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Surface active agents — Determination of interfacial tension — Drop volume method

*Agents de surface — Détermination de la tension interfaciale — Méthode au volume
de goutte*

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Reference number
ISO 9101 : 1987 (E)

Foreword

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Draft International Standards adopted by the technical committees are circulated to the member bodies for approval before their acceptance as International Standards by the ISO Council. They are approved in accordance with ISO procedures requiring at least 75 % approval by the member bodies voting.

International Standard ISO 9101 was prepared by Technical Committee ISO/TC 91, *Surface active agents*.

Users should note that all International Standards undergo revision from time to time and that any reference made herein to any other International Standard implies its latest edition, unless otherwise stated.

Surface active agents — Determination of interfacial tension — Drop volume method

0 Introduction

Interfacial tension is a fundamental property of the interface between two immiscible or partially miscible liquids. These two liquids may contain surface active agents which reduce the interfacial tension, but it is generally not possible to predict their detergent and emulsifying activities from measurements of the interfacial tension alone.

1 Scope and field of application

This International Standard specifies a test method, by measuring the drop volume, for the determination of the interfacial tension between two liquid phases.

It is particularly suitable for the measurement of the interfacial tension between two solutions of surface active agents in aqueous or organic solvents, and provides some advantages compared with the method by drawing up liquid films (ISO 6889), which are the following :

- determination of interfacial tension between two liquid phases containing all types of surface active agents, including cationic surface active agents;
- accurate measurement of low interfacial tensions of the order of 1 mN/m;
- repeatability standard deviation less than $\pm 0,5$ mN/m (acceptable for control in industrial laboratories);
- determination of interfacial tension of viscous liquids;
- determination of interfacial tension on only small amounts of the solution;
- time dependence of the interfacial tension can be measured using a simple automatic apparatus.

2 References

ISO 758, *Liquid chemical products for industrial use — Determination of density at 20 °C.*

ISO 862, *Surface active agents — Vocabulary.*

ISO 2456, *Surface active agents — Water used as a solvent for tests — Specifications and test methods.*

ISO 6889, *Surface active agents — Determination of interfacial tension by drawing up liquid films.*

3 Definition

interfacial tension : See ISO 862.

NOTE — The SI unit of interfacial tension is the newton per metre (N/m). In practice, the sub-multiple millinewton per metre (mN/m) is used.

4 Principle

Measurement of the volume of a drop of an aqueous phase formed at the end of a vertical capillary tube when it detaches from the tube in contact with the organic phase.

Obtaining the interfacial tension between two liquid phases by balancing the weight of the drop with the interfacial tension force supporting it and applying a correction factor; then calculation of the interfacial tension from the volume of the falling drop, the outer capillary radius, the density difference between the two liquid phases and the acceleration due to gravity.

5 Apparatus

5.1 Measuring instrument (see figure 1) comprising the following elements.

5.1.1 Glass syringe, accurately calibrated, of 0,5 cm³ volume, driven by a micrometer screw so graduated that the volume of the falling drop may be calculated to an accuracy of $\pm 0,000$ 1 cm³.

5.1.2 Capillary, glass or stainless steel, prepared as in 5.3, connected to the orifice of the syringe, and removable for cleaning.

5.1.3 Jacketed glass vessel having an inside diameter of 2,5 to 4 cm and a height of 5 cm or more, capable of being thermoregulated to the test temperature and of containing the syringe and the capillary.

5.2 Mounting of the apparatus

The syringe (5.1.1) and capillary (5.1.2) shall be mounted on a stand which allows movement in a vertical direction; the tip of the capillary shall be perpendicular to its axis, of uniform diameter and with an edge free from defects.

The stand is attached to a metal plate placed on a vibration-free table. Manual operation of the micrometer screw may introduce vibration so it is preferable to use a small electric motor to drive the micrometer screw.

If a d.c. motor is used, provision shall be made for the movement of the micrometer screw and its speed to be adjusted electronically, and care shall be taken to eliminate overrunning when the motor is switched off. Alternatively, a step motor may be used and the vertical displacement measured electronically.

As the circulation of water may introduce some vibrations, the thermostat shall be switched off as the drop approaches its maximum volume.

5.3 Preparation of capillary tips

a) Glass capillary

From good quality, small-bore glass capillary tubing of suitable diameter, cut a length of about 4 cm and attach to one end a ground conical 5/13 glass socket. Accurately grind the other end of the capillary on a piece of plane glass covered with moistened emery powder. To prevent rocking while polishing, embed the capillary in paraffin wax (with softening point about 70 °C) contained in a glass tube of about 1,5 cm inside diameter. The tip shall be plane and normal to the axis of the capillary; its circumference shall be undamaged.

b) Stainless steel capillary

From good quality, small-bore stainless steel capillary tubing of suitable diameter, cut a length of about 4 cm, cutting one of the ends accurately at right angles to the axis of the tube using a precision tool lathe; at the other end, fit a junction piece made of polytetrafluoroethylene.

Depending on the value of the interfacial tension and the density difference, the outer radius of the capillary will lie between 0,1 and 0,4 cm.

5.4 Thermoregulated bath capable of being controlled to within $\pm 0,5$ °C.

6 Procedure

6.1 Preparation of solution of surface active agents

6.1.1 The solutions of surface active agents for the determination shall be prepared with all necessary care. The water used for their preparation shall be double-distilled water complying with ISO 2456, checked by measuring its surface tension. Cork stoppers and, more particularly, rubber stoppers, shall on no account be used, either in the construction of the distillation apparatus or for sealing containers in which the water is kept.

6.1.2 The temperature of the solution shall be maintained to within 0,5 °C.

NOTE — Determinations carried out close to temperatures of critical solubility, such as the Krafft temperature or cloud point temperature of non-ionic surface active agents, are seriously affected by errors. It is preferable to carry out the test at a temperature higher than such temperatures or at a temperature lower than the cloud point temperature of non-ionic surface active agents.

The method provides less accurate values with solutions of surface active agents having very low concentrations, i.e. lower than 10^{-4} mol/l, unless the drop formation rate is further decreased.

6.1.3 As the interfacial tension of solutions varies with time, due to the mutual saturation of the liquid phases, and as the adsorption of surface active agents onto the interface are not instantaneous phenomena, it is difficult to recommend a standard age for the interface. It is, therefore, desirable to make several measurements over a period of time in order to obtain a curve for the interfacial tension as a function of time, and from this to determine the position of the plateau, which gives the age at which the interface reached its stationary state.

6.1.4 The surface of liquids is extremely sensitive to contamination by atmospheric dust or by vapours from solvents handled in the vicinity. Therefore no volatile products shall be handled in the room where the determinations are carried out, and all the apparatus shall be protected by a bell of the kind used for balances. These precautions also reduce variation in temperature.

6.1.5 The recommended method of taking the test portion of the liquid phases to be examined is to suck it, with a pipette, from the centre of the mass of these phases, as the surface may be liable to soiling by insoluble particles and dust.

6.2 Cleaning the measuring apparatus (5.1)

In the presence of impurities such as silicones, which are not removed by sulfo-chromic acid mixture, phosphoric acid or a solution of potassium persulfate in sulfuric acid, clean the glass cylinder of the syringe and the capillary and also the jacketed vessel using special products (for example toluene, perchloroethylene or methanolic or ethanolic potassium hydroxide solution). To minimize contamination, always rinse the syringe with the aqueous phase.

If these impurities are absent, or after cleaning with these products, wash the measuring apparatus carefully with a hot solution of potassium or ammonium persulfate in sulfuric acid. Finally, rinse in double-distilled water until the rinsings are neutral. The double-distilled water shall be freshly prepared in accordance with ISO 2456.

Before the determination, the measuring apparatus shall be completely dried.

6.3 Check of the measuring apparatus

6.3.1 Measurement of capillary radius

Measure the radius to better than 0,1 % using a travelling microscope. Take the mean of 5 to 10 measurements.

NOTE — The radius may also be obtained from accurate measurement of the maximum drop volume using a liquid of known density and interfacial tension, with the help of tables (see the annex). Use this method only when a travelling microscope or a micrometer is not available.

6.3.2 Measurement of drop volume

To obtain accurately the volume of the detached drop, check, by weighing some drops, the indicated reading obtained from the movement of the micrometer screw.

6.4 Determination

Fill the syringe (5.1.1), connected to the appropriate capillary (5.1.2) and chosen as in 6.5, with the aqueous phase and introduce the organic phase into the jacketed vessel (5.1.3).

When the density of the aqueous phase is greater than that of the organic phase, immerse the capillary in the organic phase to a depth greater than 0,5 cm.

To ensure complete wetting, completely cover the capillary tip by a drop of the aqueous phase before immersing it in the organic phase.

When the density of the aqueous phase is less than that of the organic phase, the tip shall face upwards and its orifice should be about 2 cm below the surface to allow the lighter drop to form.

Before measuring the static interfacial tension of aqueous surface active agent solutions (i.e. interfacial tension, which is not time dependent), first bring the two liquids to the desired temperature, using the thermoregulated bath (5.4), and then slowly form the drop, with a volume less than the maximum volume, at the capillary tip in 30 to 60 s. The drop should remain for 2 to 5 min whilst its volume slowly increases. Finally, the drop should become detached, on further slight enlargement.

If the drop detaches without enlargement, choose a smaller initial volume. If the enlargement is greater than 0,001 cm³, the initial volume was too small.

NOTE — Measurements with increasing age of drops (2, 5, 8 min, etc.) may thus be carried out until the volume of the detached drop remains constant. Static interfacial tension can be determined more conveniently by means of an automatic apparatus (see clause 9).

Enlarge the drop continuously at different speeds until the detached drop volume remains constant (see example of measurement in figure 2) and record the volume V determined from reading values on the scale of the micrometer screw.

A full syringe enables several measurements to be made depending on the value of the interfacial tension.

Take the mean of at least four measurements of volume of the detached drop, excluding the volume of the first drop, for calculating the interfacial tension.

6.5 Selection of capillary tips

Harkins and Brown^[1] have determined the correction factor f given in the formula (see 7.1) for different values of the ratio $r/V^{1/3}$ (r and V being defined in 7.1). The most accurate values of f lie in the region of

$$0,65 < r/V^{1/3} < 0,95$$

but the wider region of

$$0,3 < r/V^{1/3} < 1,2$$

is also acceptable.^[6]

However, in practice, the value of $r/V^{1/3}$ may lie between 0,3 and 1,6 or even greater.

To choose a capillary which will form drops of the test solution in the desired region of $r/V^{1/3}$, use figure 3 which gives $r/V^{1/3}$ as a function of the outer capillary radius for different values of the capillary constant k , expressed in centimetres to the power minus two and defined by the formula $\Delta\rho g/\gamma$ ($\Delta\rho$ and g being defined in 7.1).

Using the test solution and a capillary of arbitrary outer radius r , measure a drop volume V .

From r and $r/V^{1/3}$, read the capillary constant k from figure 3.

By following the appropriate curve of k in figure 3, the range of values of r in the desired region of $r/V^{1/3}$ is obtained.

In practice, interfacial tensions between 1 and 30 mN/m can usually be measured with capillaries of radius 0,2 cm. A radius of 0,1 cm may be used for lower interfacial tensions, and 0,3 cm or greater for higher interfacial tensions.

7 Expression of results

7.1 Method of calculation

The interfacial tension, γ , expressed in millinewtons per metre, is given by the formula

$$\frac{V \Delta\rho g}{2\pi r f}$$

where

V is the volume, in cubic centimetres, of the falling drop;

$\Delta\rho$ is the density difference, in grams per cubic centimetre, at the temperature of the determination, between the two liquid phases, measured according to ISO 758;

g is the acceleration due to gravity, equal to 981 cm·s⁻²;

r is the outer radius, in centimetres, of the capillary used;

f is the correction factor corresponding to the ratio $r/V^{1/3}$ given in tables 1 and 2.

NOTE — Values of f are given in table 1 for values of $r/V^{1/3}$ between 0,65 and 0,95 in steps of 0,001.

In table 2, values are given in the region $0,3 < r/V^{1/3} < 1,2$, also in steps of 0,001. The values in table 1 are more accurate than those in table 2, so the former should be used whenever possible.

7.2 Precision

The accuracy in the determination of the interfacial tension results from errors in V , $\Delta\rho$, r and f , and may be estimated to better than 1 mN/m. With care it may be reduced to about 1 %.

The precision of the measurements can be improved significantly by limiting vibrations, by temperature control and by mutual presaturation of the liquids.

8 Test report

The test report shall include the following information :

- a) all information necessary for the complete identification of the test liquid phases, particularly the surface active agent concentration and the densities;
- b) the reference of the method used (reference to this International Standard); also the outer radius of the capillary tip used, and the measuring method used [microscope or standard liquids (see the annex)];
- c) the nature of the water or the solvent used and the concentration of the solutions;
- d) the temperature of the determination;
- e) the time of formation and the age of the drop before detachment;
- f) the individual and mean values of the measured drop volumes, and the measured interfacial tensions;

g) any operating details not specified in this International Standard or in the International Standards to which reference is made, or regarded as optional, as well as any incidents liable to have affected the results.

9 Bibliography

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Annex

Determination of capillary radius with liquids of known density and interfacial tension

(This annex does not form part of the standard.)

Using a capillary of unknown radius, accurately measure the maximum drop volume with liquids of tabulated density and interfacial tension. From this volume V and the capillary constant k , given in table 3, calculate the minimum volume $V_0 = V k^{3/2}$. The corresponding value of $r/V^{1/3}$ is read from table 4. The capillary radius is calculated from the value of $r/V^{1/3}$ and the measured maximum drop volume V .

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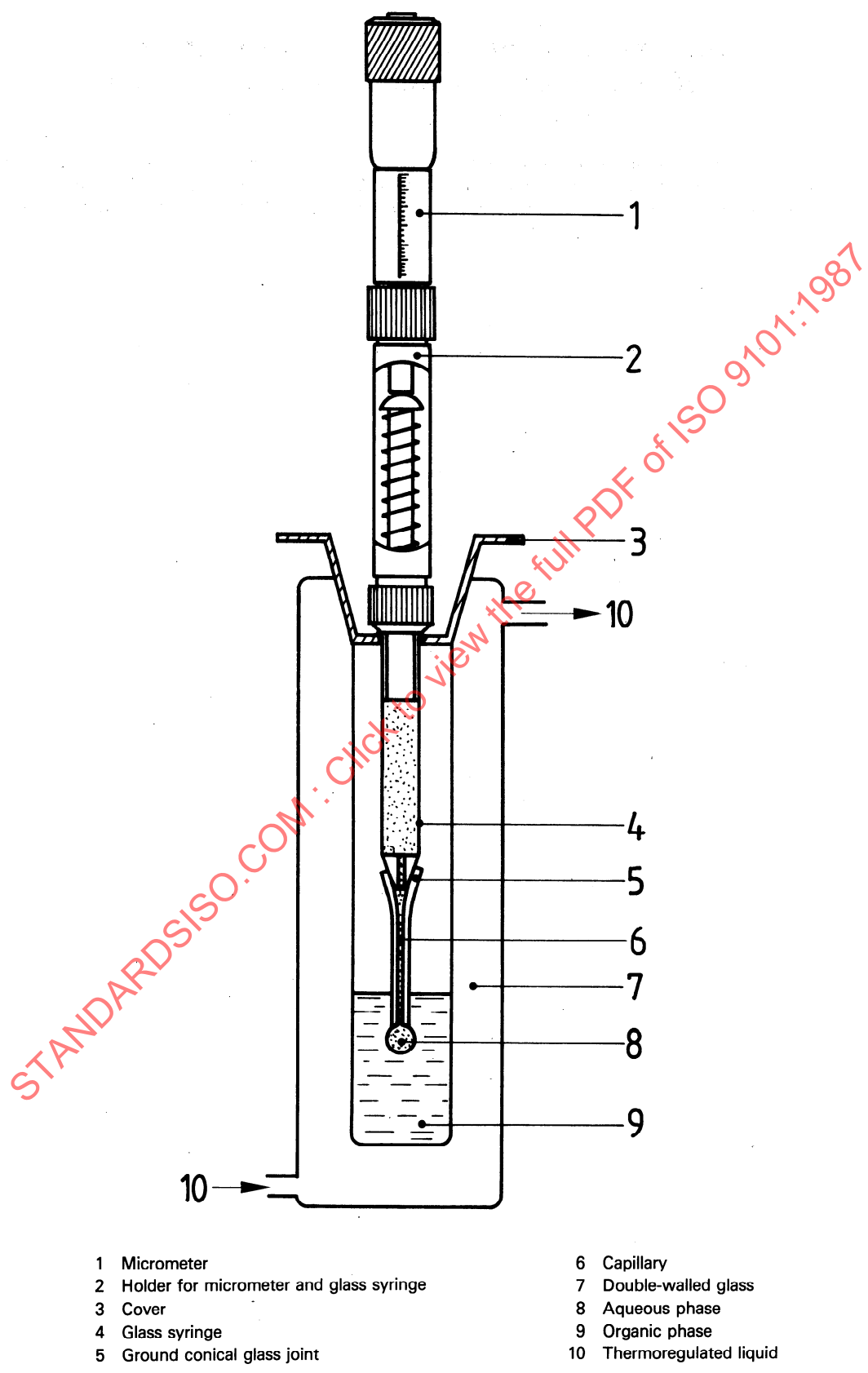


Figure 1 — Apparatus for measuring the interfacial tension by the drop volume method

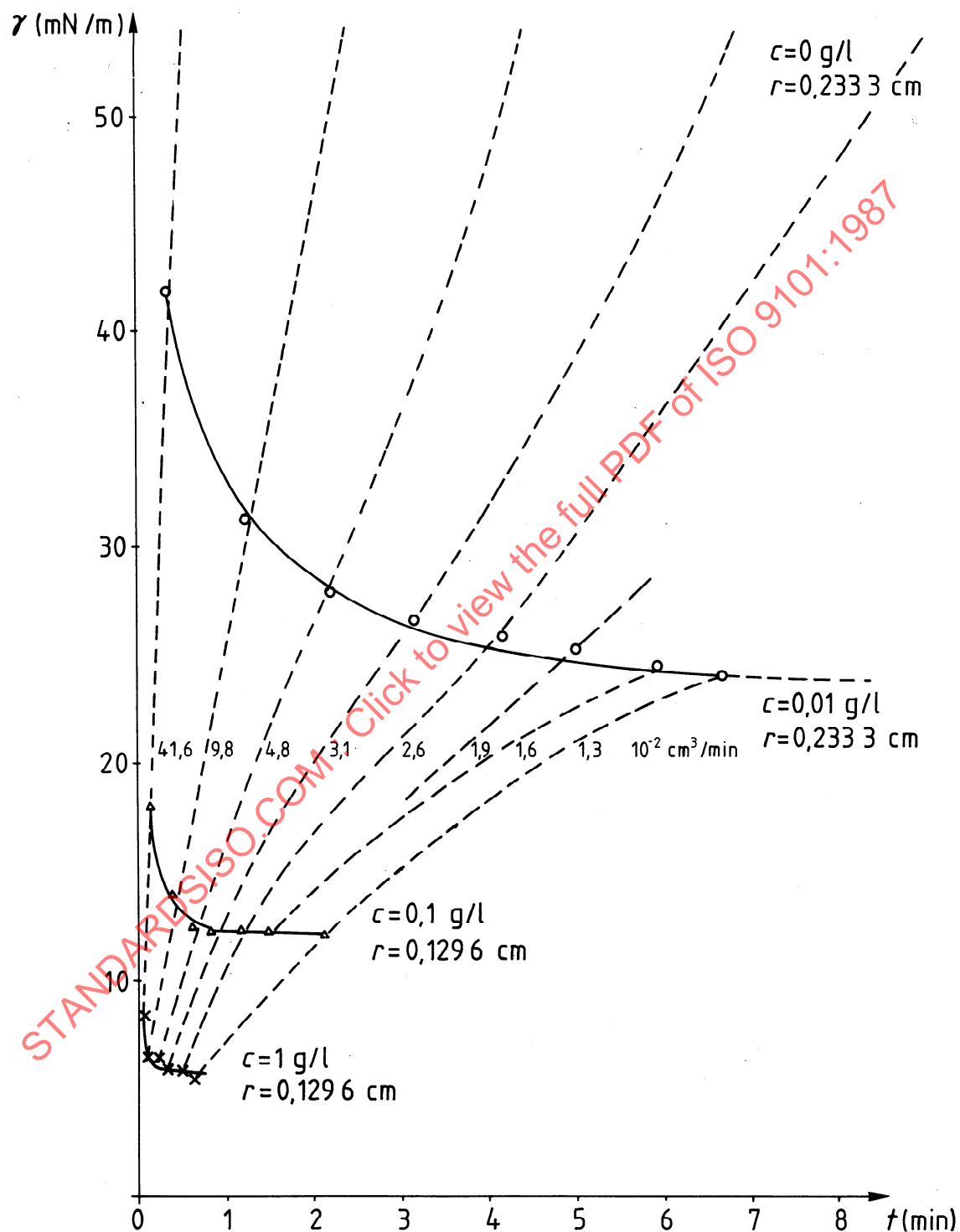


Figure 2 — Influence of the life of the drop on the interfacial tension for the system water/paraffin oil/surfactant (0 to 1 g/l)
(The drop formation rate is also shown : 0,013 to 0,416 cm³/min.)

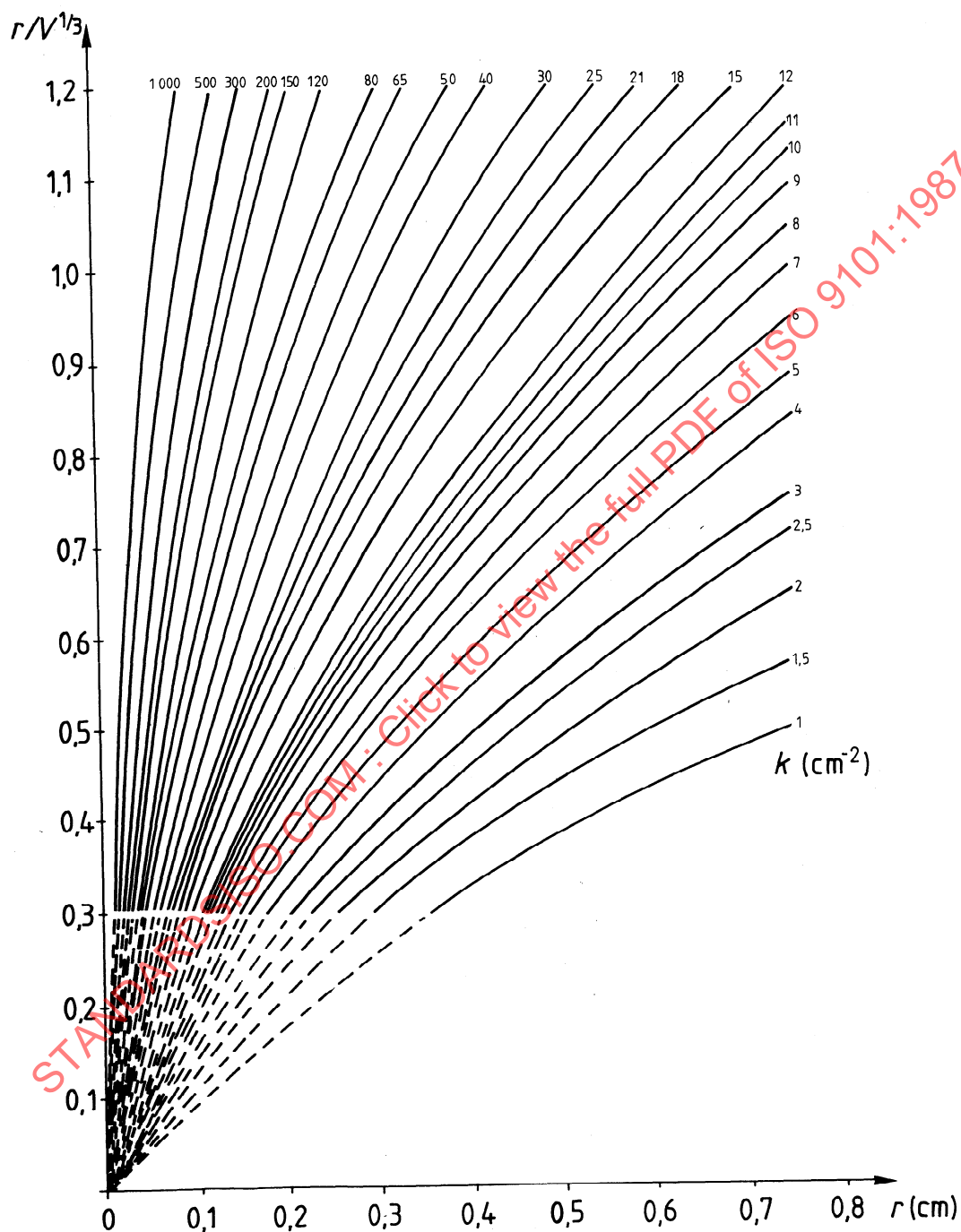


Figure 3 — Variation of capillary radius r (in centimetres) with the ratio $r/V^{1/3}$ for different values of $k = \Delta\rho g/\gamma$ (in centimetres to the power minus two) for $0,3 < r/V^{1/3} < 1,2$ (full curves)

Table 1 — Values of correction factor f for different values of the ratio $r/V^{1/3}$ between 0,65 and 0,95 in steps of 0,001^[5]

$r/V^{1/3}$	0	1	2	3	4	5	6	7	8	9
0,65	0,617 2	0,617 1	0,616 9	0,616 7	0,616 7	0,616 3	0,616 2	0,616 0	0,615 8	0,615 6
0,66	0,615 5	0,615 3	0,615 1	0,614 9	0,614 8	0,614 6	0,614 4	0,614 3	0,614 1	0,613 9
0,67	0,613 8	0,613 6	0,613 4	0,613 3	0,613 1	0,613 0	0,612 8	0,612 6	0,612 5	0,612 3
0,68	0,612 2	0,612 0	0,611 9	0,611 7	0,611 5	0,611 4	0,611 2	0,611 1	0,610 9	0,610 8
0,69	0,610 7	0,610 5	0,610 4	0,610 2	0,610 1	0,609 9	0,609 8	0,609 6	0,609 5	0,609 4
0,70	0,609 2	0,609 1	0,609 0	0,608 8	0,608 7	0,608 6	0,608 4	0,608 3	0,608 2	0,608 0
0,71	0,607 9	0,607 8	0,607 6	0,607 5	0,607 4	0,607 3	0,607 1	0,607 0	0,606 9	0,606 8
0,72	0,606 7	0,606 5	0,606 4	0,606 3	0,606 2	0,606 1	0,606 0	0,605 8	0,605 7	0,605 6
0,73	0,605 5	0,605 4	0,605 3	0,605 3	0,605 1	0,605 0	0,604 9	0,604 8	0,604 7	0,604 5
0,74	0,604 4	0,604 3	0,604 2	0,604 1	0,604 0	0,603 9	0,603 9	0,603 9	0,603 7	0,603 6
0,75	0,603 5	0,603 4	0,603 3	0,603 2	0,603 1	0,603 0	0,602 9	0,602 8	0,602 8	0,602 7
0,76	0,602 6	0,602 5	0,602 4	0,602 3	0,602 3	0,602 2	0,602 1	0,602 0	0,602 0	0,601 9
0,77	0,601 8	0,601 7	0,601 7	0,601 6	0,601 5	0,601 4	0,601 4	0,601 3	0,601 2	0,601 2
0,78	0,601 1	0,601 0	0,601 0	0,600 9	0,600 8	0,600 8	0,600 7	0,600 7	0,600 6	0,600 6
0,79	0,600 5	0,600 4	0,600 4	0,600 3	0,600 3	0,600 2	0,600 2	0,600 1	0,600 1	0,600 0
0,80	0,600 0	0,599 9	0,599 9	0,599 8	0,599 8	0,599 7	0,599 7	0,599 7	0,599 6	0,599 6
0,81	0,599 5	0,599 5	0,599 5	0,599 4	0,599 4	0,599 4	0,599 3	0,599 3	0,599 3	0,599 2
0,82	0,599 2	0,599 2	0,599 2	0,599 1	0,599 1	0,599 1	0,599 0	0,599 0	0,599 0	0,599 0
0,83	0,599 0	0,598 9	0,598 9	0,598 9	0,598 9	0,598 9	0,598 9	0,598 8	0,598 8	0,598 8
0,84	0,598 8	0,598 8	0,598 8	0,598 8	0,598 8	0,598 8	0,598 7	0,598 7	0,598 7	0,598 7
0,85	0,598 7	0,598 7	0,598 7	0,598 7	0,598 7	0,598 7	0,598 7	0,598 7	0,598 7	0,598 7
0,86	0,598 8	0,598 8	0,598 8	0,598 8	0,598 8	0,598 8	0,598 8	0,598 8	0,598 8	0,598 9
0,87	0,598 9	0,598 9	0,598 9	0,598 9	0,598 9	0,599 0	0,599 0	0,599 0	0,599 0	0,599 0
0,88	0,599 1	0,599 1	0,599 1	0,599 1	0,599 3	0,599 2	0,599 2	0,599 3	0,599 3	0,599 3
0,89	0,599 4	0,599 4	0,599 4	0,599 5	0,599 5	0,599 5	0,599 6	0,599 6	0,599 7	0,599 7
0,90	0,599 7	0,599 8	0,599 8	0,599 9	0,599 9	0,600 0	0,600 0	0,600 1	0,600 1	0,600 2
0,91	0,600 2	0,600 3	0,600 3	0,600 4	0,600 4	0,600 5	0,600 5	0,600 6	0,600 7	0,600 7
0,92	0,600 8	0,600 8	0,600 9	0,601 0	0,601 0	0,601 1	0,601 2	0,601 2	0,601 3	0,601 4
0,93	0,601 4	0,601 5	0,601 6	0,601 6	0,601 7	0,601 8	0,601 9	0,601 0	0,602 0	0,602 1
0,94	0,602 2	0,602 3	0,602 3	0,602 4	0,602 5	0,602 6	0,602 7	0,602 8	0,602 8	0,602 9

Table 2 — Values of correction factor f for different values of the ratio $r/V^{1/3}$ between 0,3 and 1,2 in steps of 0,001^[6]

$r/V^{1/3}$	0	1	2	3	4	5	6	7	8	9
0,300	0,726 3	0,725 8	0,725 3	0,724 8	0,724 3	0,723 8	0,723 4	0,722 9	0,722 4	0,721 9
0,310	0,721 4	0,720 9	0,720 4	0,720 0	0,719 5	0,719 0	0,718 5	0,718 0	0,717 6	0,717 1
0,320	0,716 6	0,716 1	0,715 7	0,715 2	0,714 7	0,714 3	0,713 8	0,713 3	0,712 9	0,712 4
0,330	0,711 9	0,711 5	0,711 0	0,710 6	0,710 1	0,709 7	0,709 2	0,708 8	0,708 3	0,707 9
0,340	0,707 4	0,707 0	0,706 5	0,706 1	0,705 6	0,705 2	0,704 7	0,704 3	0,703 9	0,703 4
0,350	0,703 0	0,702 5	0,702 1	0,701 7	0,701 2	0,700 8	0,700 4	0,700 0	0,699 5	0,699 1
0,360	0,698 7	0,698 2	0,697 8	0,697 4	0,697 0	0,696 6	0,696 1	0,695 7	0,695 3	0,694 9
0,370	0,694 5	0,694 1	0,693 7	0,693 2	0,692 8	0,692 4	0,692 0	0,691 6	0,691 2	0,690 8
0,380	0,690 4	0,690 0	0,689 6	0,689 2	0,688 8	0,688 4	0,688 0	0,687 6	0,687 2	0,686 8
0,390	0,686 4	0,686 0	0,685 7	0,685 3	0,684 9	0,684 5	0,684 1	0,683 7	0,683 3	0,683 0
0,400	0,682 6	0,682 2	0,681 8	0,681 4	0,681 1	0,680 7	0,680 3	0,679 9	0,679 6	0,679 2
0,410	0,678 8	0,678 4	0,678 1	0,677 7	0,677 3	0,677 0	0,676 6	0,676 2	0,675 9	0,675 5
0,420	0,675 2	0,674 8	0,674 4	0,674 1	0,673 7	0,673 4	0,673 0	0,672 6	0,672 3	0,672 0
0,430	0,671 6	0,671 3	0,670 9	0,670 6	0,670 2	0,669 9	0,669 5	0,669 2	0,668 8	0,668 5
0,440	0,668 2	0,667 8	0,667 5	0,667 1	0,666 8	0,666 5	0,666 1	0,665 8	0,665 5	0,665 1
0,450	0,664 8	0,664 5	0,664 2	0,663 8	0,663 5	0,663 2	0,662 9	0,662 5	0,662 2	0,661 9
0,460	0,661 6	0,661 2	0,660 9	0,660 6	0,660 3	0,660 0	0,659 7	0,659 3	0,659 0	0,658 7
0,470	0,658 4	0,658 1	0,657 8	0,657 5	0,657 2	0,656 9	0,656 6	0,656 2	0,655 9	0,655 6
0,480	0,655 3	0,655 0	0,654 7	0,654 4	0,654 1	0,653 8	0,653 5	0,653 3	0,653 0	0,652 7
0,490	0,652 4	0,652 1	0,651 8	0,651 5	0,651 2	0,650 9	0,650 6	0,650 3	0,650 1	0,649 8
0,500	0,649 5	0,649 2	0,648 9	0,648 6	0,648 4	0,648 1	0,647 8	0,647 5	0,647 3	0,647 0
0,510	0,646 7	0,646 4	0,646 2	0,645 9	0,645 6	0,645 3	0,645 1	0,644 8	0,644 5	0,644 3
0,520	0,644 0	0,643 7	0,643 5	0,643 2	0,642 9	0,642 6	0,642 4	0,642 2	0,641 9	0,641 6
0,530	0,641 4	0,641 1	0,640 9	0,640 6	0,640 4	0,640 1	0,639 9	0,639 6	0,639 4	0,639 1
0,540	0,638 9	0,638 6	0,638 4	0,638 1	0,637 9	0,637 6	0,637 4	0,637 1	0,636 9	0,636 7
0,550	0,636 4	0,636 2	0,635 9	0,635 7	0,635 5	0,635 2	0,635 0	0,634 8	0,634 5	0,634 3
0,560	0,634 1	0,633 8	0,633 6	0,633 4	0,633 1	0,632 9	0,632 7	0,632 5	0,632 2	0,632 0
0,570	0,631 8	0,631 6	0,631 3	0,631 1	0,630 9	0,630 7	0,630 5	0,630 3	0,630 0	0,629 8
0,580	0,629 6	0,629 4	0,629 2	0,629 0	0,628 8	0,628 5	0,628 3	0,628 1	0,627 9	0,627 7
0,590	0,627 5	0,627 3	0,627 1	0,626 9	0,626 7	0,626 5	0,626 3	0,626 1	0,625 9	0,625 7
0,600	0,625 5	0,625 3	0,625 1	0,624 9	0,624 7	0,624 5	0,624 3	0,624 1	0,623 9	0,623 7
0,610	0,623 5	0,623 3	0,623 1	0,623 0	0,622 8	0,622 6	0,622 4	0,622 2	0,622 0	0,621 8
0,620	0,621 7	0,621 5	0,621 3	0,621 1	0,620 9	0,620 8	0,620 6	0,620 4	0,620 2	0,620 0
0,630	0,619 9	0,619 7	0,619 5	0,619 4	0,619 2	0,619 0	0,618 8	0,618 7	0,618 5	0,618 3
0,640	0,618 2	0,618 0	0,617 8	0,617 7	0,617 5	0,617 3	0,617 2	0,617 0	0,616 9	0,616 7
0,650	0,616 5	0,616 4	0,616 2	0,616 1	0,615 9	0,615 8	0,615 6	0,615 4	0,615 3	0,615 1
0,660	0,615 0	0,614 8	0,614 7	0,614 7	0,614 5	0,614 2	0,614 1	0,613 9	0,613 8	0,613 7
0,670	0,613 5	0,613 4	0,613 2	0,613 1	0,612 9	0,612 8	0,612 7	0,612 5	0,612 4	0,612 3
0,680	0,612 1	0,612 0	0,611 8	0,611 7	0,611 6	0,611 4	0,611 3	0,611 2	0,611 1	0,610 9
0,690	0,610 8	0,610 7	0,610 5	0,610 4	0,610 3	0,610 2	0,610 0	0,609 9	0,609 8	0,609 7
0,700	0,609 5	0,609 4	0,609 3	0,609 2	0,609 1	0,609 0	0,608 8	0,608 7	0,608 6	0,608 5
0,710	0,608 4	0,608 3	0,608 2	0,608 0	0,607 9	0,607 8	0,607 7	0,607 6	0,607 5	0,607 4
0,720	0,607 3	0,607 2	0,607 1	0,607 0	0,606 9	0,606 8	0,606 7	0,606 6	0,606 5	0,606 4
0,730	0,606 3	0,606 2	0,606 1	0,606 0	0,605 9	0,605 8	0,605 7	0,605 6	0,605 5	0,605 4
0,740	0,605 3	0,605 2	0,605 2	0,605 1	0,605 0	0,604 9	0,604 8	0,604 7	0,604 6	0,604 6
0,750	0,604 5	0,604 4	0,604 3	0,604 2	0,604 1	0,604 1	0,604 0	0,603 9	0,603 8	0,603 8
0,760	0,603 7	0,603 6	0,603 5	0,603 5	0,603 4	0,603 3	0,603 2	0,603 2	0,603 1	0,603 0
0,770	0,603 0	0,602 9	0,602 8	0,602 8	0,602 7	0,602 6	0,602 6	0,602 5	0,602 5	0,602 4
0,780	0,602 3	0,602 3	0,602 2	0,602 2	0,602 1	0,602 0	0,602 0	0,601 9	0,601 9	0,601 8
0,790	0,601 8	0,601 7	0,601 7	0,601 6	0,601 6	0,601 5	0,601 5	0,601 4	0,601 4	0,601 3
0,800	0,601 3	0,601 3	0,601 2	0,601 2	0,601 1	0,601 1	0,601 0	0,601 0	0,601 0	0,600 9
0,810	0,600 9	0,600 9	0,600 8	0,600 8	0,600 7	0,600 7	0,600 7	0,600 7	0,600 6	0,600 6
0,820	0,600 6	0,600 5	0,600 5	0,600 5	0,600 5	0,600 4	0,600 4	0,600 4	0,600 4	0,600 3
0,830	0,600 3	0,600 3	0,600 3	0,600 3	0,600 2	0,600 2	0,600 2	0,600 2	0,600 2	0,600 2
0,840	0,600 1	0,600 1	0,600 1	0,600 1	0,600 1	0,600 1	0,600 1	0,600 1	0,600 1	0,600 1
0,850	0,600 0	0,600 0	0,600 0	0,600 0	0,600 0	0,600 0	0,600 0	0,600 0	0,600 0	0,600 0
0,860	0,600 0	0,600 0	0,600 0	0,600 0	0,600 0	0,600 1	0,600 1	0,600 1	0,600 1	0,600 1
0,870	0,600 1	0,600 1	0,600 1	0,600 1	0,600 1	0,600 2	0,600 2	0,600 2	0,600 2	0,600 2
0,880	0,600 2	0,600 3	0,600 3	0,600 3	0,600 3	0,600 3	0,600 4	0,600 4	0,600 4	0,600 4
0,890	0,600 5	0,600 5	0,600 5	0,600 6	0,600 6	0,600 6	0,600 6	0,600 7	0,600 7	0,600 7
0,900	0,600 8	0,600 8	0,600 9	0,600 9	0,600 9	0,601 0	0,601 0	0,601 1	0,601 1	0,601 1
0,910	0,601 2	0,601 2	0,601 3	0,601 3	0,601 4	0,601 4	0,601 5	0,601 5	0,601 6	0,601 6
0,920	0,601 7	0,601 7	0,601 8	0,601 8	0,601 9	0,601 9	0,602 0	0,602 0	0,602 1	0,602 1
0,930	0,602 2	0,602 3	0,602 3	0,602 4	0,602 5	0,602 5	0,602 6	0,602 7	0,602 7	0,602 8