. The content of copper and ammonia in solution was then estimated. Additions of distilled water were then

made, and after each addition the solution was shaken at 15° for twenty-four hours before estimating the copper and ammonia again. Two sets of experiments were carried out, and the results were as follows:

Solution.	Cu, grams per litre.	NH ₃ , grams per litre.
Original No. 1	11.0	267.1
After 1st dilution.....	11.8	247.5
„ 2nd „	12.6	237.2
„ 3rd „	13.3	228.4
„ 4th „	15.7	207.0
„ 5th „	17.8	192.2
„ 6th „	19.4	182.0
„ 7th „	23.3	145.6
„ 8th „	26.4	129.7
Original No. 2.	11.3	237.8
After 1st dilution.....	13.5	218.0
„ 2nd „	14.6	206.7
„ 3rd „	16.7	194.4
„ 4th „	17.4	181.5
„ 5th „	17.6	171.7

The results obtained are not quite coincident, indicating the existence of different modifications of the basic copper salt, but the differences are scarcely larger than those obtained in the determination of the solubility of copper hydroxide in ammonia. This experiment confirms the impossibility of obtaining constant solutions by Ost's method, owing to the variation in solubility of copper hydroxide in ammonia.

If the solubility curves of basic sulphate, tetrammine copper sulphate, and cupric hydroxide are considered together, it will be seen (Fig. 3) that the solubility curve for basic copper sulphate represents a solution saturated with both tetrammine copper sulphate and copper hydroxide, the solubility being the sum of the solubility of tetrammine copper sulphate and of the solubility of some modification of copper hydroxide, as the general form of the curve and the value of the solubility found is consistent with this view.

The study of Ost's cuprammonium solution described above rendered it evident that the precise adjustment of this solution to definite concentration of copper and ammonia would be difficult, and as it was apparent that the value of the viscosity obtained for a sample of cotton depended to an appreciable extent on the concentration of ammonia and copper in the solution, Ost's method of preparation of the solution was abandoned.

Since copper hydroxide prepared by Dawson's method showed a fairly definite solubility in ammonia, an unsaturated solution of this form of copper hydroxide was chosen as the standard cuprammonium solution.

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The method adopted was the following. Sixty grams of copper sulphate are dissolved in 1 litre of hot water, a few drops of sulphuric acid added, the solution is allowed to cool to 50° , and ammonia (D 0.880) added until the precipitation of basic copper sulphate is complete, any excess of ammonia being neutralised with a few drops of sulphuric acid. The precipitated basic sulphate is allowed to settle, and the supernatant liquid decanted.

FIG. 3.

Solubilities in ammonia at 15° .

The precipitate is washed by decantation with hot water, 200 c.c. of 20 per cent. sodium hydroxide solution are added, and the whole is well shaken at the ordinary temperature. The precipitate is converted into blue cupric hydroxide, which is then allowed to settle, the supernatant liquid decanted, and the precipitate washed by decantation with cold water until the filtrate is free from alkali and sulphate. The precipitate is then collected, washed with distilled water, and dried on a porous plate at 40° .

The dried cupric hydroxide is transferred to an aspirator bottle and 800 c.c. of ammonia, containing 200—210 grams of ammonia per litre, are added. The whole is well shaken and the excess of cupric hydroxide allowed to settle. The supernatant liquid is run off through a glass wool filter and the volume measured. The copper in the solution is determined, and the theoretical quantity of ammonia is added to give a solution containing 11 grams of copper and 200—210 grams of ammonia per litre.

The composition of the finished solution is finally checked by analysis.

Trouble may be experienced in preparing this solution unless the conditions laid down are strictly maintained.

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RESEARCH DEPARTMENT,
ROYAL ARSENAL,
WOOLWICH.

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