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EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

**LIQUID METALS FOR HEAT-PIPES, PROPERTIES,
PLOTS AND DATA SHEETS**

by

H.E.J. SCHINS

1967

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SUMMARY

For the calculation of the heat transfer in a heat-pipe many liquid and vapor data of the working fluid (metal) are required. These data are required in the region of the boiling point.

Measurements were made for the twelve most current metals of the vapor pressure and the surface tension. For calcium, strontium and barium density measurements were also made.

The vapor pressure measurements were made with the new method of Bohdanský, using the heat-pipe effect in an open tube. This method is described elsewhere. Surface tension and density measurements were made by the maximum bubble pressure method. The vapor pressure measurements make it possible to evaluate the boiling point, the heat of vaporization and the vapor density.

Data sheets are presented for surface tension, vapor pressure and vapor density. Data sheets for density, viscosity and vapor viscosity selected from the literature are also presented.

It is shown that it is still impossible to predict a certain property theoretically with adequate accuracy.

KEYWORDS

HEAT TRANSFER
HEAT PIPES
SURFACE TENSION
CALCIUM
BARIUM
STRONTIUM

METALS
DENSITY
MEASUREMENT
BOILING POINTS
EVAPORATION HEAT
VISCOSITY

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LIQUID METALS FOR HEAT-PIPES, PROPERTIES, PLOTS
AND DATA SHEETS

1. INTRODUCTION (*)

Heat-pipes (1) are simple structures made for the conduction of gross amounts of heat from one end to the other of the pipe, with a negligible drop in temperature along it. Working conditions are e.g. the transfer of some kilowatts at a working temperature of 1600°C in a tube of one inch diameter. Heat-pipes consist of closed cylindrical refractory metal tubes with some special internal structure and contain a small amount of low melting metal. At the working temperature the low melting metal serves as the heat-transfer medium. For a mathematical treatment of the heat-conduction many liquid metal data are required.

Most of the experimental data in the past were taken in the neighbourhood of the melting point. The working temperature of the heat-pipe however is near to the boiling point of the liquid metal and because of this fact, we were obliged to repeat several measurements at higher temperatures, especially vapor pressure and surface tension measurements.

This was the more urgent where extrapolations are mostly difficult because of the wide scattering in both absolute values and temperature dependencies of the data by earlier investigators.

Herewith data sheets are presented for the current liquid metals used in heat-pipes: lithium, sodium, potassium, rubidium, caesium, silver, calcium, strontium, barium, thallium, lead and bismuth.

The atomic diameters calculated from the atomic volume were taken from Zwicker (132). The melting points are from Nesmeyanov (17). This author gives also reliable data for boiling points and heats of vaporization but we preferred our own measurements (5) as they are more consistent.

For the critical temperatures we refer to the publications of Grosse and collaborators (9).

The liquid densities were found by averaging the most reliable values, only in the case of calcium, strontium and barium our values were preferred. The liquid viscosity plots were chosen after examination of the data in literature on a $\log \eta$ versus $1/T$ scale. As to the surface tension we give our own values which mostly agree to others when extrapolated to the melting temperature range. This was not the case for barium and calcium. For rubidium and caesium no other values were found.

(*) Manuscript received on September 8, 1967.

A summary of densities can be found in McGonigal (10) and Strauss (65). Of viscosities in Grosse (12, 22), Chapman (61), Strauss (13), Menz-Sauerwald (96), Budde (133) and Nikolskiy (134). Of surface tensions in Taylor (18, 28), Grosse (11), Strauss (66), Zadumkin (19) and Flint (104).

Metals of interest here are also treated by Liquid Metals Handbook (23), Stull and Sinke (25), Bainton (38), Spiller (51, Na, K), Dunning (55, Na), Sittig (92, Na), Lemmon (131, K), Ellis (114, Li), Weatherford (130) and Kutateladze (156).

As to the vapor pressure we preferred our data to Nesmeyanov's (17) because ours were taken in the boiling point region. Mostly they agree to extrapolations of low or very low temperature regions. For the calculation of the ideal gas density we used these vapor pressures.

For the calculation of the vapor viscosity we used a formula put forward by Hirschfelder (125). We found that it fitted to the measured values of mercury (126, 127) and caesium (120) when we put in the characteristic temperatures from Chapman (61), and the atomic diameter of Zwikker (132).

Thus we give an abundant evaluation for liquid density and viscosity, but for surface tension, vapor pressure and vapor density we rely on our own values and mention only the references.

2. Experimental Data

2.1. <u>Lithium</u>	Value / Formula	References
Atomic number	3	
Atomic weight	6,94	
Atomic diameter	2,80 Å	132
Melting point	454°K	17
Boiling point	1613°K	5
Critical temp.	4110°K; 3540°K	10,93
Heat of vapor.	35,4 Cal/mol	5
Density	$0,555-0,934 \cdot 10^{-4} \cdot T \text{ g/cm}^3$	36,67,83,134
Viscosity	$1,42 \exp 1309/RT \text{ mP}$	36,100,101,134
Surface tension	$453-0,148 T \text{ dyne/cm}$	6,83,109
Vapor pressure	$\log P = 7,67-7740/T \text{ torr}$	4,5,158
Vapor density	$1,113 \cdot 10^{-4} P/T \text{ (P in torr)}$	
approx. formula	$3,72-7740/T - \log T$	
Vapor viscosity	$9,0 \cdot 10^{-6} \sqrt{T/\Omega}^{(2,2)*} \text{ poise}$	125,61
approx. formula	$1,20 \cdot 10^{-7} \cdot T - 6,0 \cdot 10^{-6}$	

2.1.1. The viscosity of lithium

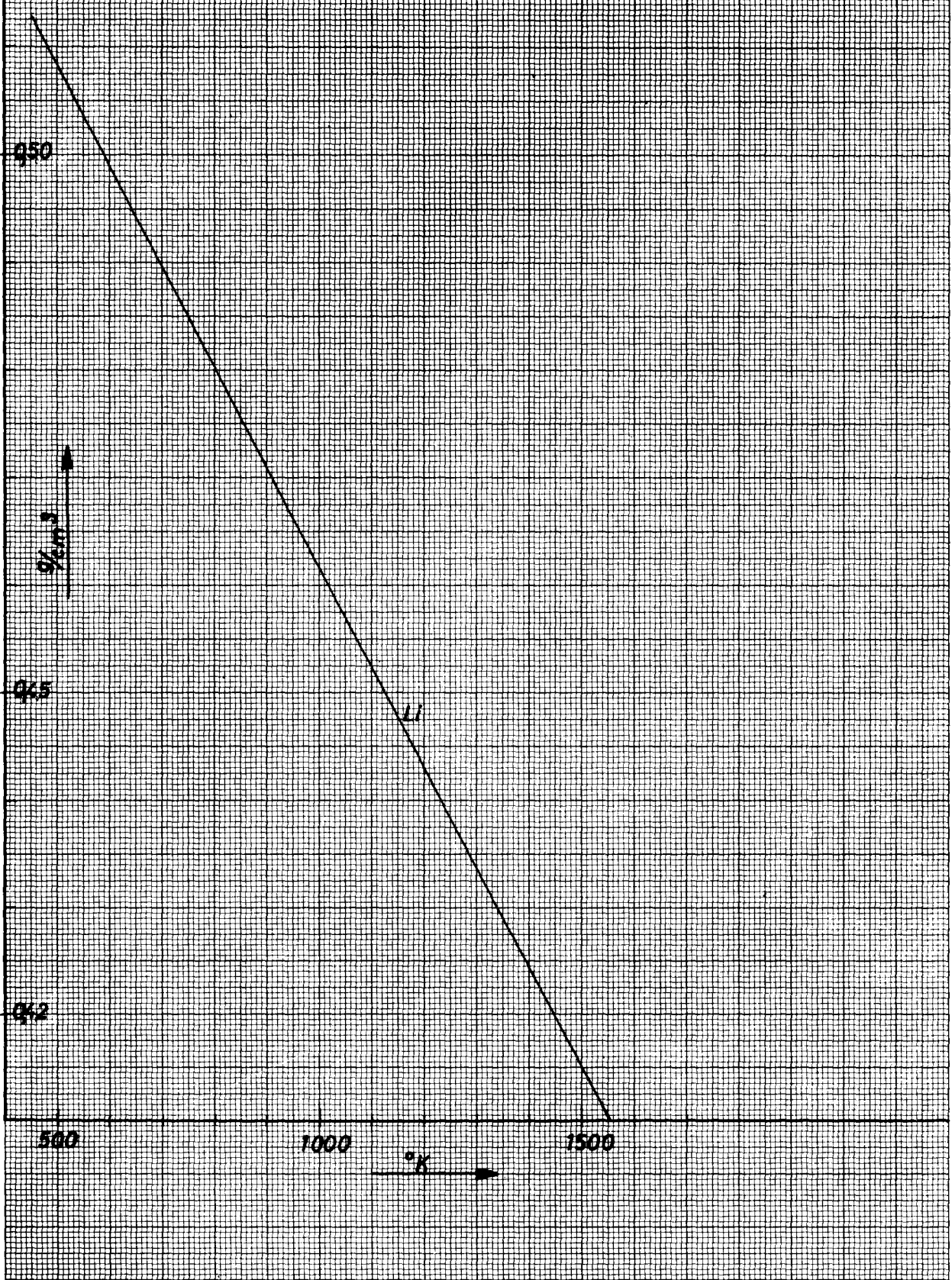
The measurements of Anadrade (36) and Novikov (101) fall together and also the lowest temperature measurements of Rigney (100) fall on this regression line. The higher temperature measurements of Rigney fall clearly away from this curve. Nikolskiy (134) gives the same values as Novikov.

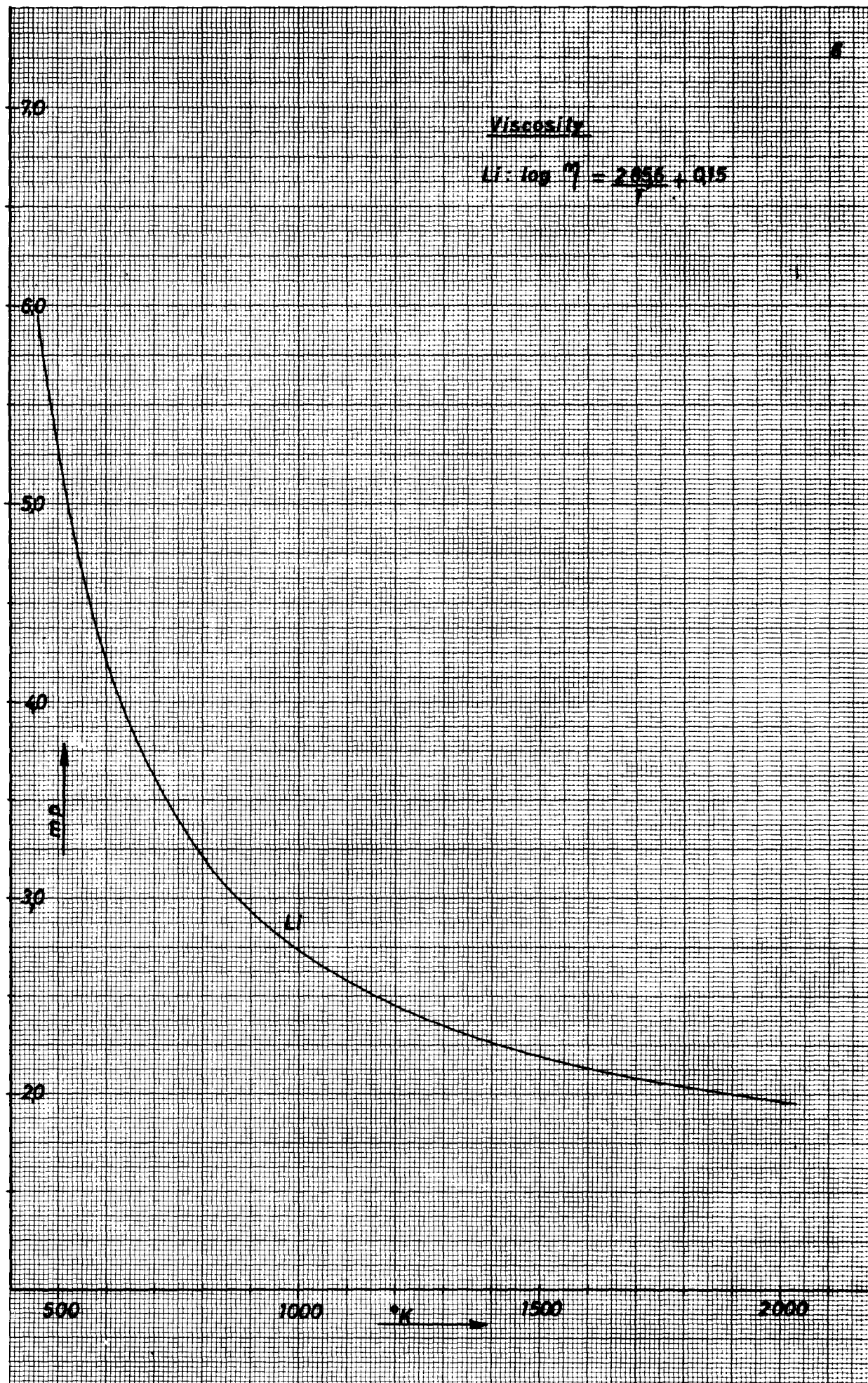
2.1.2. The specific weight of lithium

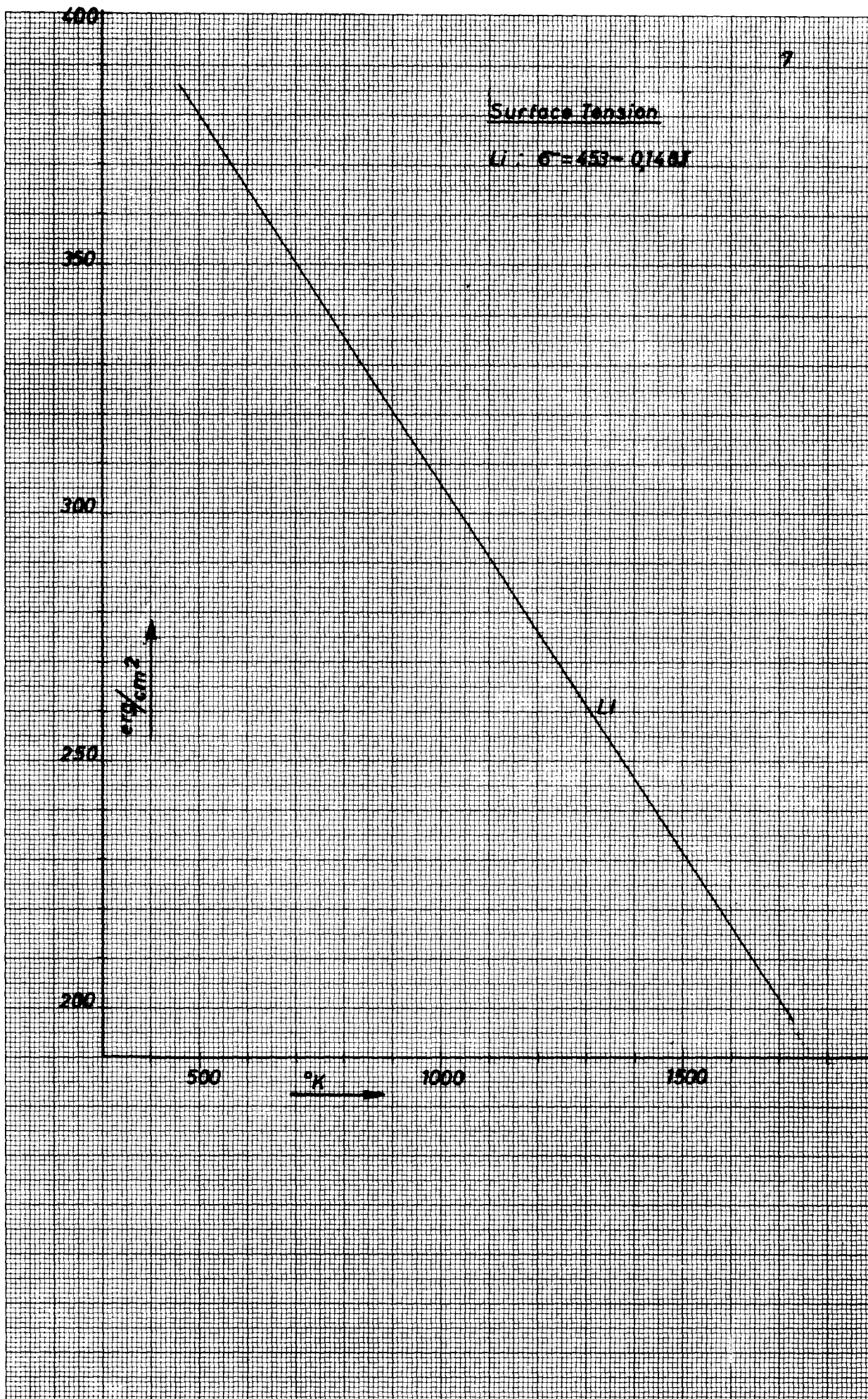
Author	Year	Method	Max. Temp.	Formula
Bernini see (36)	1914	dilatometric	230°C	$0,5567-0,9913 \cdot 10^{-4} T$
(67)	1950	Max Bubble P.	1600°C	$0,5454-0,82 \cdot 10^{-4} T$
(83)	1955	Max Bubble P.	500°C	$0,547-0,855 \cdot 10^{-4} T$
(134)	1959	-	-	$0,564-1,00 \cdot 10^{-4} T$

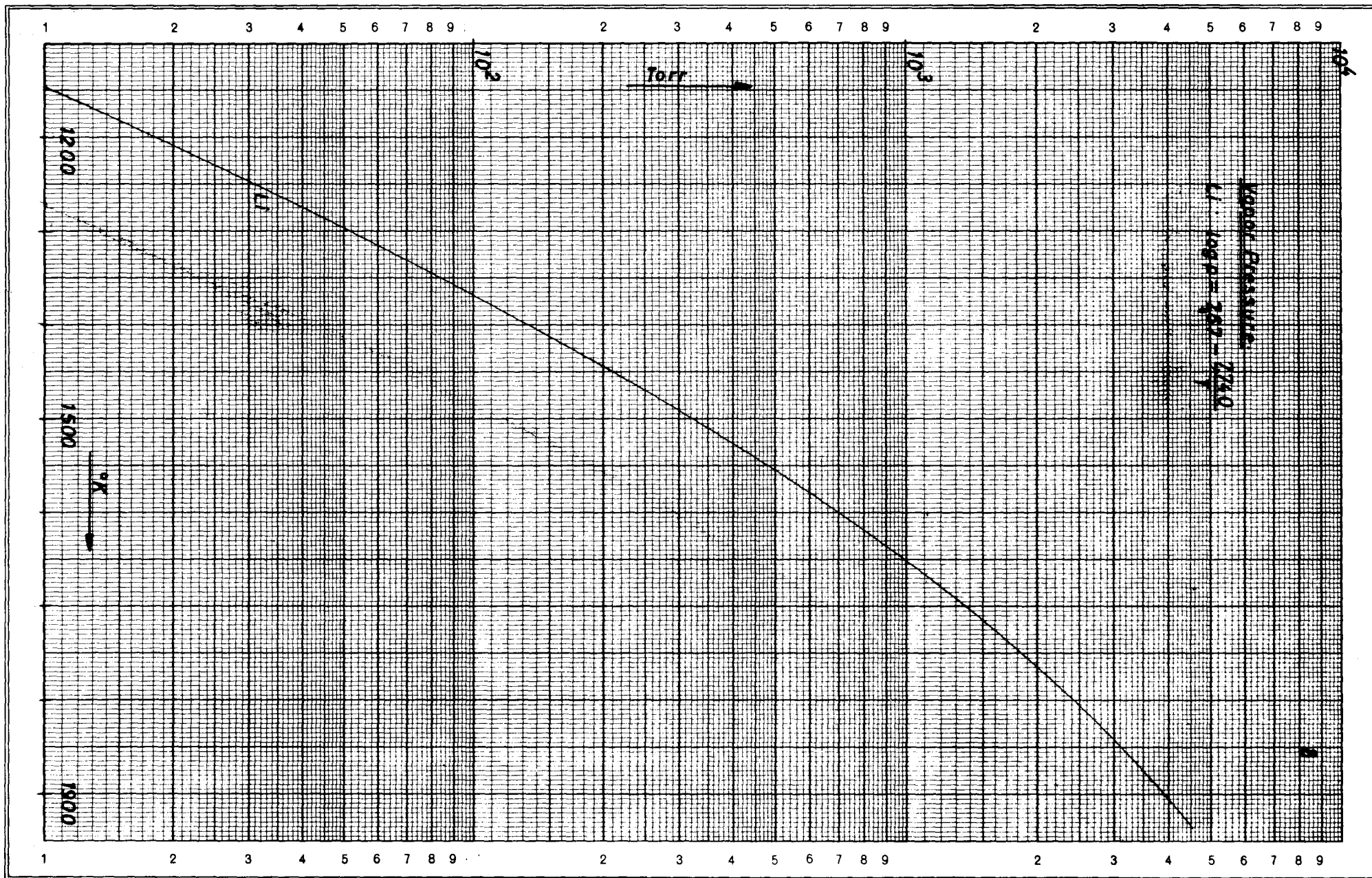
Specific Weight

$\gamma = 0.555 - 0.0004 \cdot T$

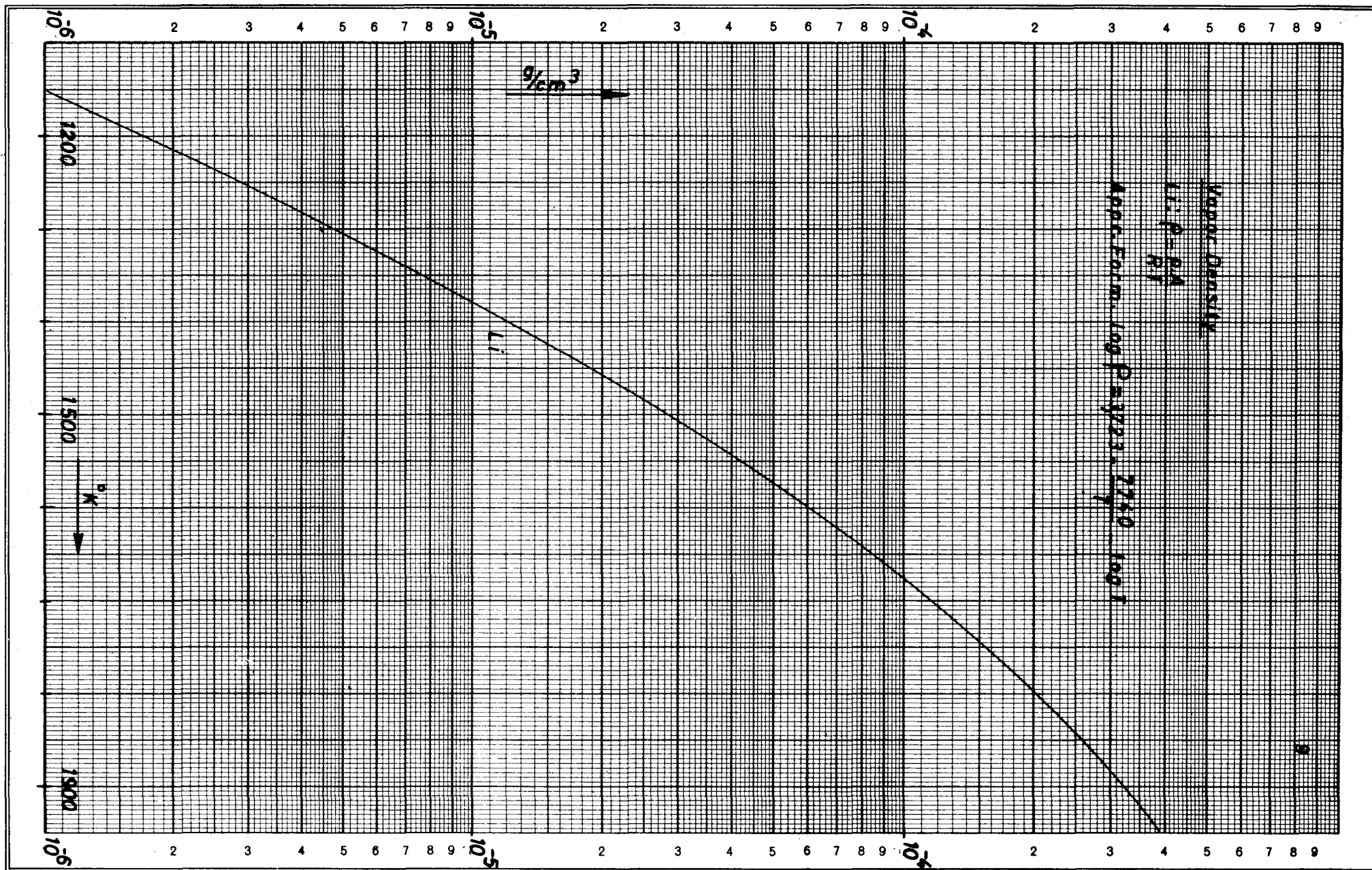




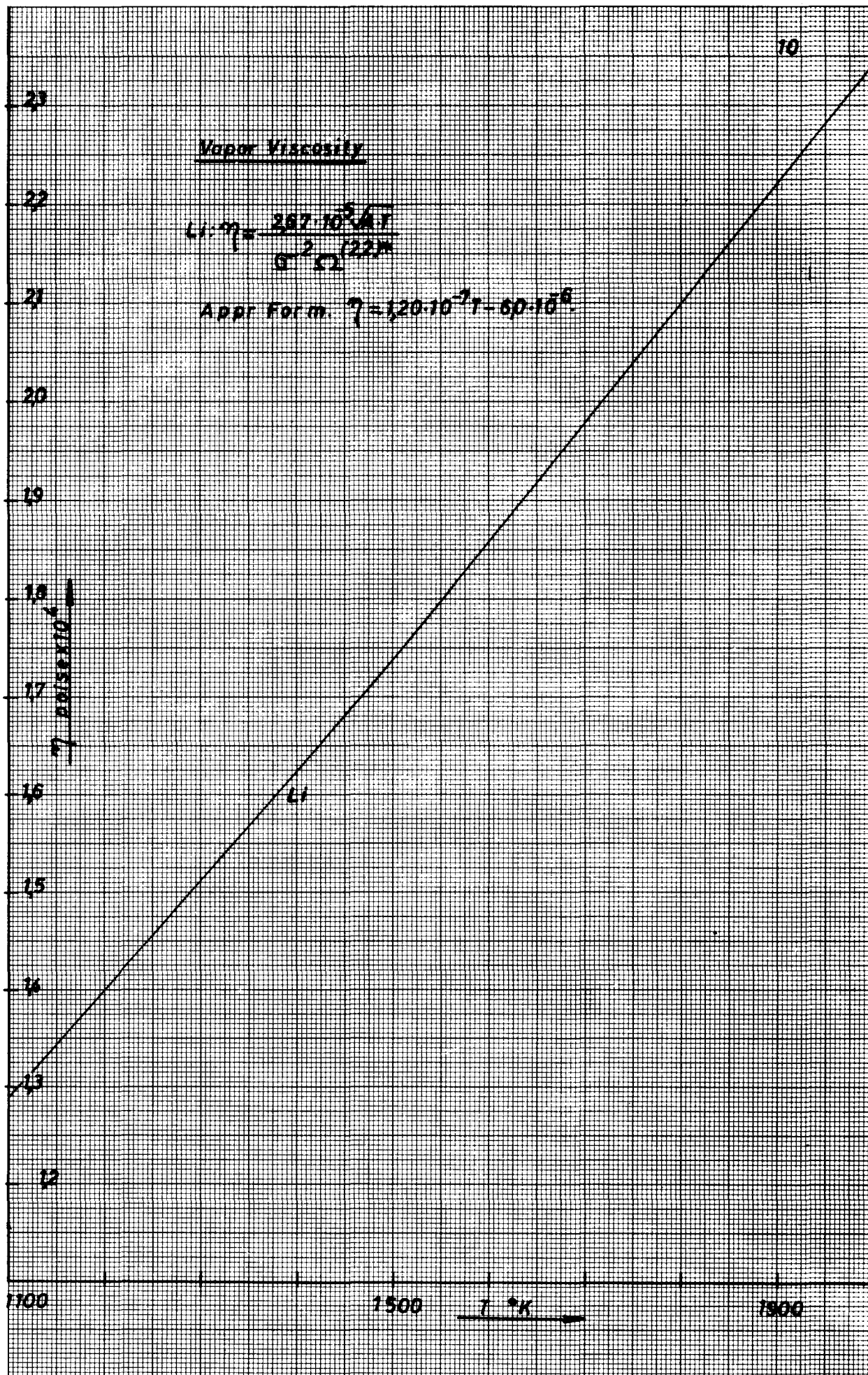




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2.2. <u>Sodium</u>	Value / Formula	References
Atomic number	11	
Atomic weight	22,991	
Atomic diameter	3,40 Å	132
Melting point	372°K	17
Boiling point	1156°K	5, 52, 53
Critical temp.	2800°K; 2600°K	9, 157
Heat of vapor.	24,2 Cal/mol	5
Density	$1,018-2,34 \cdot 10^{-4} \cdot T \text{ g/cm}^3$	54, 56, 93, 94, 101, 117, 134
Viscosity	$0,893 \exp 1517/RT \text{ mP}$	54, 57, 101, 102, 103
Surface tension	$220-0,091 T \text{ dyne/cm}$	6, 56, 151, 152
Vapor pressure	$\log P = 7,46-5290/T \text{ torr}$	5, 50, 137, 158, 159
Vapor density	$3,683 \cdot 10^{-4} P/T \text{ (P in torr)}$	93, 130
approx. formula	$4,03-5290/T - \log T$	
Vapor viscosity	$1,102 \cdot 10^{-5} \sqrt{T/\Omega}^{(2,2)*} \text{ poise}$	125, 61
approx. formula	$1,60 \cdot 10^{-7} \cdot T - 5,0 \cdot 10^{-6}$	

2.2.1. The viscosity of sodium

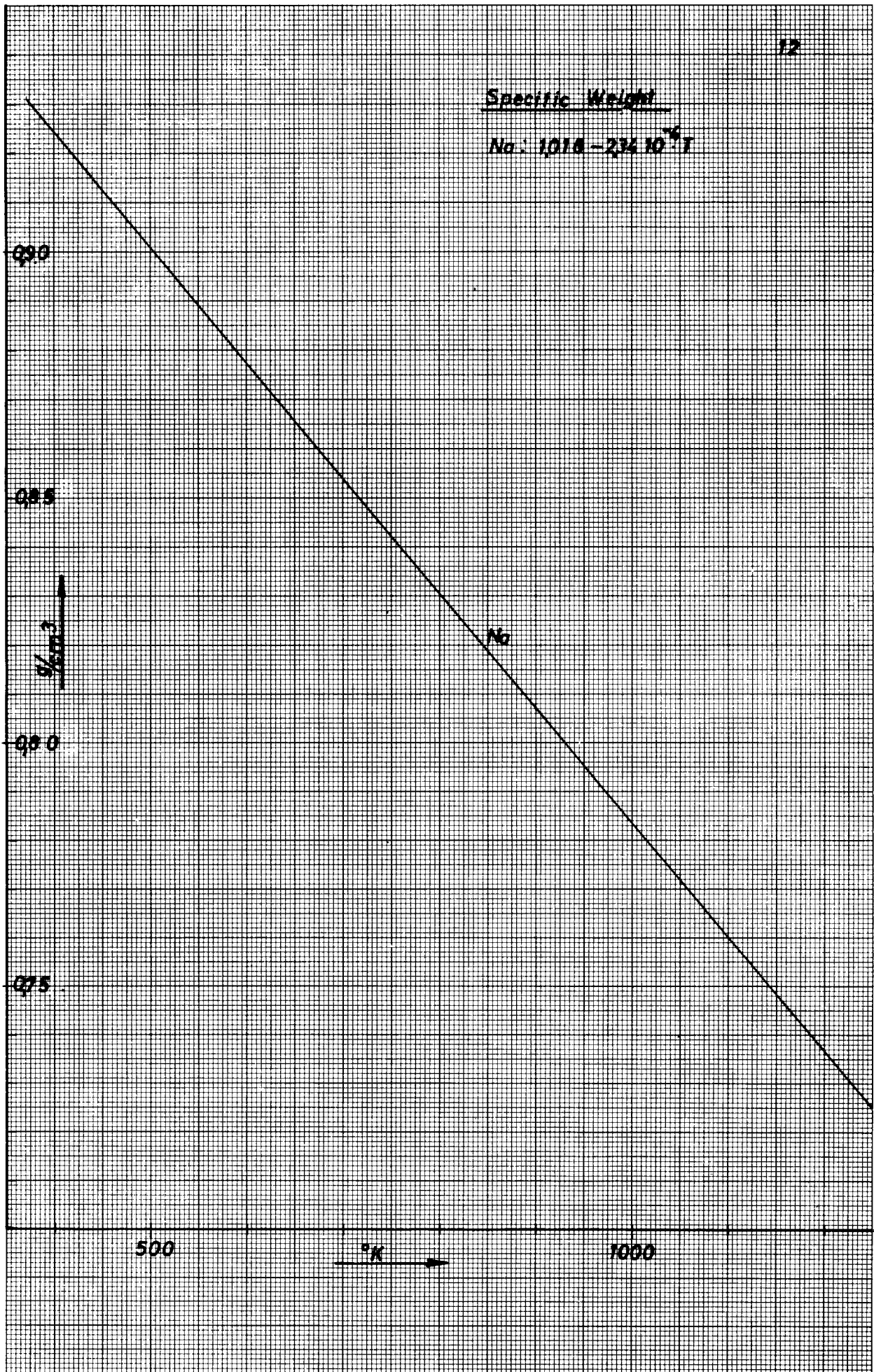
There is no doubt about the viscosity of sodium as all the measured values fall on a perfectly straight line.

2.2.2. The specific weight of sodium

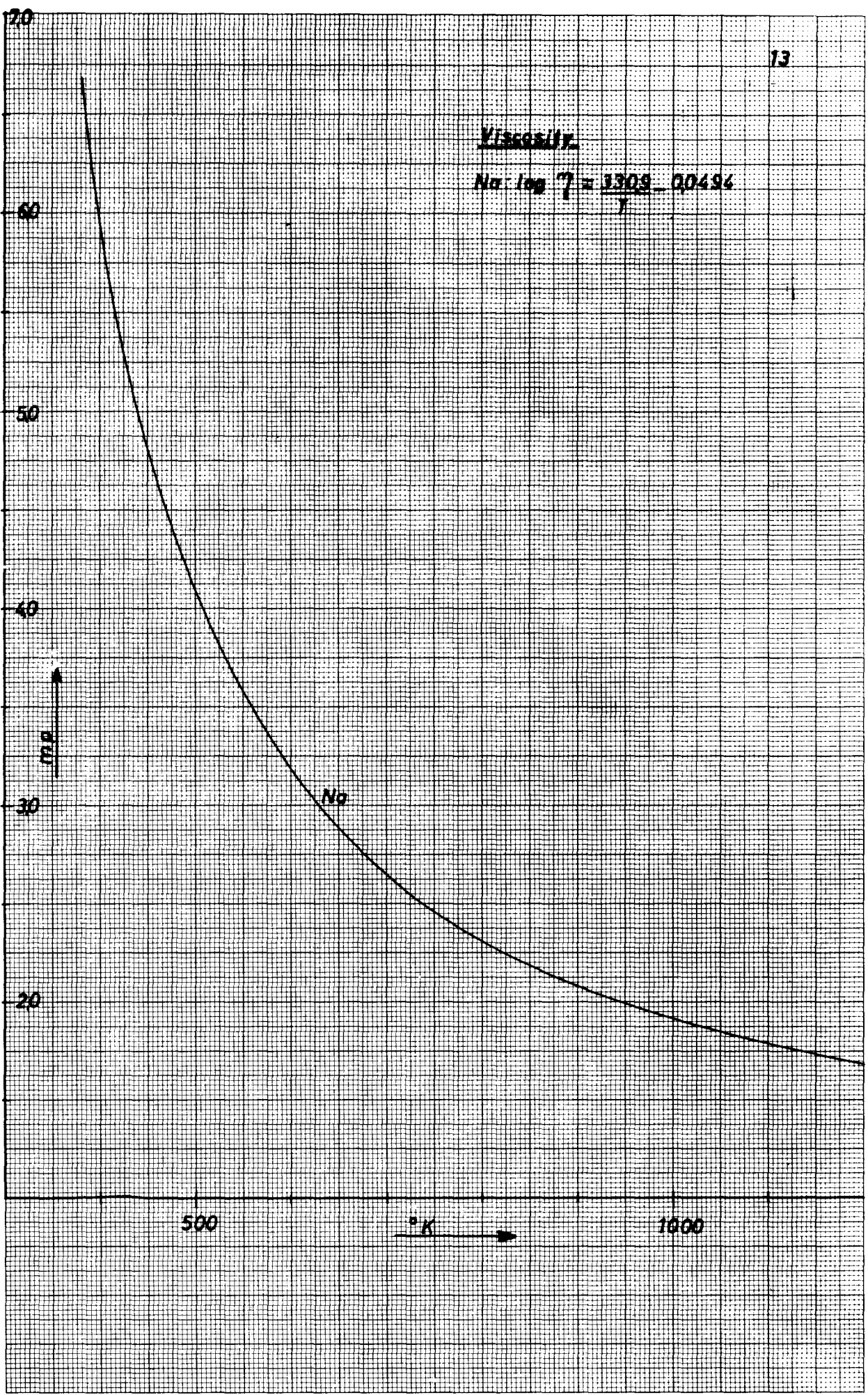
Author	Year	Method	Max. Temp.	Formula
(116)	1951	Pycnometer	193°C	$1,0127-2,322 \cdot 10^{-4} T$
(56, 117)	1953	Max. Bubble P.	500°C	$0,99-1,9 \cdot 10^{-4} T$
Hagen see (92)	1956	dilatometric	-	$1,010-2,23 \cdot 10^{-4} T$
(51)	1963	Survey	-	$1,039-2,60 \cdot 10^{-4} T$
(51)	1963	"	-	$1,017-2,415 \cdot 10^{-4} T$
(93)	1965	"	-	$1,020-2,40 \cdot 10^{-4} T$
(93)	1965	γ-radiation	1260°C	$1,0456-2,57 \cdot 10^{-4} T$
(94)	1966	Pycnometer	1000°C	$1,0127-2,292 \cdot 10^{-4} T$
(101)	1957	Archimedian	750°C	$1,009-2,21 \cdot 10^{-4} T$
(134)	1959	-	600°C	$1,020-2,47 \cdot 10^{-4} T$

Specific Weight

No: 1016-234 10⁴ T

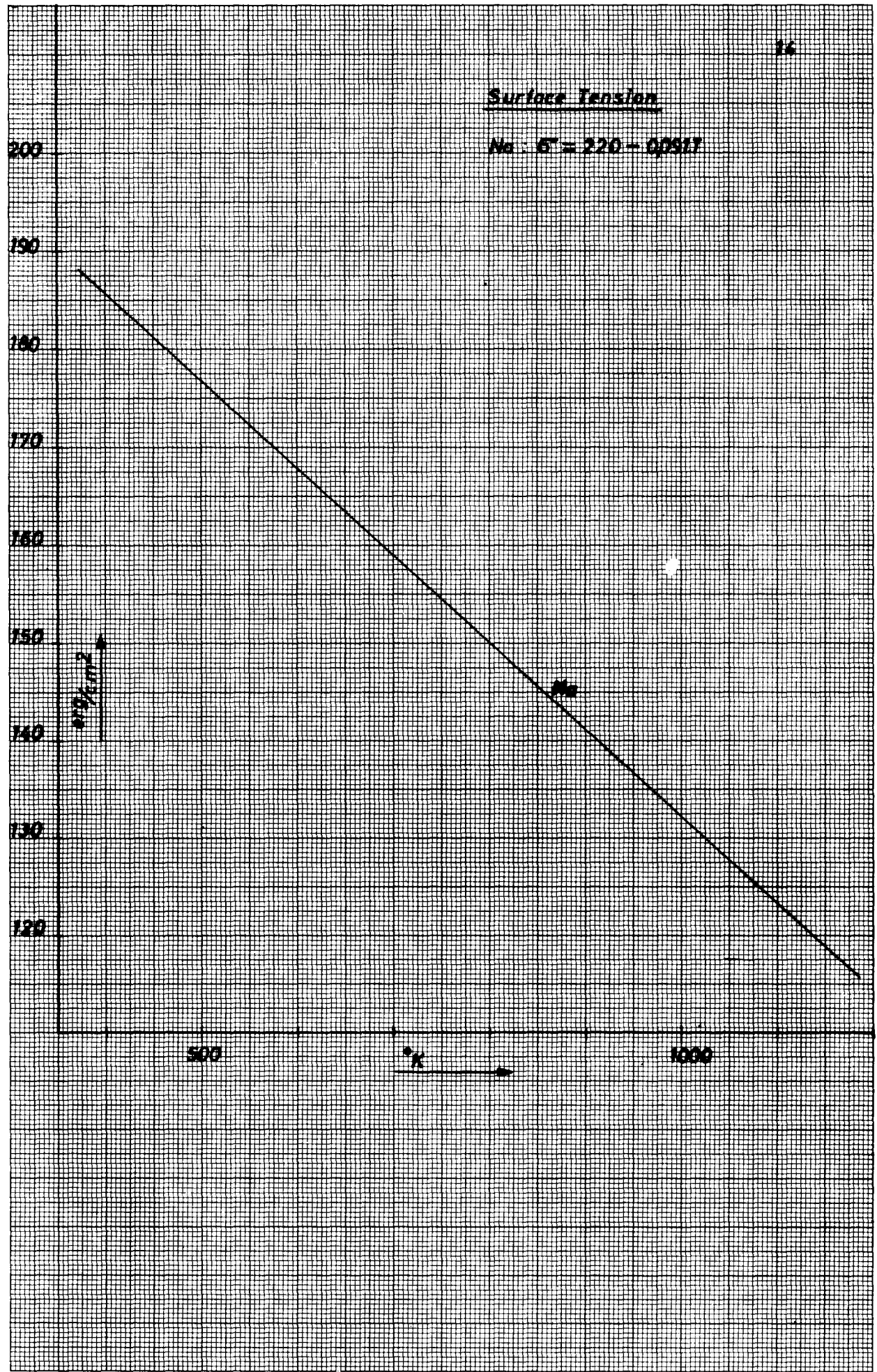


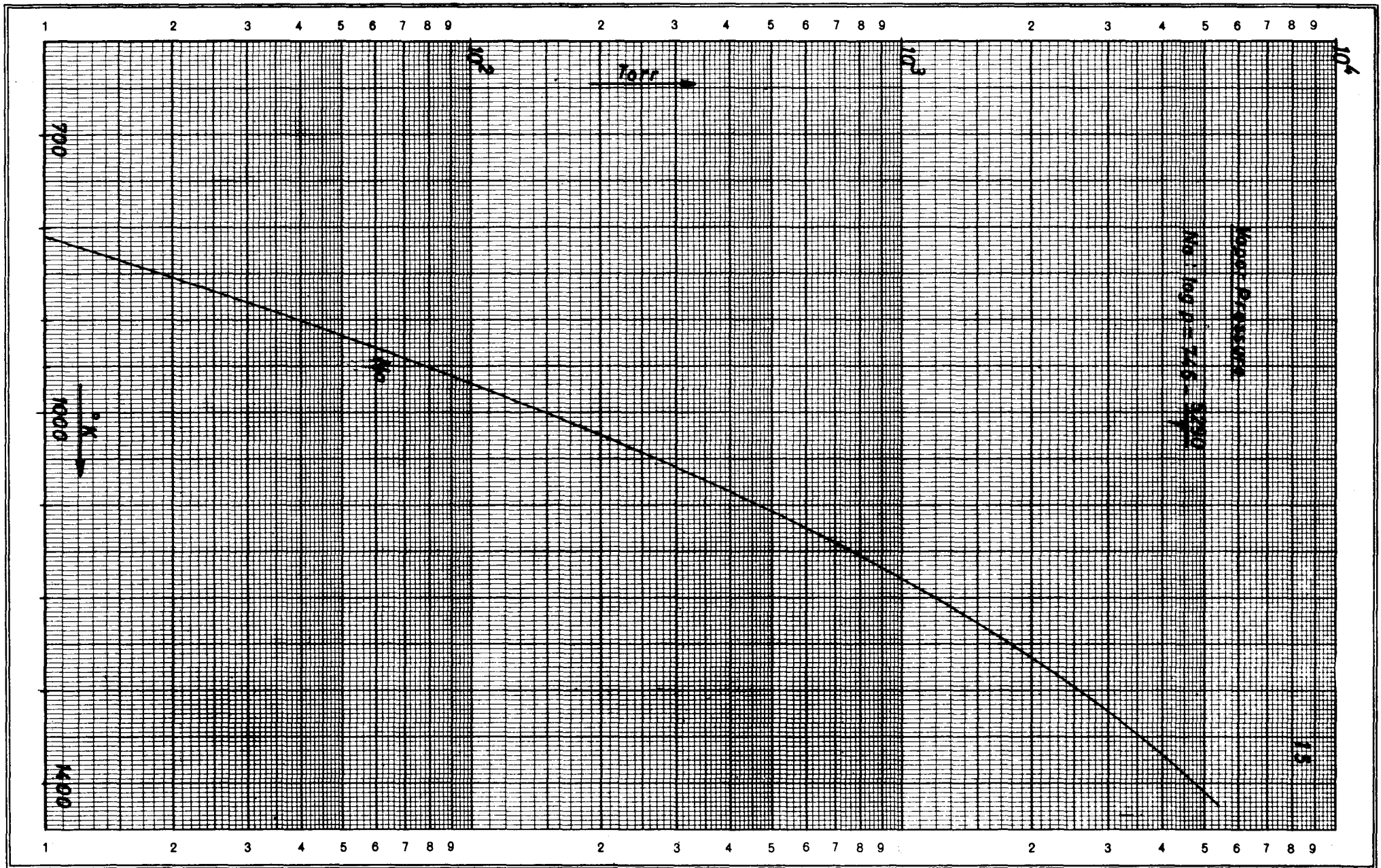
Viscosity
No: $\log \eta = \frac{3309}{T} - 0.0494$



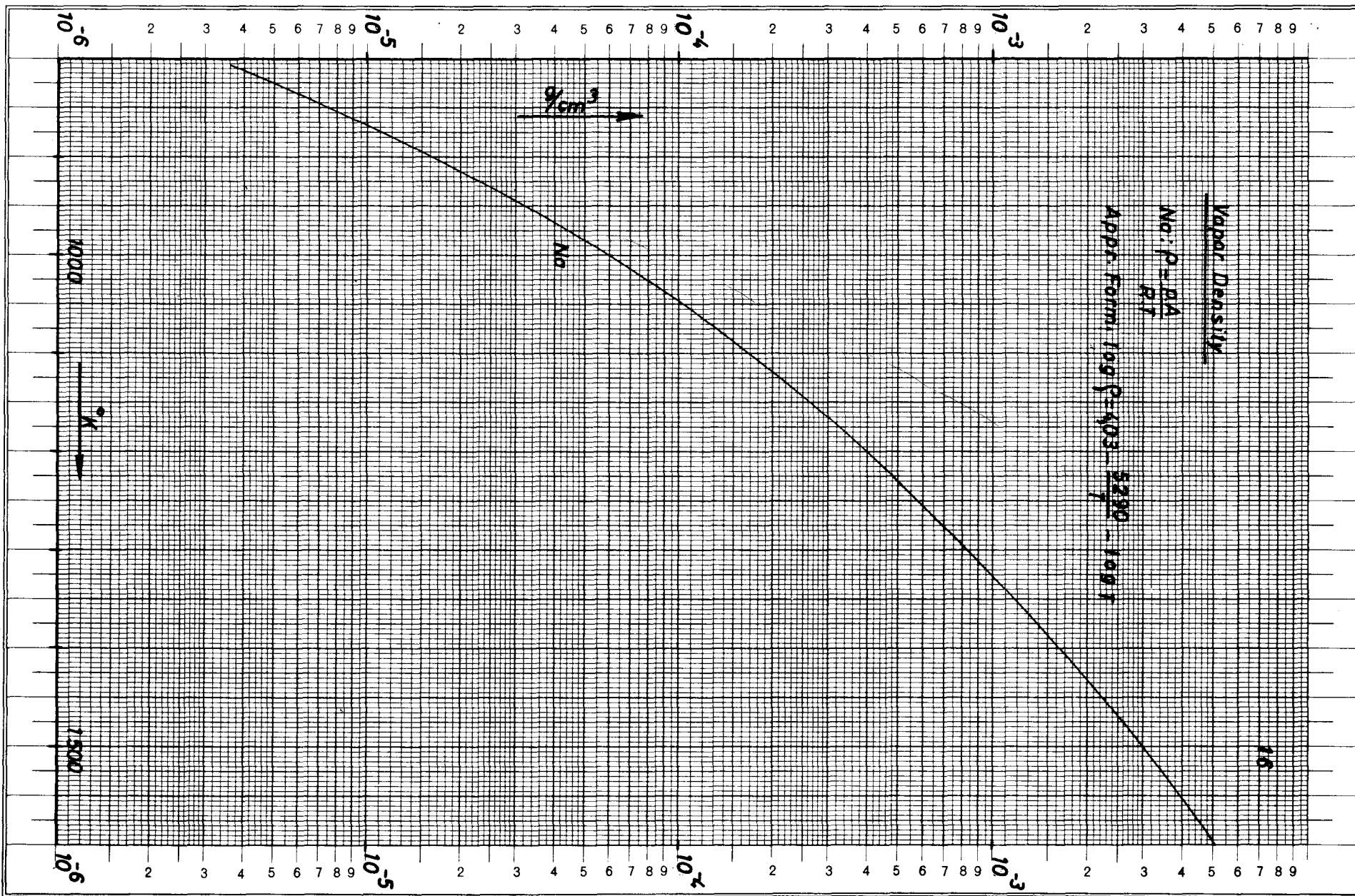
Surface Tension

No. $\sigma = 220 - 0.0917T$



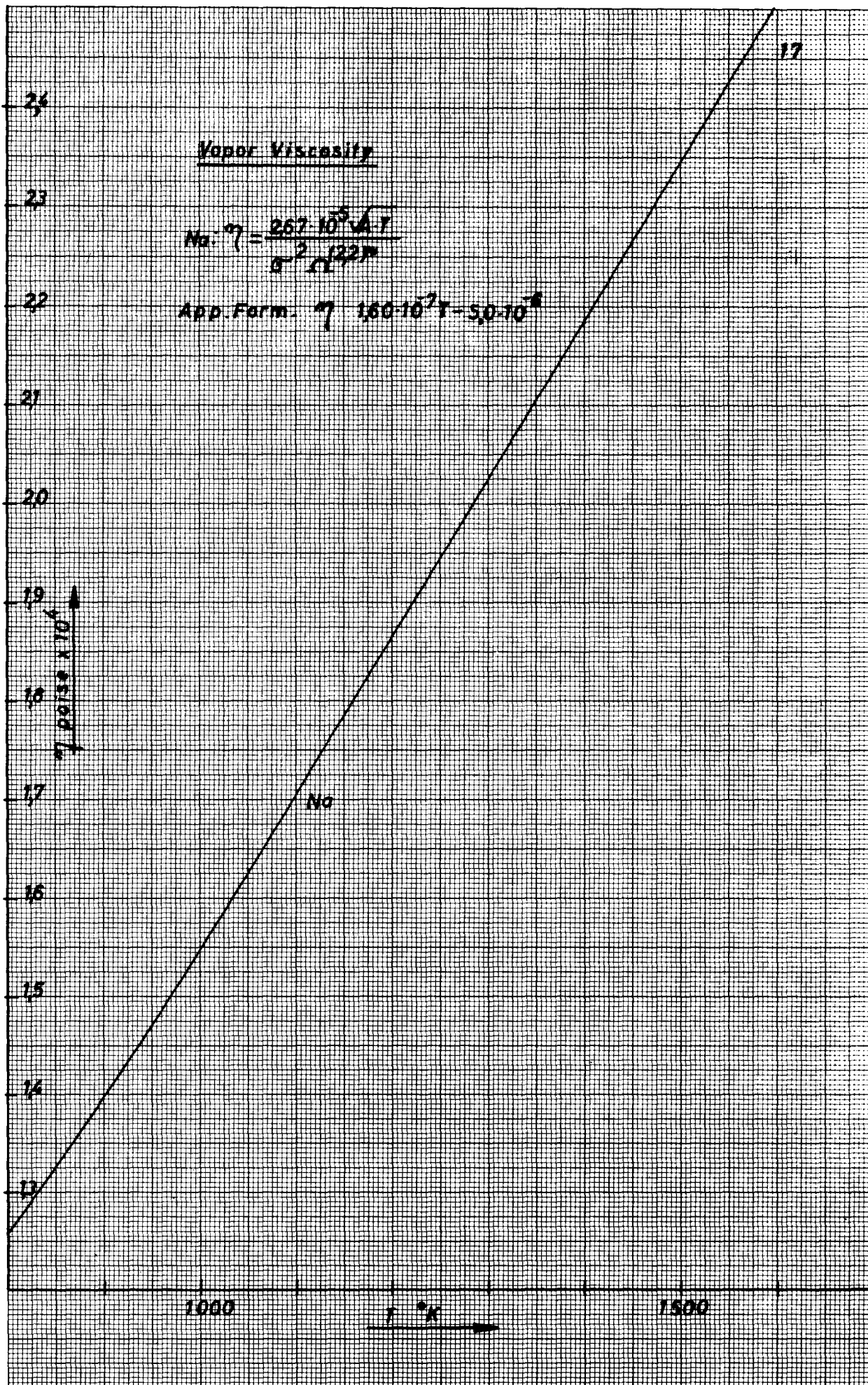


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2.3. <u>Potassium</u>	Value / Formula	References
Atomic number	19	
Atomic weight	39, 1	
Atomic diameter	4, 20 Å	132
Melting point	337°K	17
Boiling point	1049°K	17
Critical temp.	2440°K; 2300°K	9, 157, 135
Heat of vapor.	20, 0 Cal/mol	this work
Density	$0, 909-2, 41 \cdot 10^{-4} \cdot T \text{ g/cm}^3$	54, 57, 93, 94, 101, 103, 116, 134, 136, 138, 139
Viscosity	$1, 001 \exp 1121/RT \text{ mP}$	44, 54, 57, 101, 116, 129, 131, 133, 134, 139
Surface tension	$125-0, 062 T \text{ dyne/cm}$	6, 117, 138
Vapor pressure	$\log P = 7, 22-4440/T \text{ torr}$	5, 131, 158, 159, 160, 161
Vapor density	$6, 27 \cdot 10^{-4} P/T \text{ (P in torr)}$	93, 130.
approx. formula	$4, 02-4440/T - \log T$	
Vapor viscosity	$9, 47 \cdot 10^{-6} \sqrt{T/\Omega}^{(2, 2)*} \text{ poise}$	125, 61
approx. formula	$1, 46 \cdot 10^{-7} \cdot T - 5, 0 \cdot 10^{-6}$	

2.3.1. The viscosity of potassium

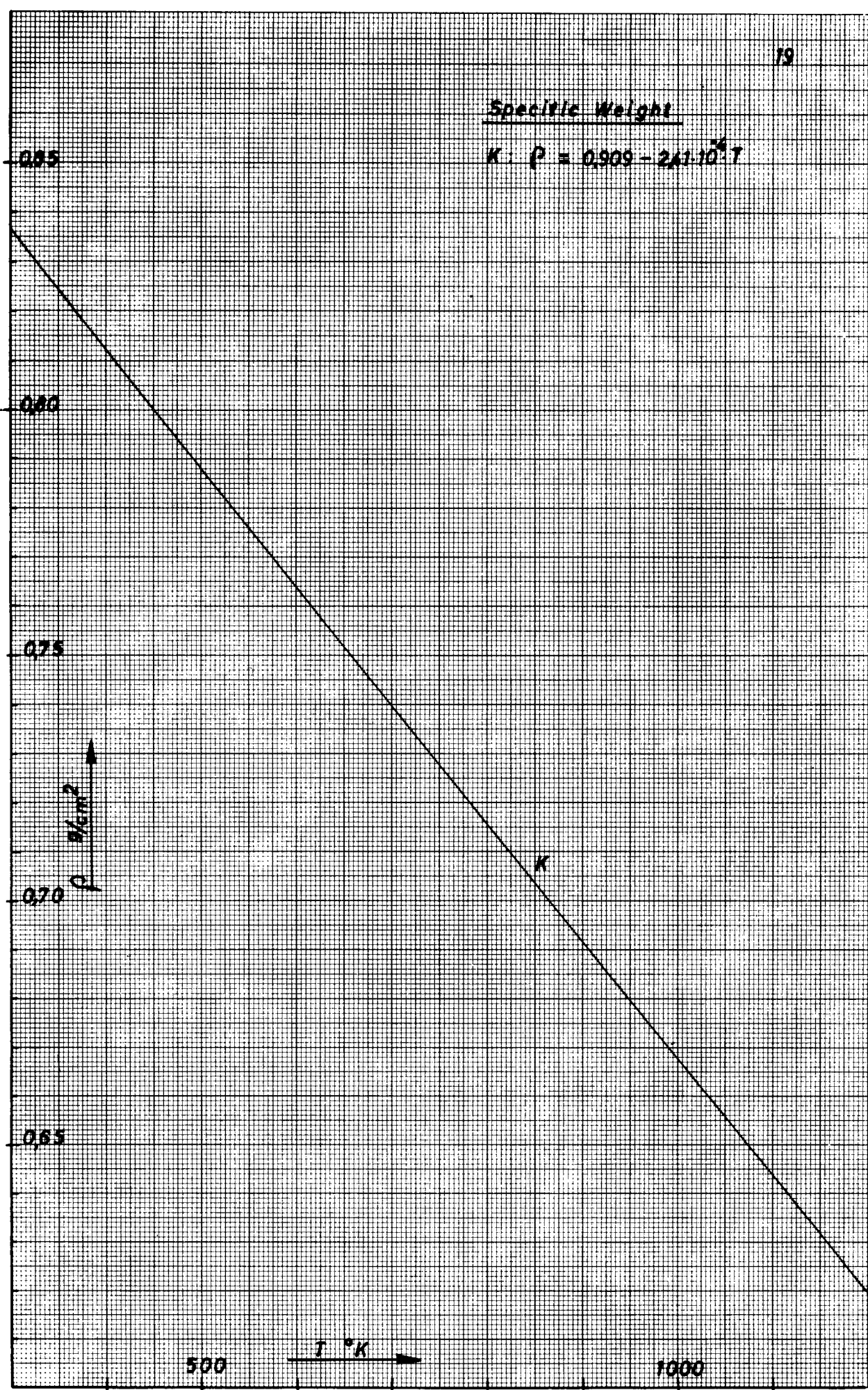
Two regression lines are possible; that of Chiong (57), Gering-Sauerwald (44), Roeder (139) and that of Ewing (54, 94). A complication is that Lemmon (131), Novikov (101) and Nikolskiy (134) cross over: at low temperature they confirm Chiong at very high Ewing. We give the Chiong values.

2.3.2. The density of potassium

Author	Year	Method	Max. Temp.	Formula
54, 94, 116	1951/54/66	Pycnometer	1250°C	$0, 9105-2, 44 \cdot 10^{-4} T$
139	1956	Archimedean	-	$0, 909-2, 35 \cdot 10^{-4} T$
101	1956	Archimedean	750°C	$0, 907-2, 36 \cdot 10^{-4} T$
134	1959	-	700°C	$0, 903-2, 28 \cdot 10^{-4} T$
138	1964	Max. Bubble P.	750°C	$0, 907-2, 375 \cdot 10^{-4} T$
93	1965	γ-radiation	1250°C	$0, 888-2, 24 \cdot 10^{-4} T$

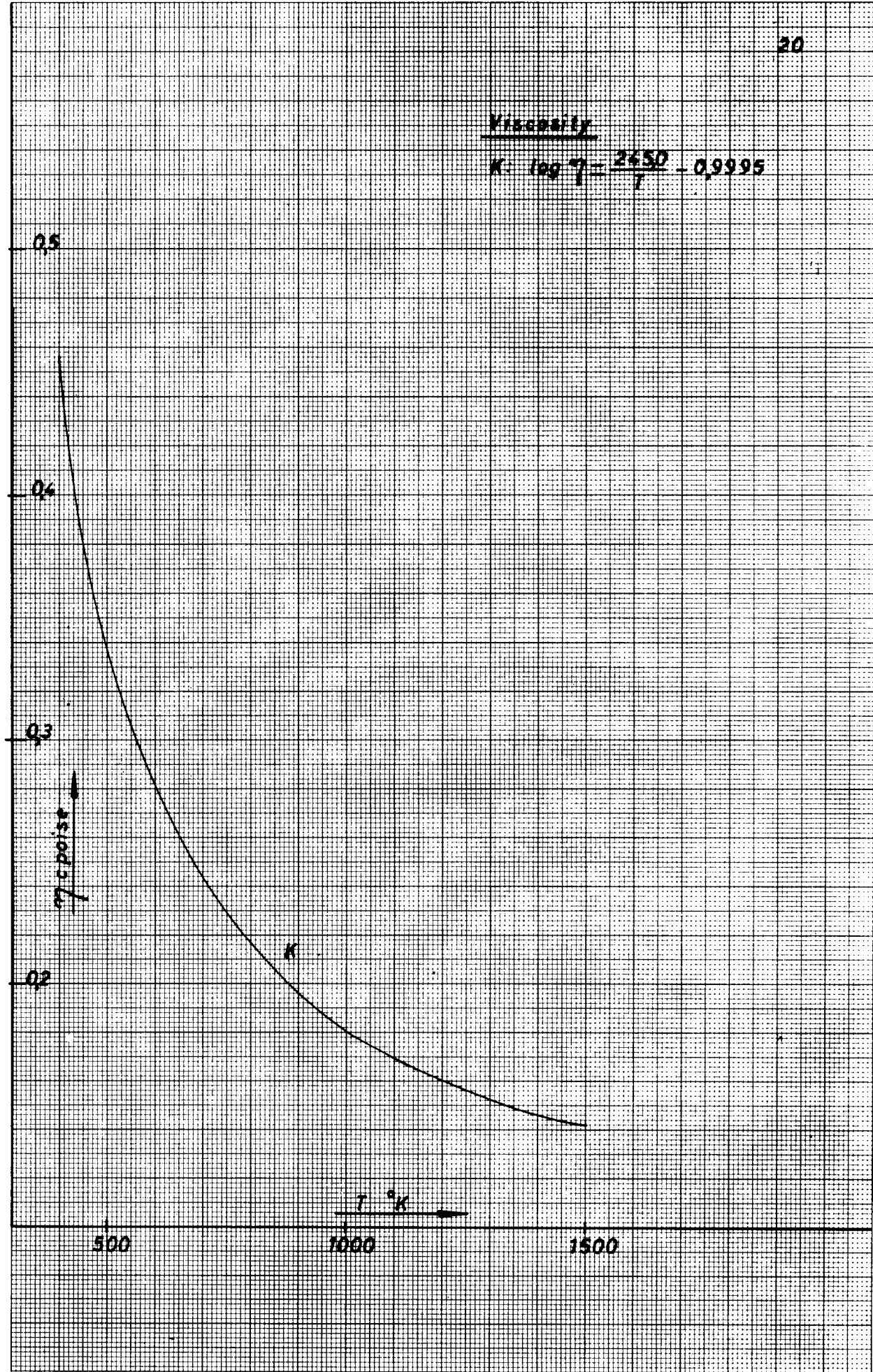
Specific Weight

$$K: \rho = 0.909 - 2.41 \cdot 10^{-4} T$$



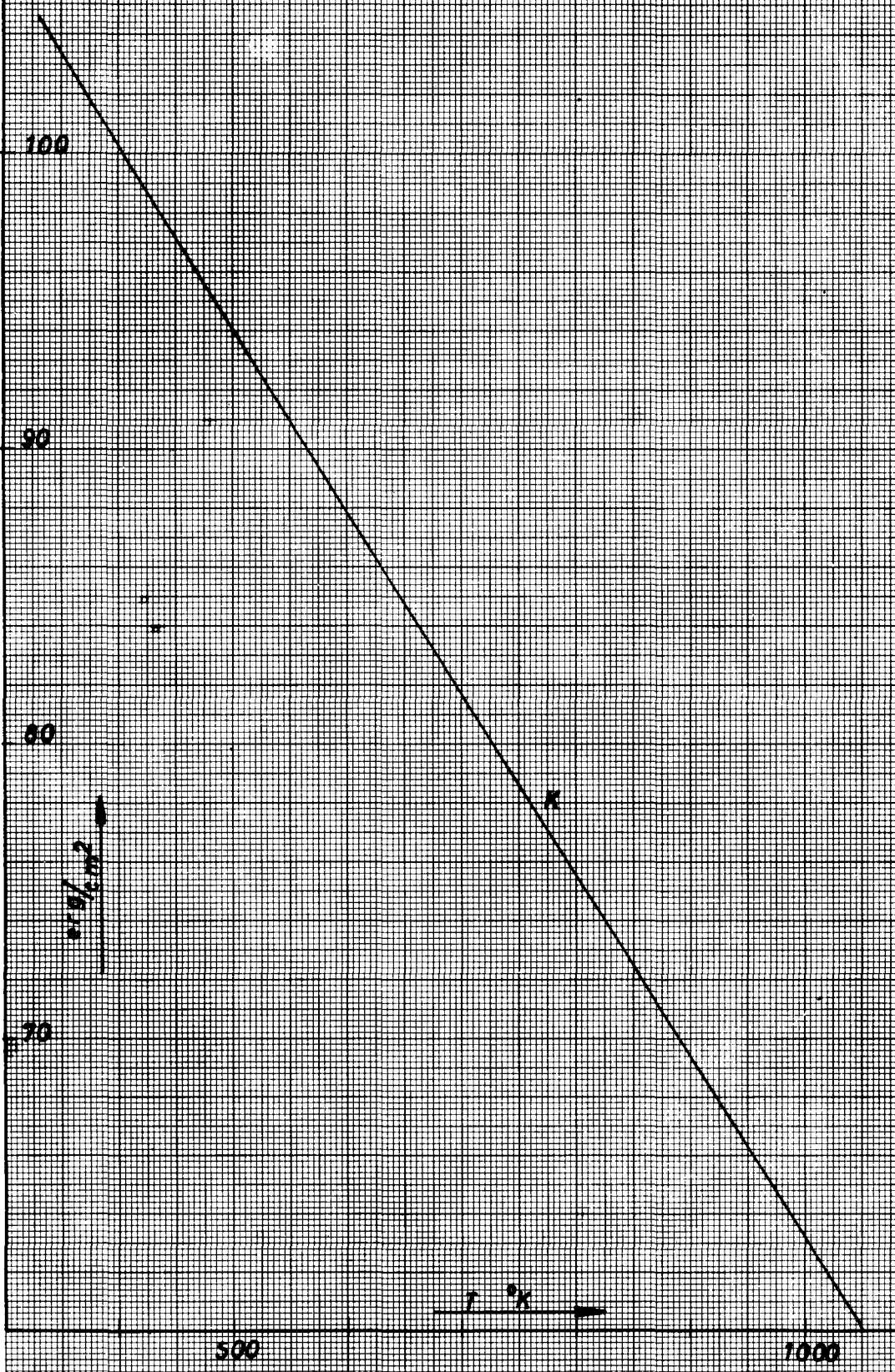
Viscosity

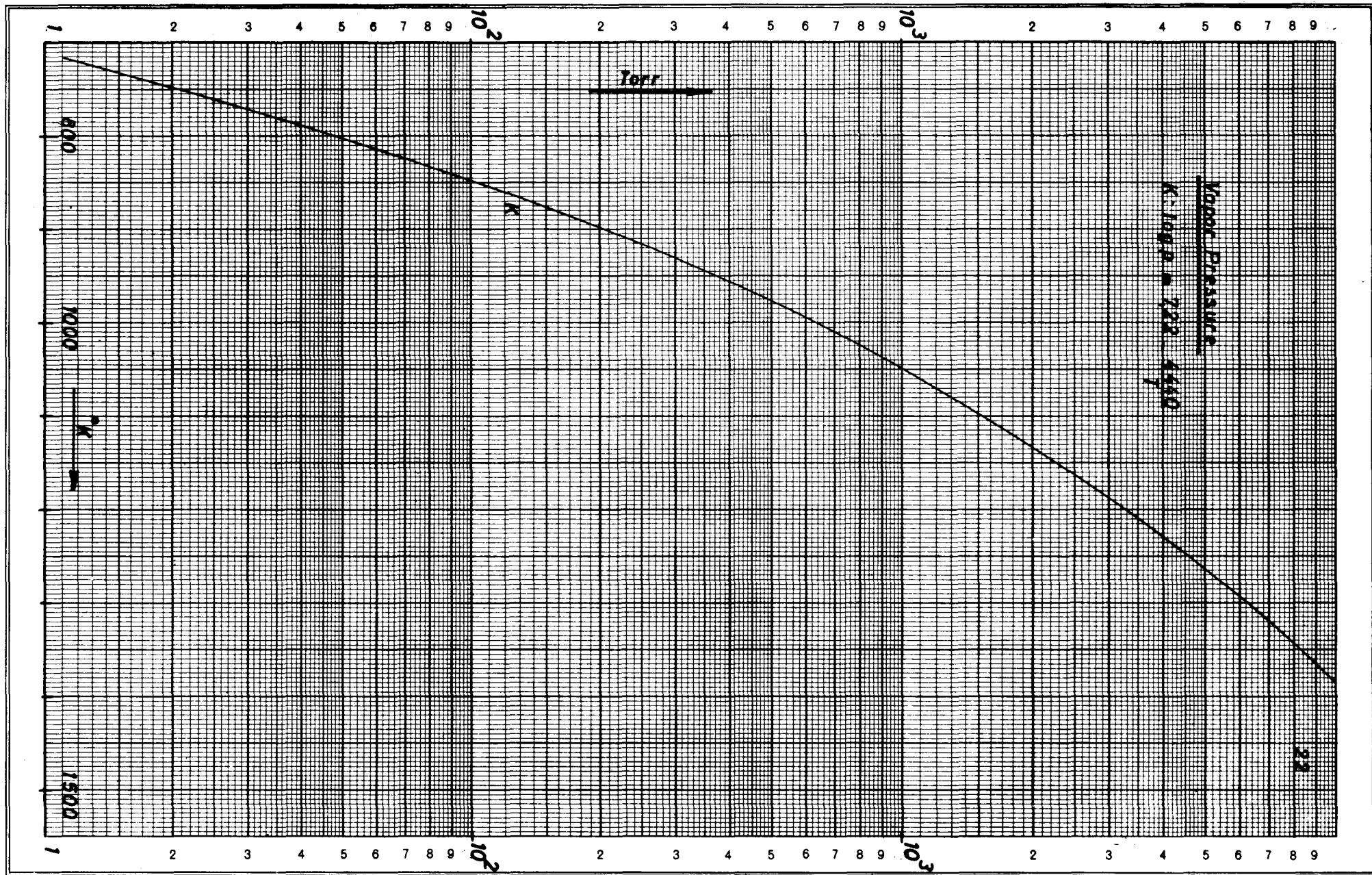
$$K: \log \eta = \frac{2450}{T} - 0.9995$$



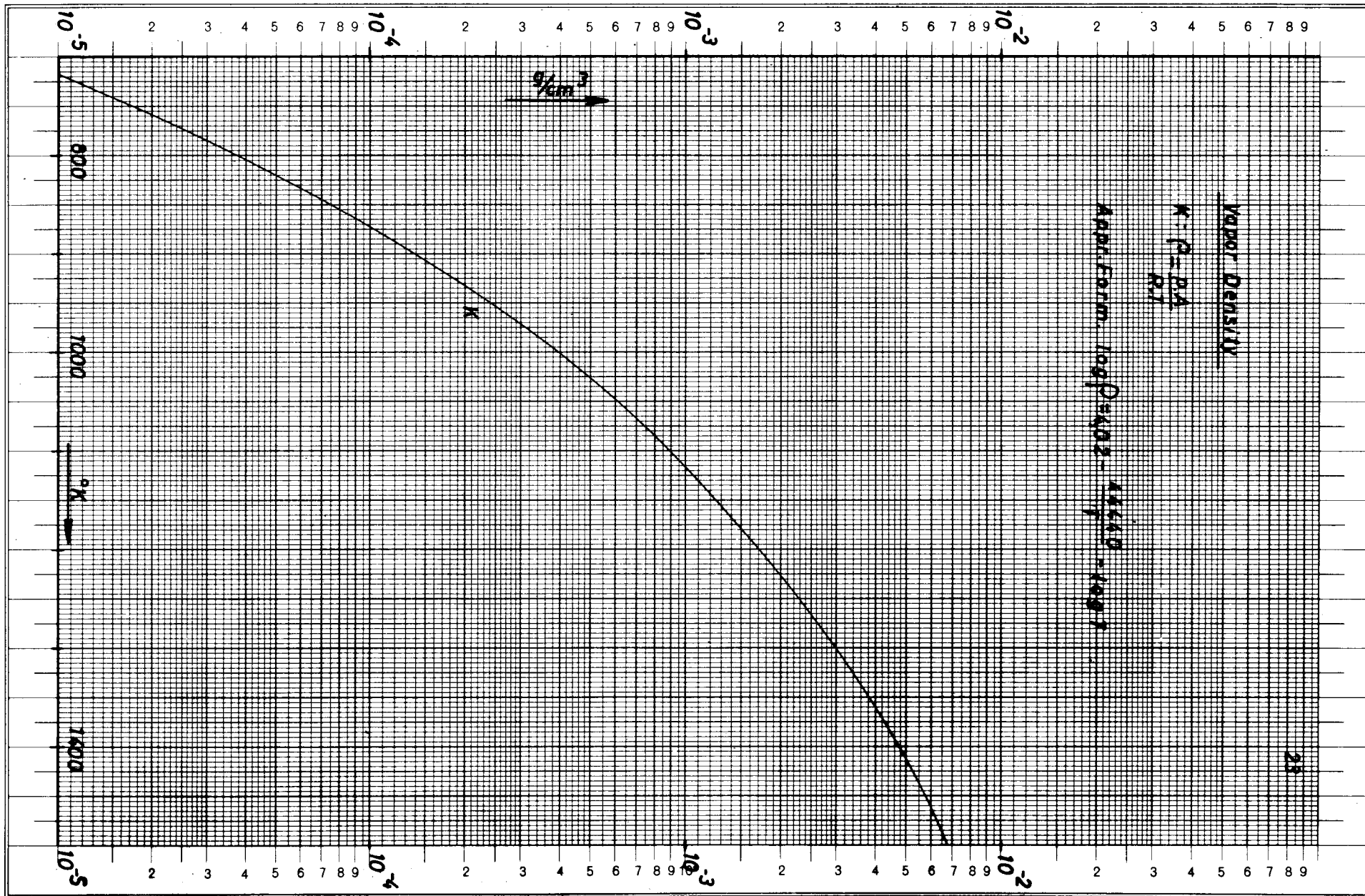
Surface Tension

$\sigma = 125 - 0.002T$

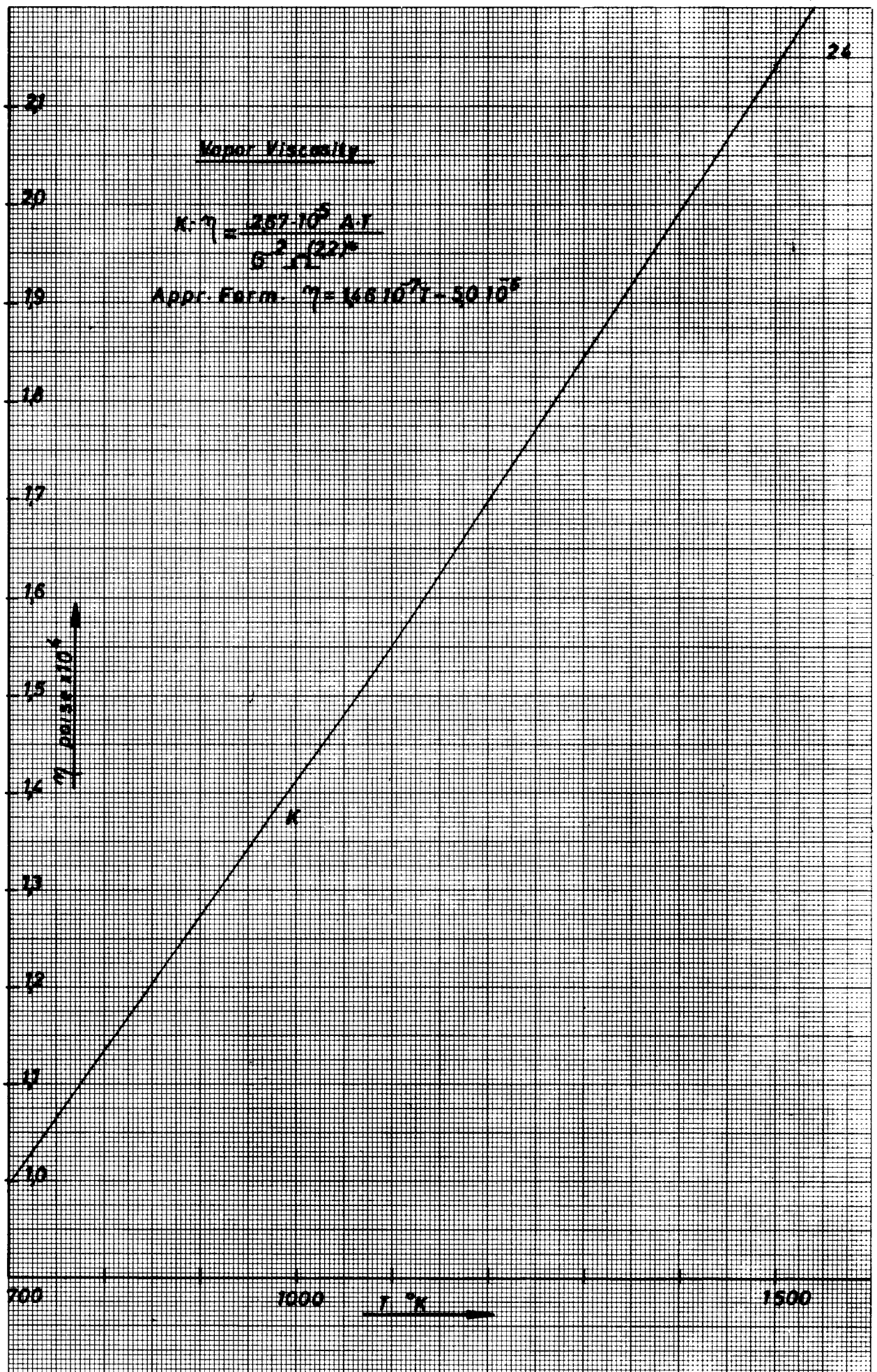




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2.4.

<u>Rubidium</u>	Value / Formula	References
Atomic number	37	
Atomic weight	85,48	
Atomic diameter	4,50 Å	132
Melting point	312°K	17
Boiling point	962°K	5
Critical temp.	2110°K	77
Heat of vapor.	18,3 Cal/mol	5
Density	$1,614-4,32 \cdot 10^{-4} T \text{ g/cm}^3$	33, 34, 36, 93, 103, 140, 141
Viscosity	$0,93 \exp 1213/RT \text{ mP}$	36, 142
Surface tension	$95 - 0,052 T \text{ dyne/cm}$	6
Vapor pressure	$\log P = 7,05-4010/T \text{ torr}$	5, 33, 34, 141, 137
Vapor density	$1,37 \cdot 10^{-3} P/T (P \text{ in torr})$	79, 93, 130
approx. formula	$4,19-4010/T - \log T$	
Vapor viscosity	$1,183 \cdot 10^{-5} \sqrt{T/\Omega}^{(2,2)*} \text{ poise}$	130, 125, 61
approx. formula	$1,88 \cdot 10^{-7} \cdot T - 2,0 \cdot 10^{-6}$	

2.4.1. The viscosity of rubidium

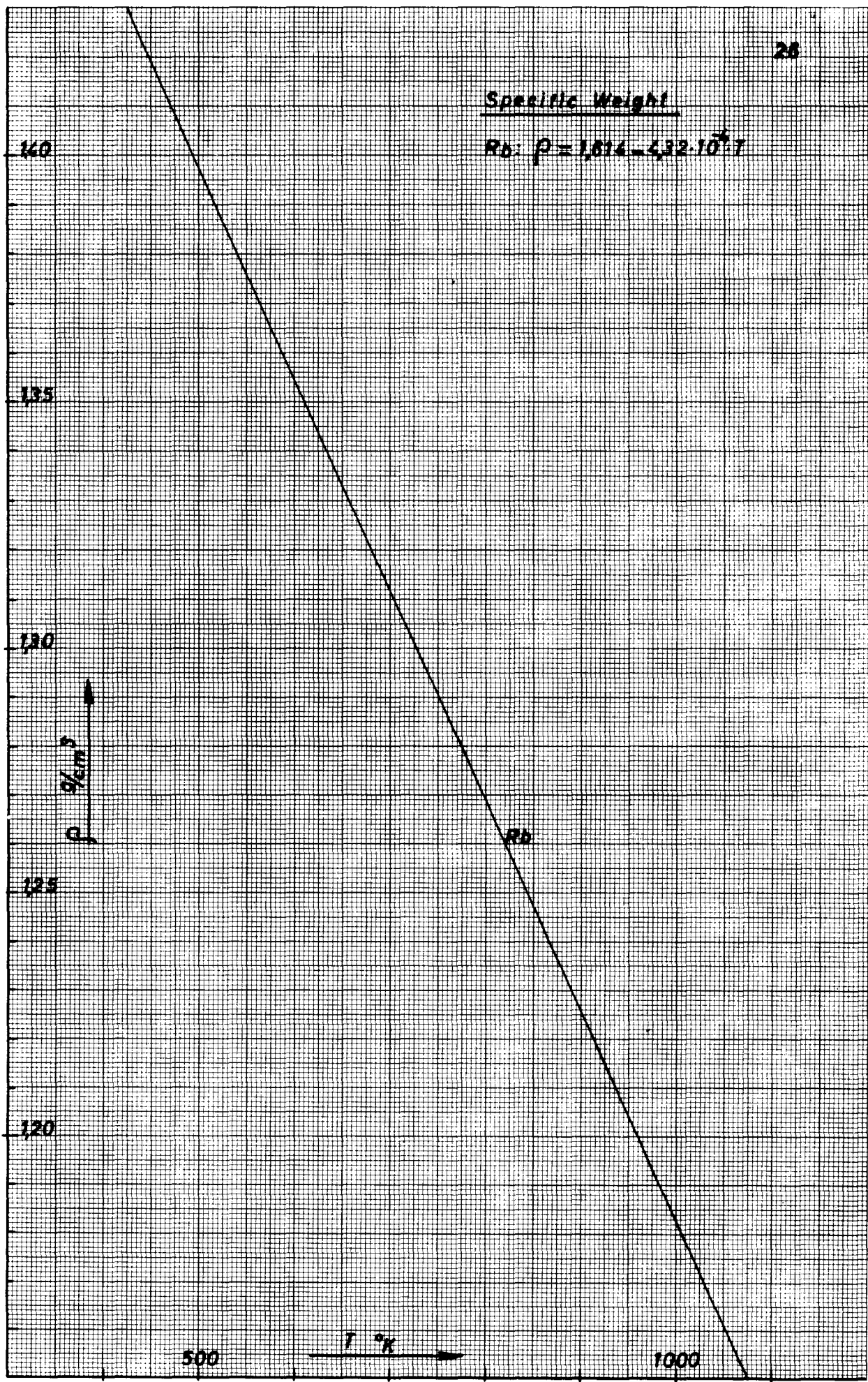
The data of Weatherford (142) confirm excellently the original values of Andrade (36).

2.4.2. The density of rubidium

Author	Year	Method	Max. Temp.	Formula
36	1952	Pycnometer	220°C	$1,587-4,30 \cdot 10^{-4} T$
140	1957	Archimedean	500°C	$1,689-5,40 \cdot 10^{-4} T$
141	1962	Dilatometer	750°C	$1,600-4,04 \cdot 10^{-4} T$
33	1964	-	1100°C	$1,652-4,55 \cdot 10^{-4} T$
34	1964	-	950°C	$1,602-4,10 \cdot 10^{-4} T$
93	1965	γ-radiation	1720°C	$1,689-4,77 \cdot 10^{-4} T$

Specific Weight

Rb: $\rho = 1,014 - 4,32 \cdot 10^{-4} T$



140

135

130

125

120

ρ g/cm³

Rb

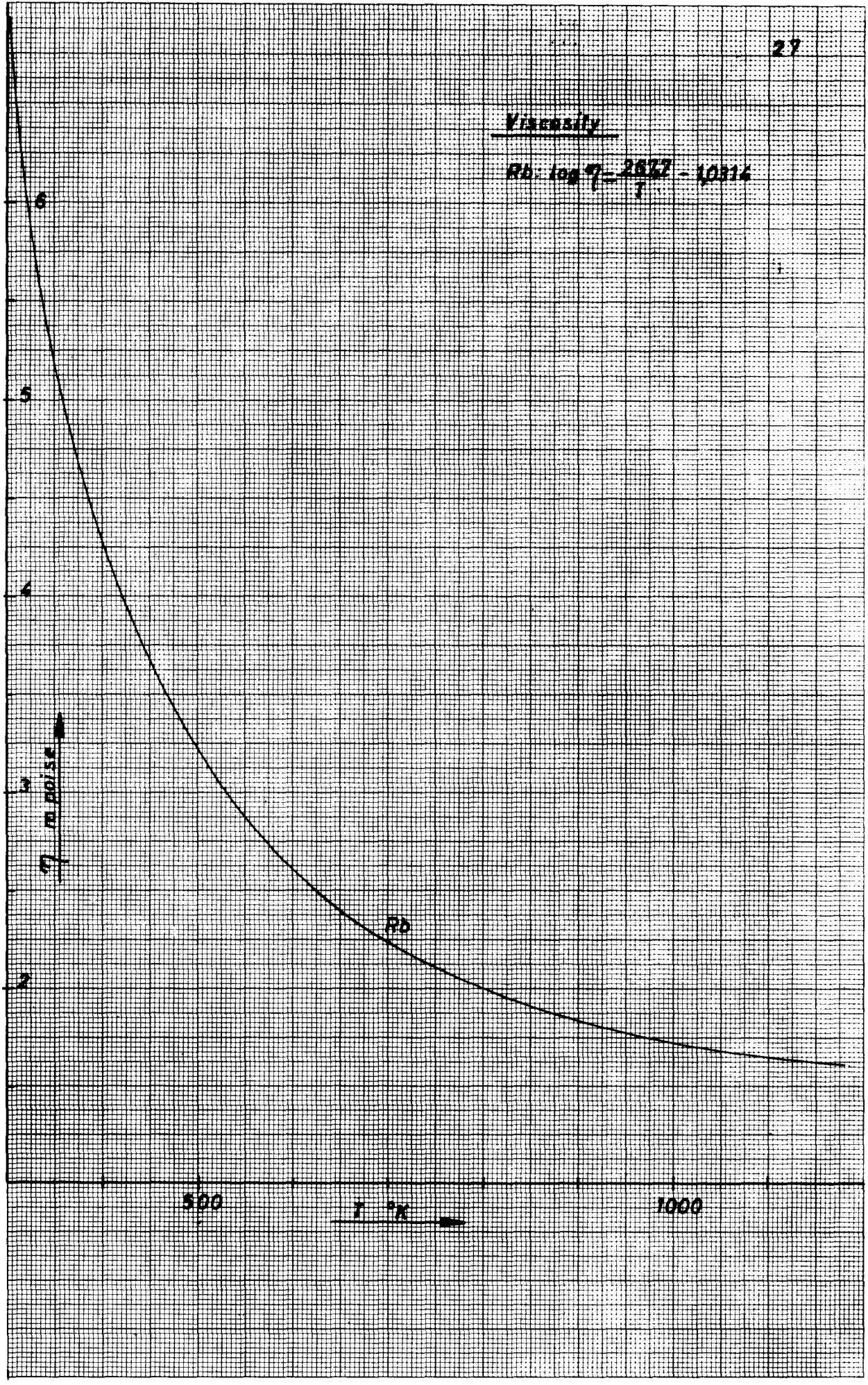
500

T °K

1000

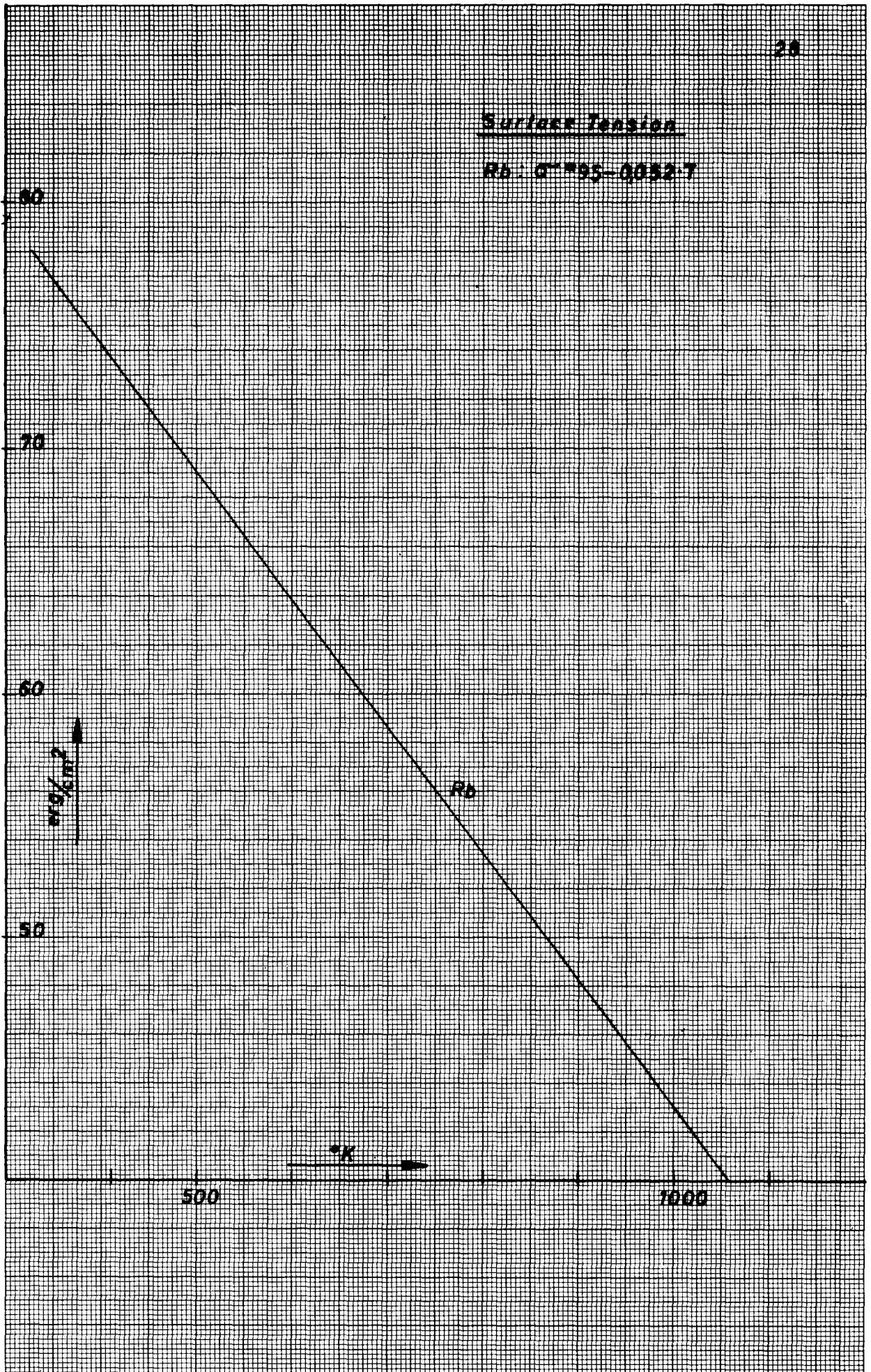
Viscosity

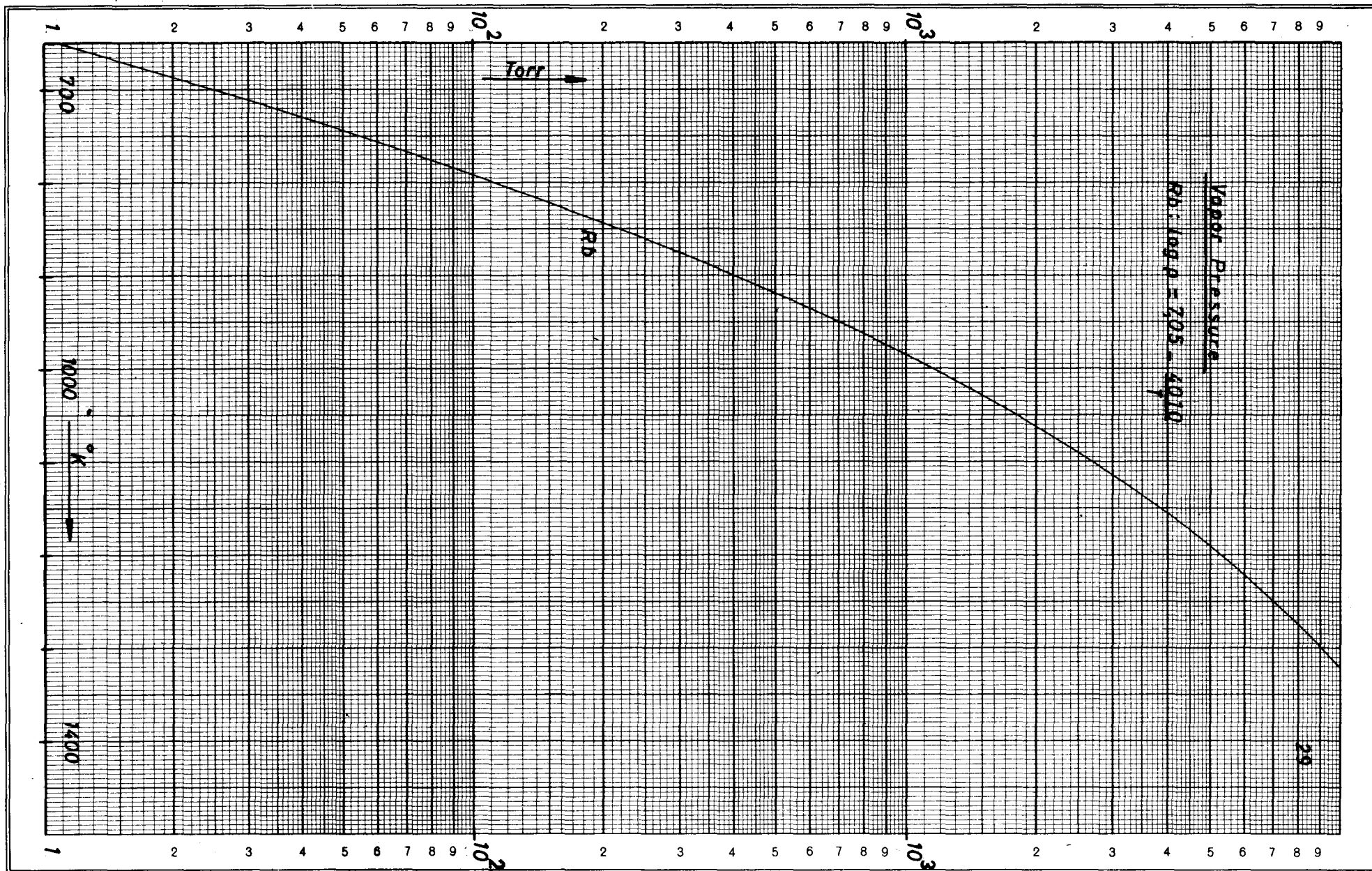
$RD: \log \eta = \frac{2837}{T} - 10.314$



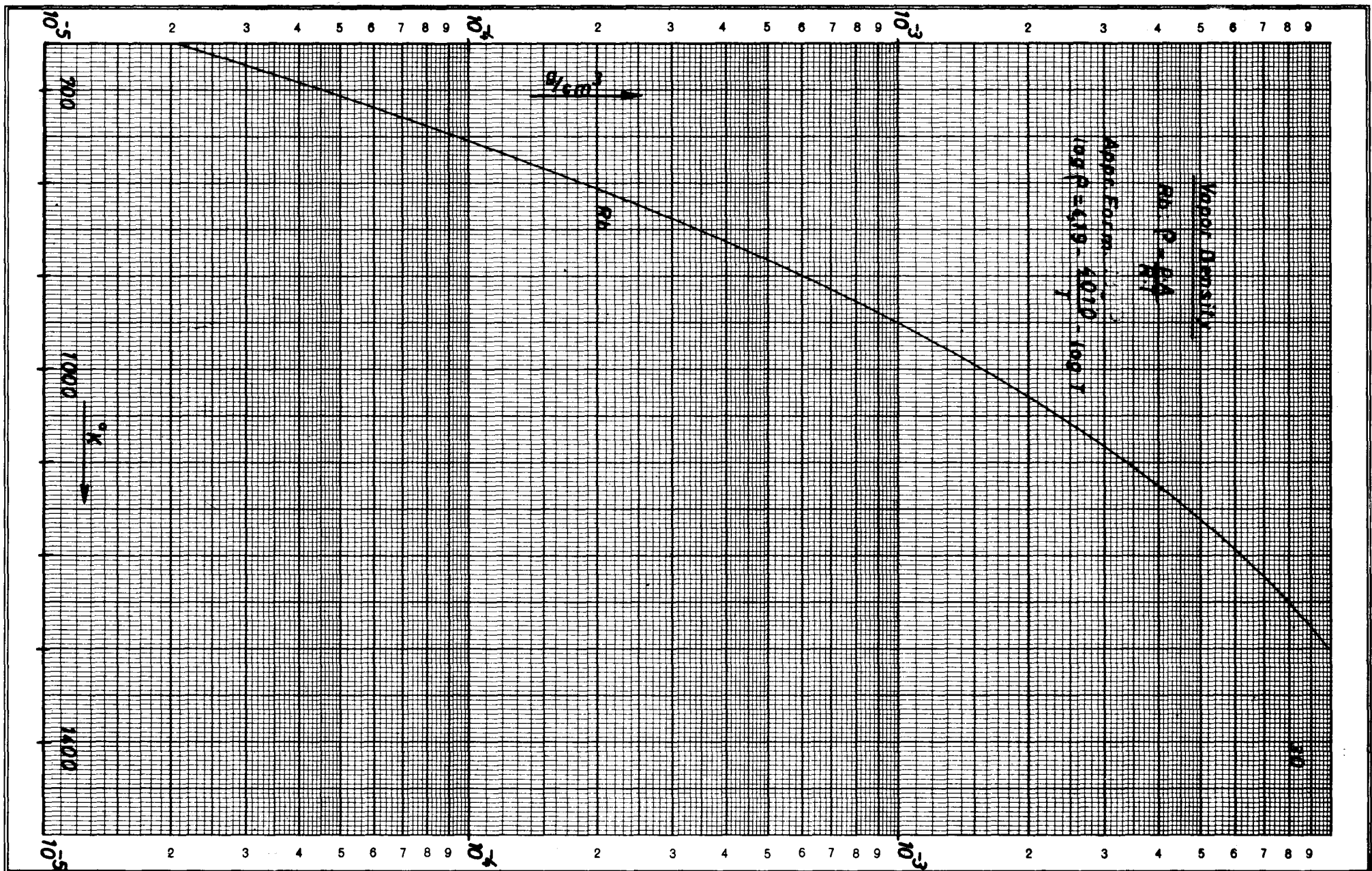
Surface Tension

Rb: C-95-0052-7

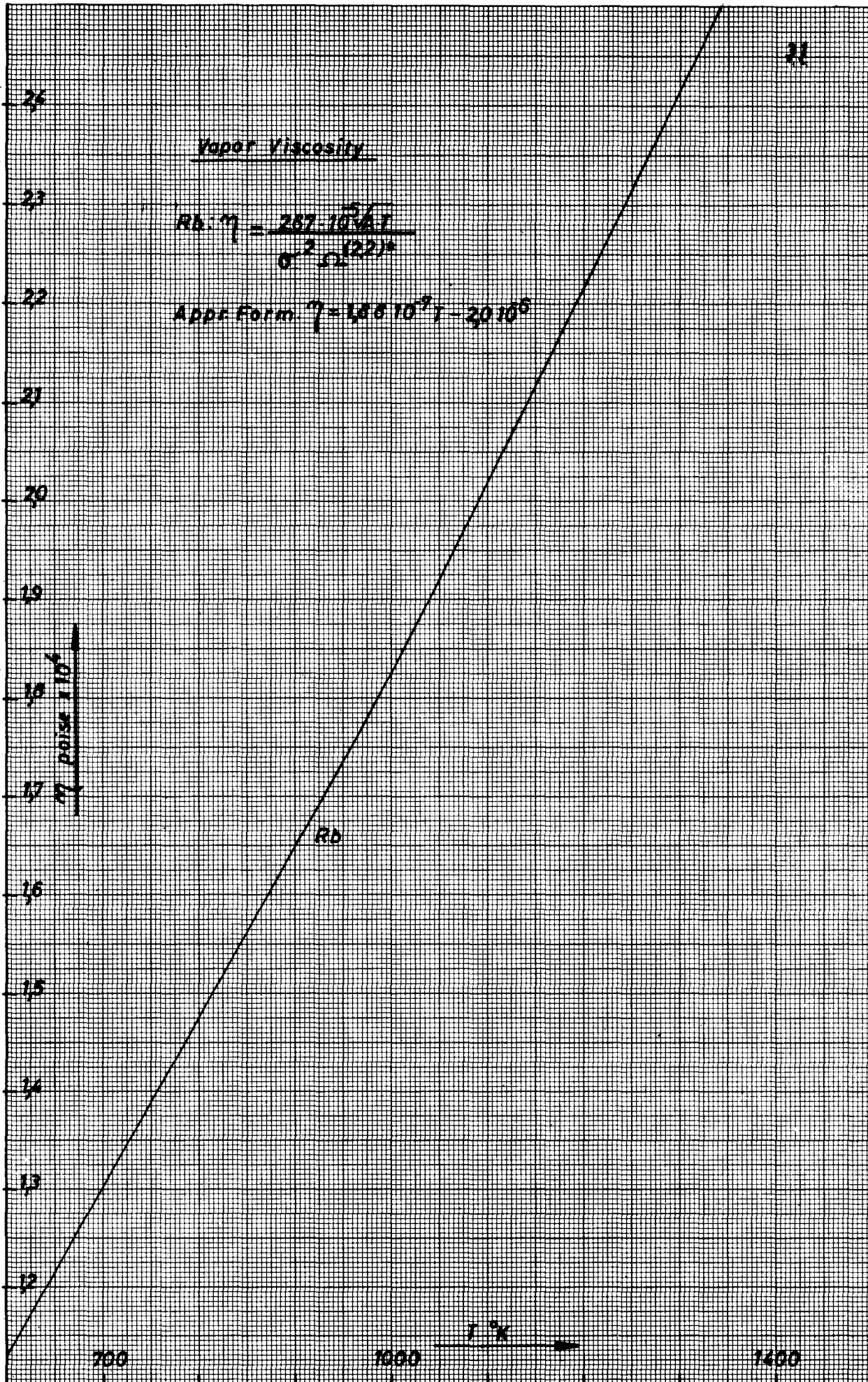




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2.5. Caesium	Value / Formula	References
Atomic number	55	
Atomic weight	132,91	
Atomic diameter	4,90 Å	132
Melting point	302°K	17
Boiling point	939°K	5
Critical temp.	2050°K	77
Heat of vapor.	17,2 Cal/mol	5
Density	$2,028-5,81 \cdot 10^{-4} \cdot T \text{ g/cm}^3$	33,34,36,77,78, 79,94,141
Viscosity	$1,135 \exp 1060/RT \text{ mP}$	36
Surface tension	$80-0,05 T \text{ dyne/cm}$	6
Vapor pressure	$\log P = 6,88-3750/T \text{ torr}$	5,159,33,34,162, 141,136
Vapor density	$2,127 \cdot 10^{-3} P/T \text{ (P in torr)}$	78,93,130
approx. formula	$4,21-3750/T - \log T$	
Vapor viscosity	$1,28 \cdot 10^{-5} \sqrt{T/\Omega}^{(2,2)*} \text{ poise}$	128,130,125,61
approx. formula	$2,10 \cdot 10^{-7} \cdot T - 6,0 \cdot 10^{-6}$	

2.5.1. The viscosity of caesium

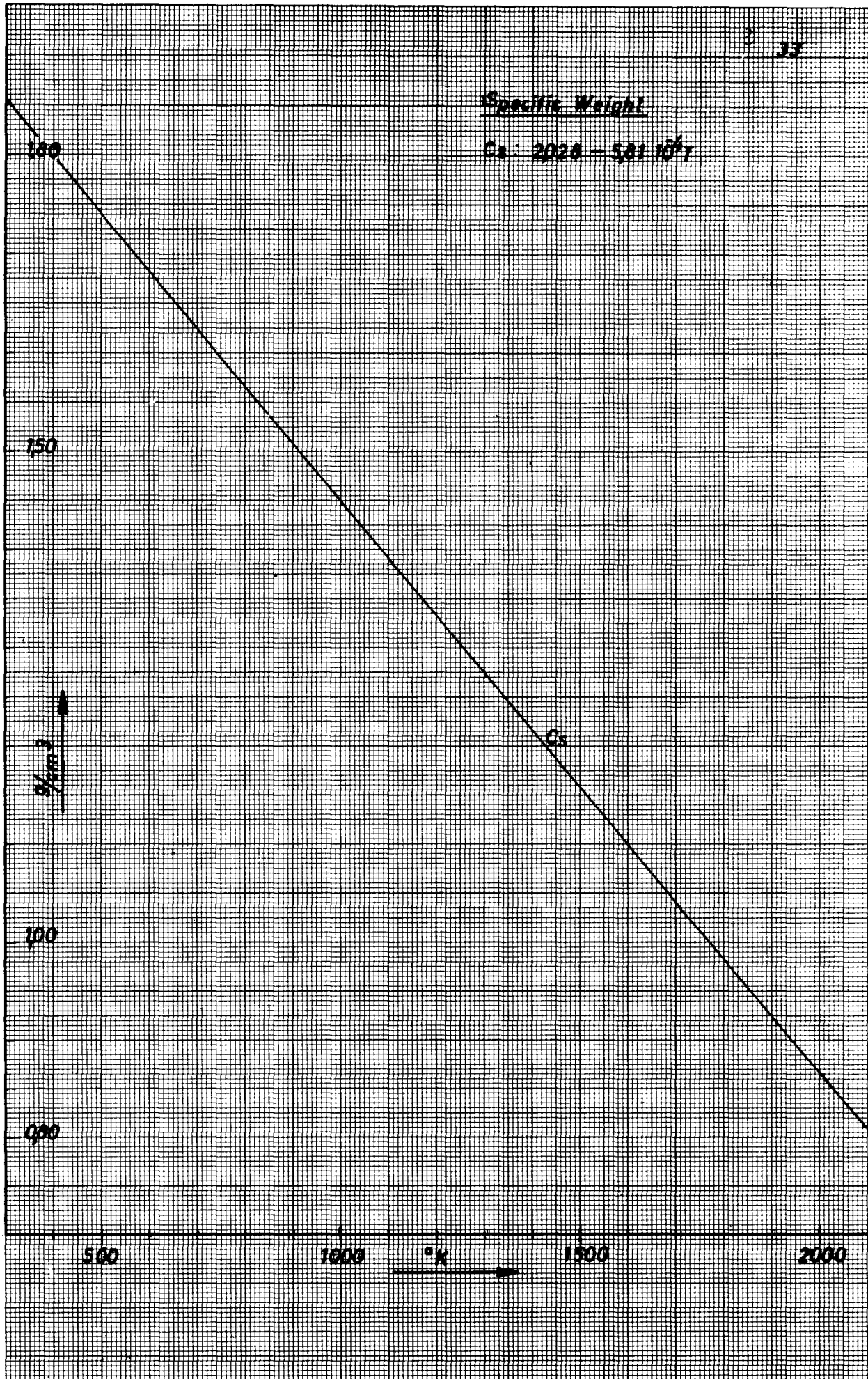
Only one measurement has been made, i. e. by Andrade and Dobbs (36). It appears nevertheless very reliable, because of the exactness of all their alkali measurements.

2.5.2. The density of caesium

Author	Year	Method	Max. Temp.	Formula
(36)	1952	volumetric	210°C	$2,020-6,0 \cdot 10^{-4} T$
(141)	1962	pycnometer	750°C	$2,001-5,198 \cdot 10^{-4} T$
(33)	1963	dilatometric	-	$2,0372-5,964 \cdot 10^{-4} T$
(34)	1964	pycnometer	910°C	$2,055-5,97 \cdot 10^{-4} T$
(79)	1965	radiation	1750°C	$2,024-5,6676 \cdot 10^{-4} T$
(78)	1965	dilatometric	1650°C	$2,0104-5,4639 \cdot 10^{-4} T$
(94)	1966	pycnometer	1400°C	$1,9950-5,0527 \cdot 10^{-4} T$
(77)	1966	survey		$2,006-5,46 \cdot 10^{-4} T$

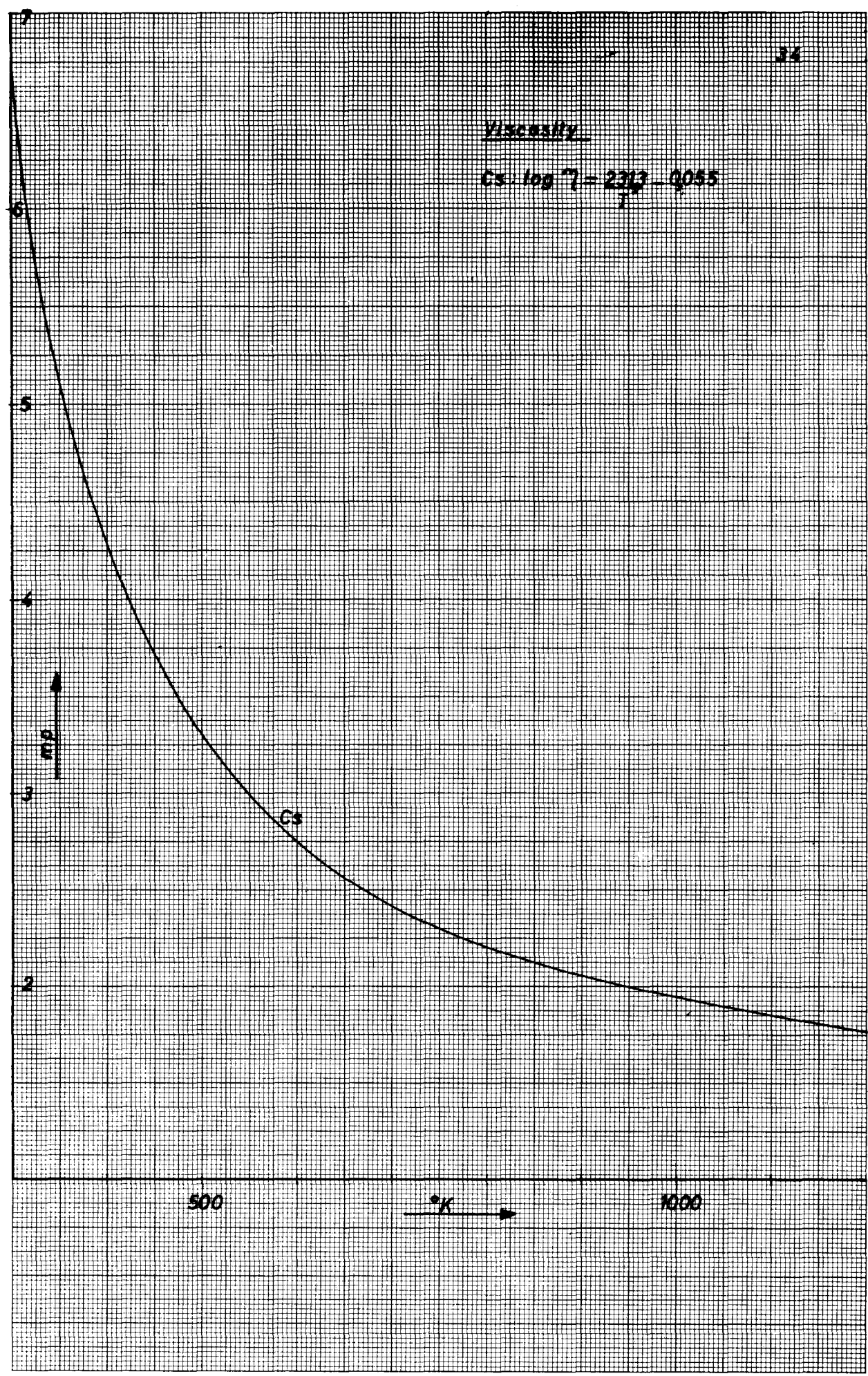
Specific Weight

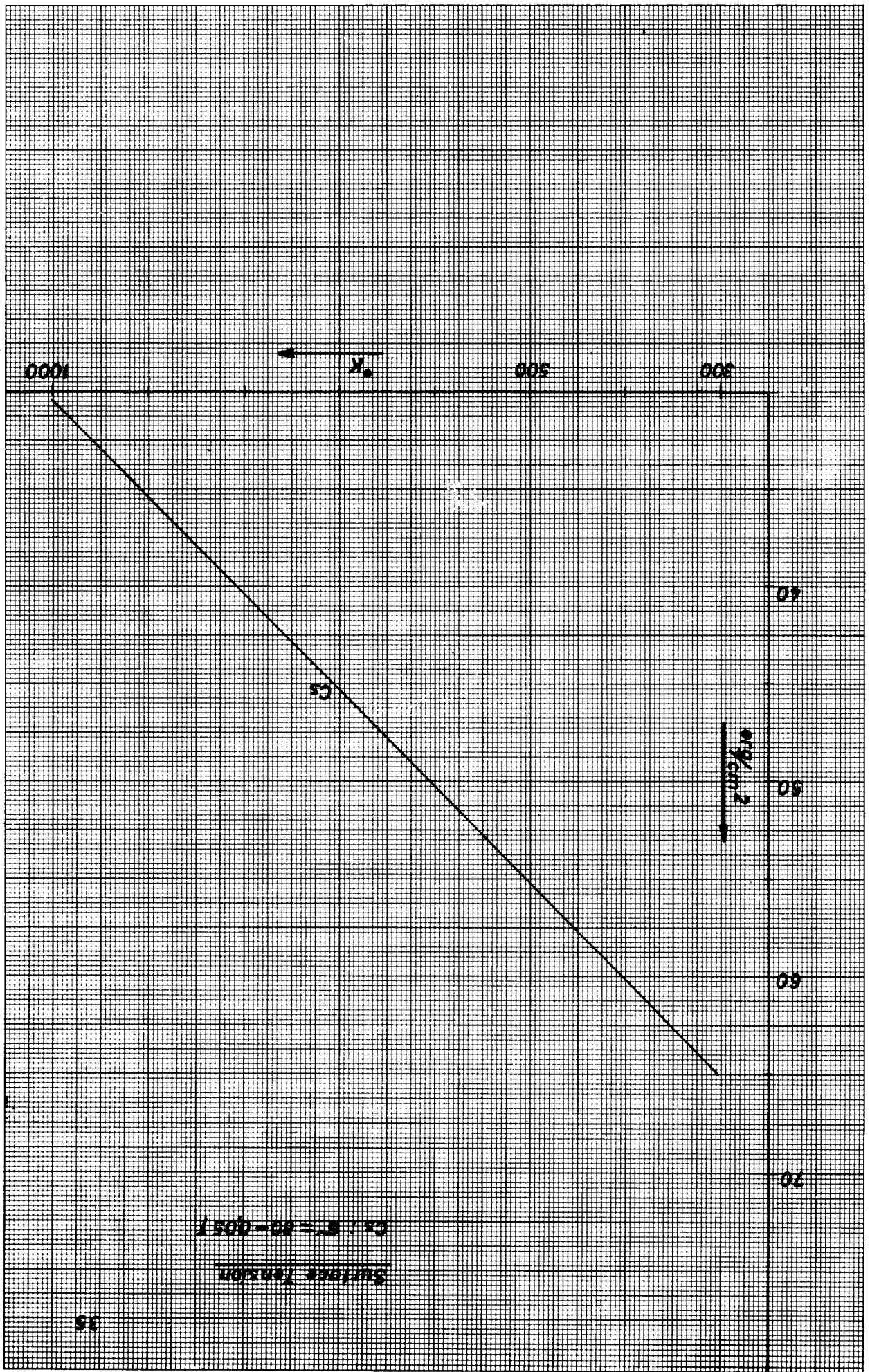
Ca: 2028 - 581 10⁶ T



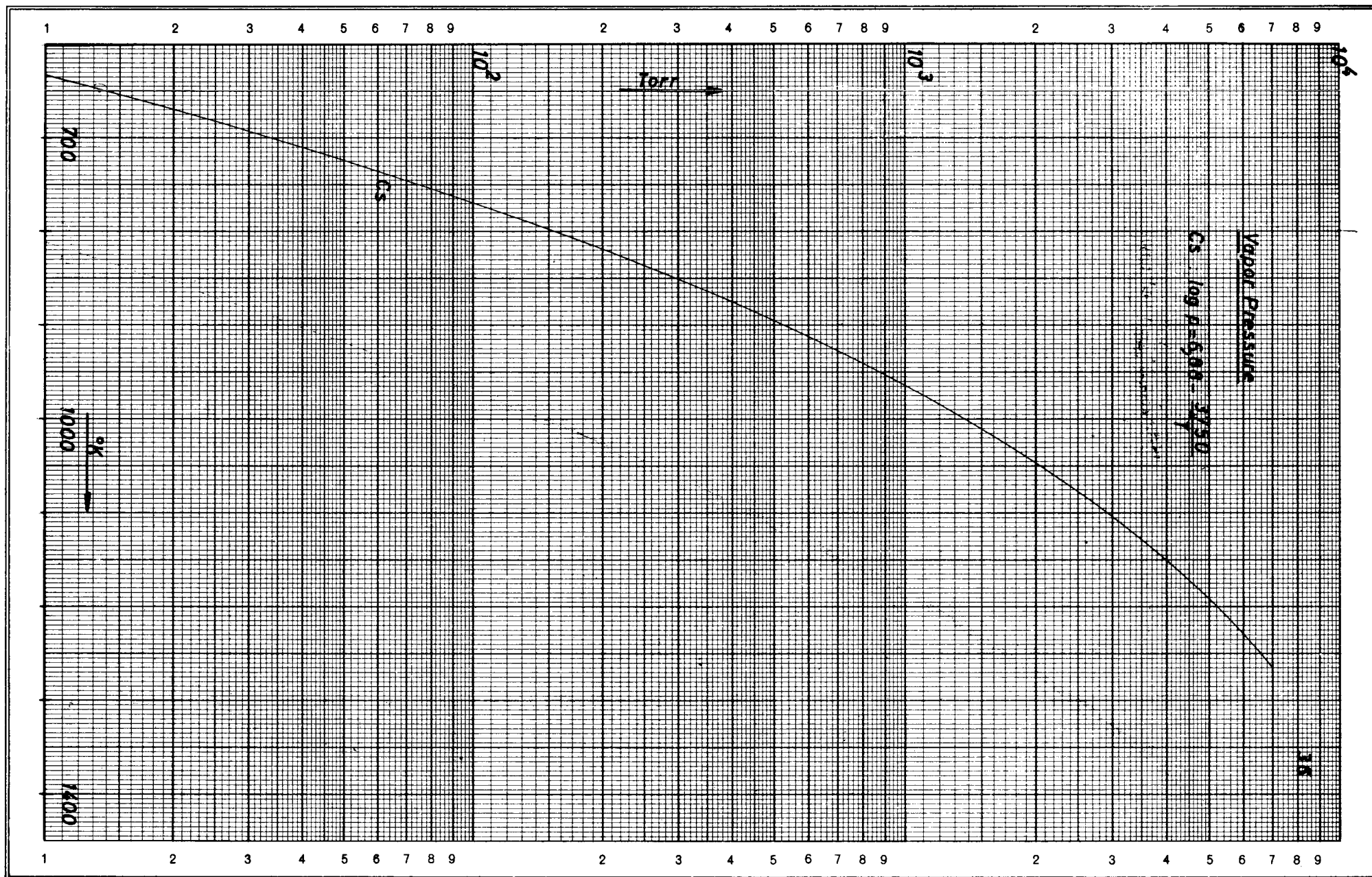
VISCOSITY

$Cs \cdot \log \eta = 2313 - 0.055 T$

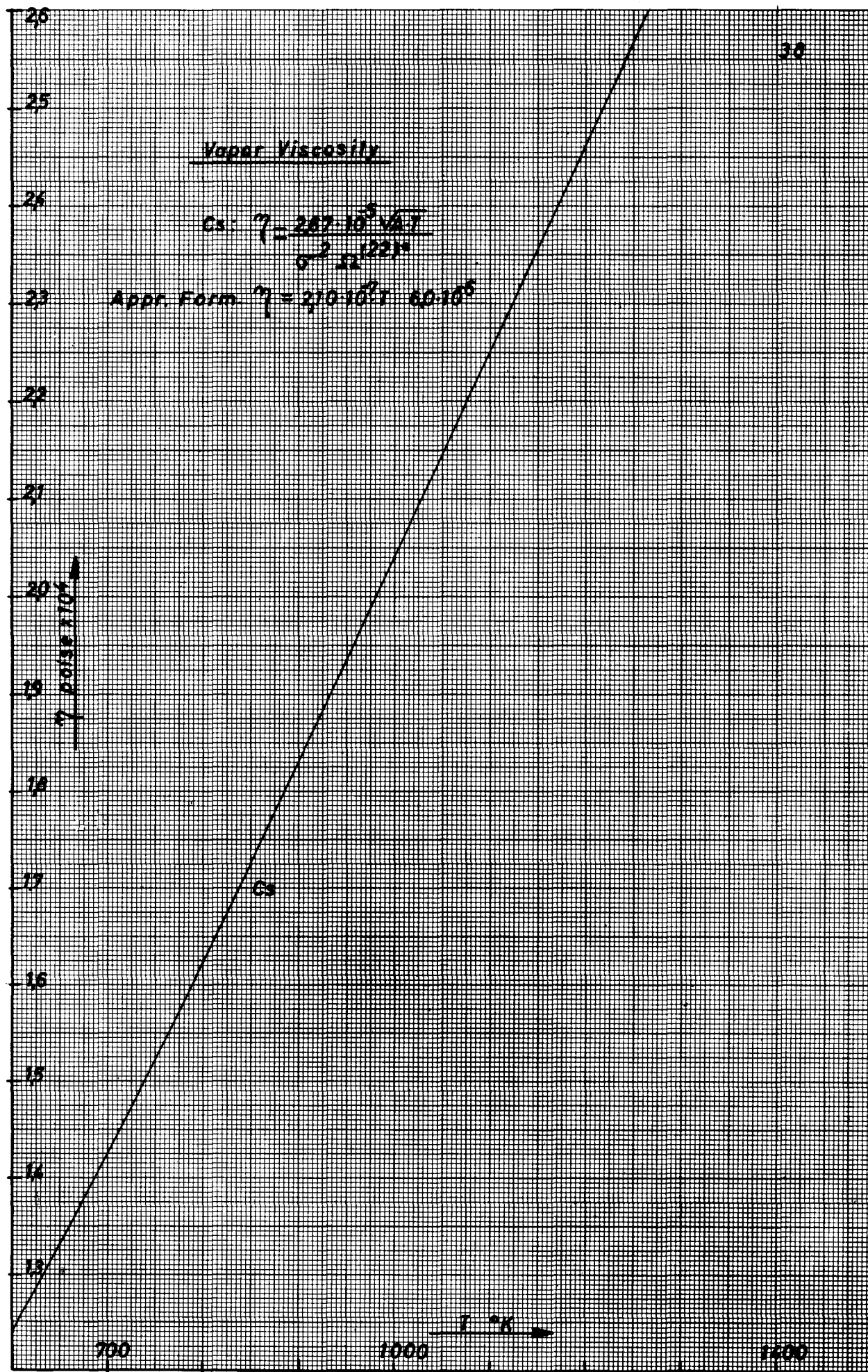




Surface Tension
 C x 10³ (g/100 ml)



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2. 6.	Silver	Value/Formula	References
	Atomic number	47	
	Atomic weight	107, 88	
	Atomic diameter	2, 58 Å	132
	Melting point	1234°K	17
	Boiling point	2450°K	5
	Critical temp.	7500°K	39
	Heat of vapor.	58, 3 Cal/mol	5
	Density	$10, 53-9, 86 \cdot 10^{-4} \cdot T \text{ g/cm}^3$	48, 49, 84-91, 99
	Viscosity	$0, 316 \exp 6400/RT \text{ cP}$	44, 49, 97-99, 143
	Surface tension	$1136-0, 174 T \text{ dyne/cm}$	76, 110-112, 150
	Vapor pressure	$\log P = 8, 06-12720/T \text{ torr}$	4, 5, 46, 47
	Vapor density	$1, 73 \cdot 10^{-3} P/T \text{ (P in torr)}$	
	approx. formula	$5, 30-12720/T - \log T$	
	Vapor viscosity	$4, 16 \cdot 10^{-5} \sqrt{T}/\Omega \text{ (2, 2)* poise}$	125, 61
	approx. formula	$3, 0 \cdot 10^{-7} \cdot T + 8, 5 \cdot 10^{-5}$	

2. 6. 1. The viscosity of silver

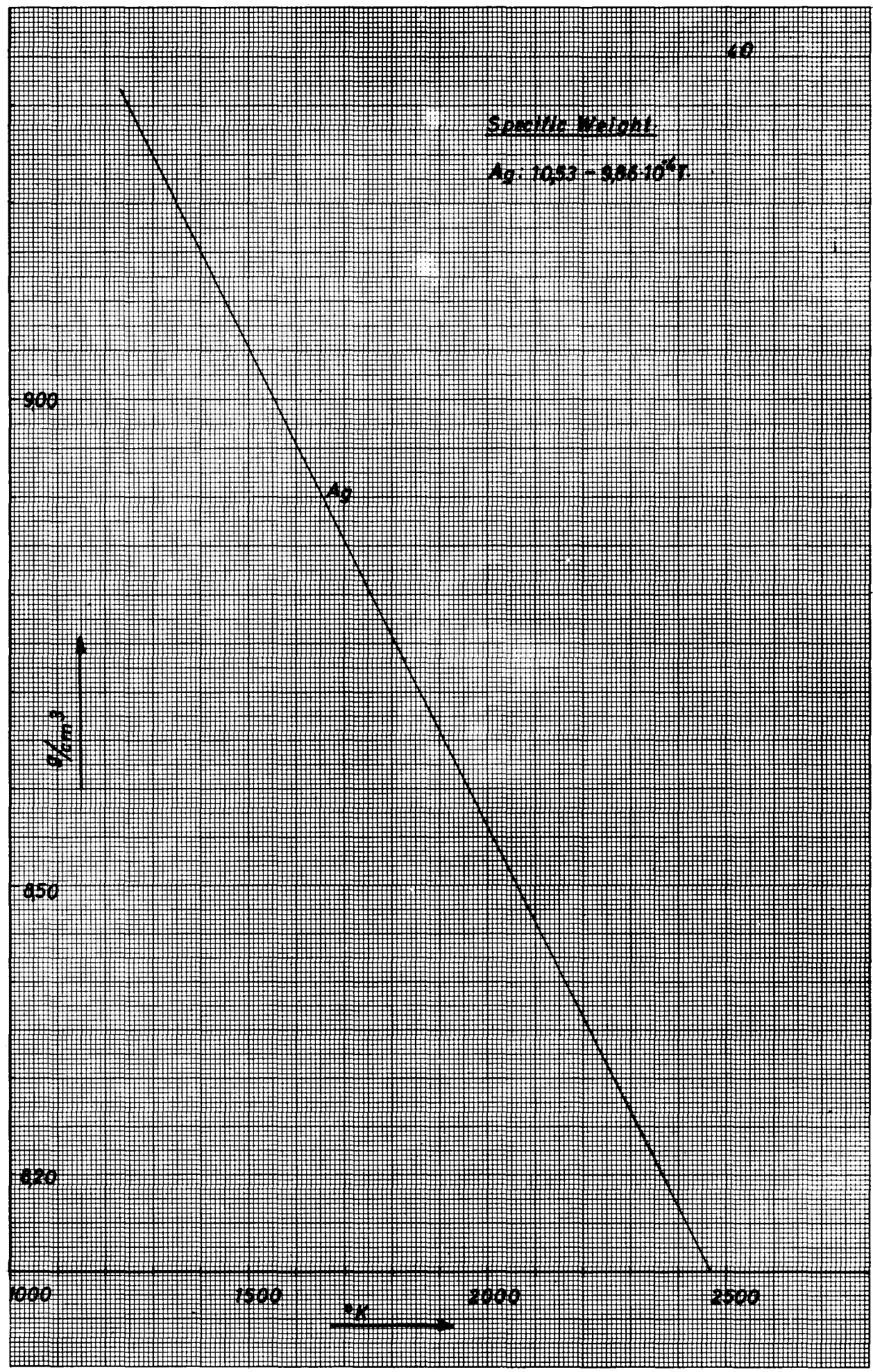
The values of Grosse (97), Anderson (143) and Wertman (99) coincide and give a greater inclination, i. e. H-value, than those of Sauerwald (44, 98) and Gebhardt (49).

2. 6. 2. The density of silver

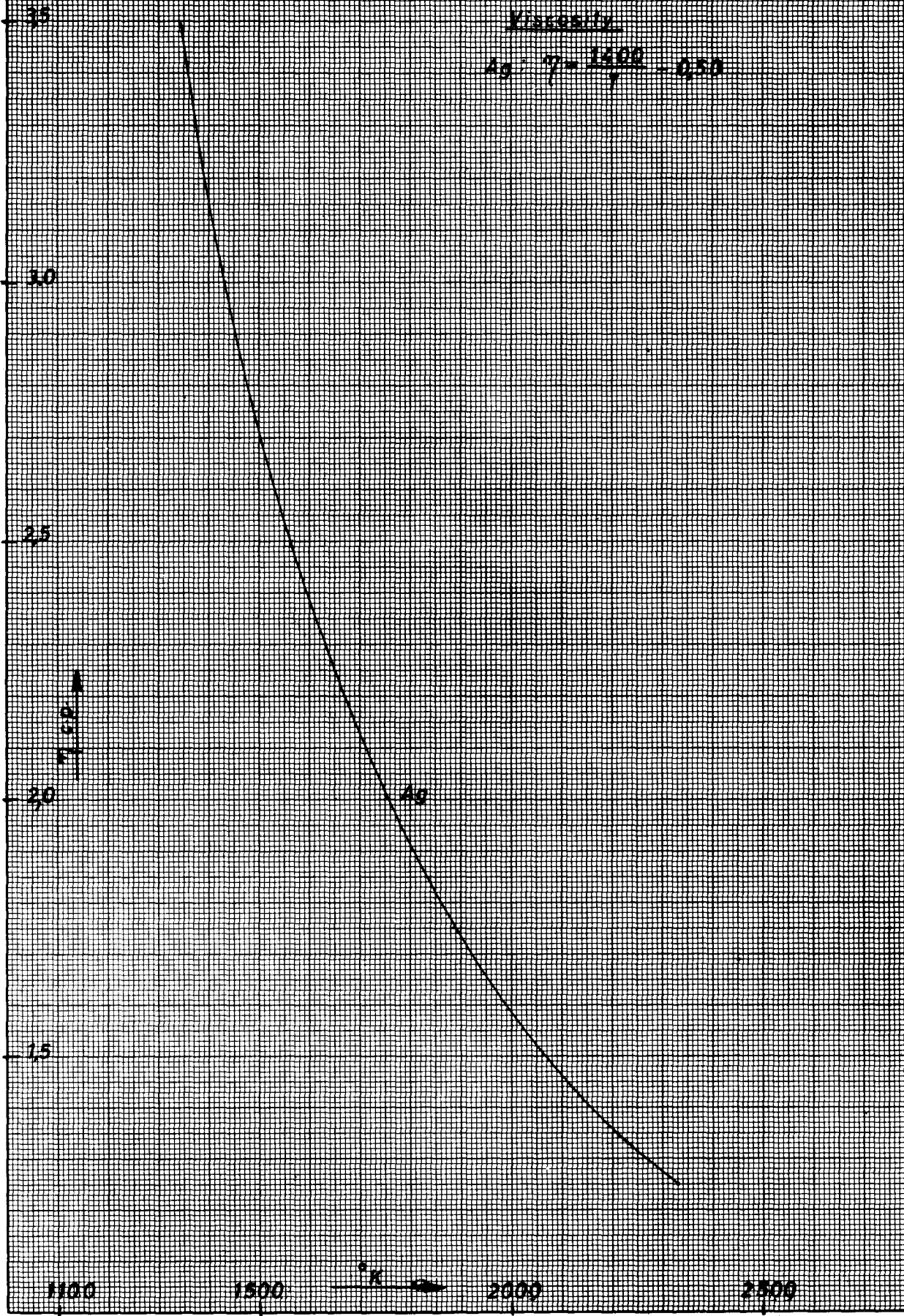
Author	Year	Method	Max. Temp.	Formula
(91)	1922	Archimedean		$10, 77-11, 74 \cdot 10^{-4} T$
(88)	1929	Pycnometer	1670°C	$10, 439-9, 279 \cdot 10^{-4} T$
(90)	1930	Archimedean	1300°C	$10, 50-9, 52 \cdot 10^{-4} T$
(85)	1930	"		$10, 543-10, 00 \cdot 10^{-4} T$
(89)	1951	"	1300°C	$10, 3347-8, 6222 \cdot 10^{-4} T$
(99)	1960	-	-	$10, 50-9, 75 \cdot 10^{-4} T$
(86)	1961	Max Bubble P.	1300°C	$10, 6213-10, 442 \cdot 10^{-4} T$
(84)	1961	Max Bubble P.	1150°C	$10, 666-10, 7 \cdot 10^{-4} T$
(48)	1962	Archimedean	2000°C	$10, 465-9, 067 \cdot 10^{-4} T$

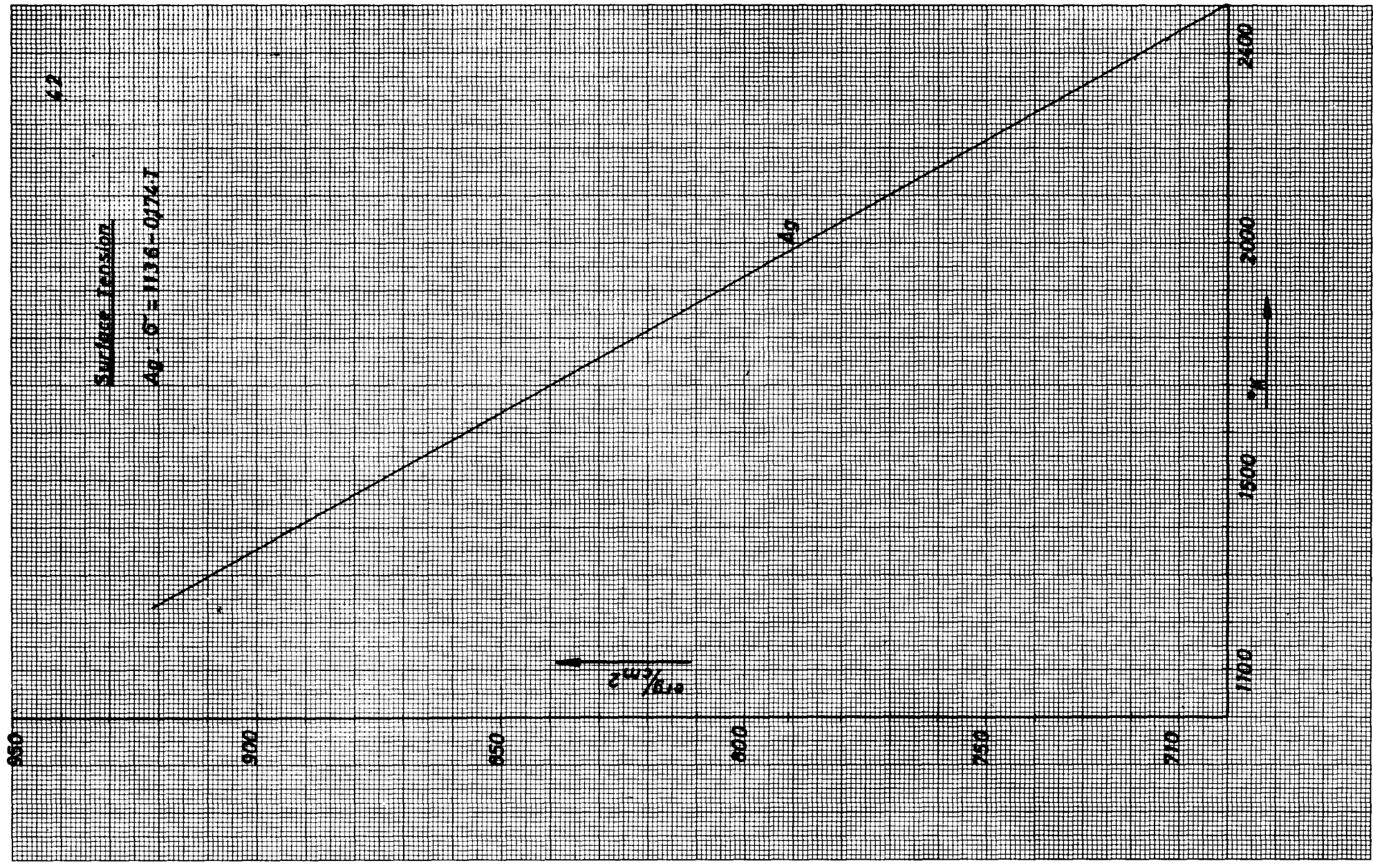
Specific Weight

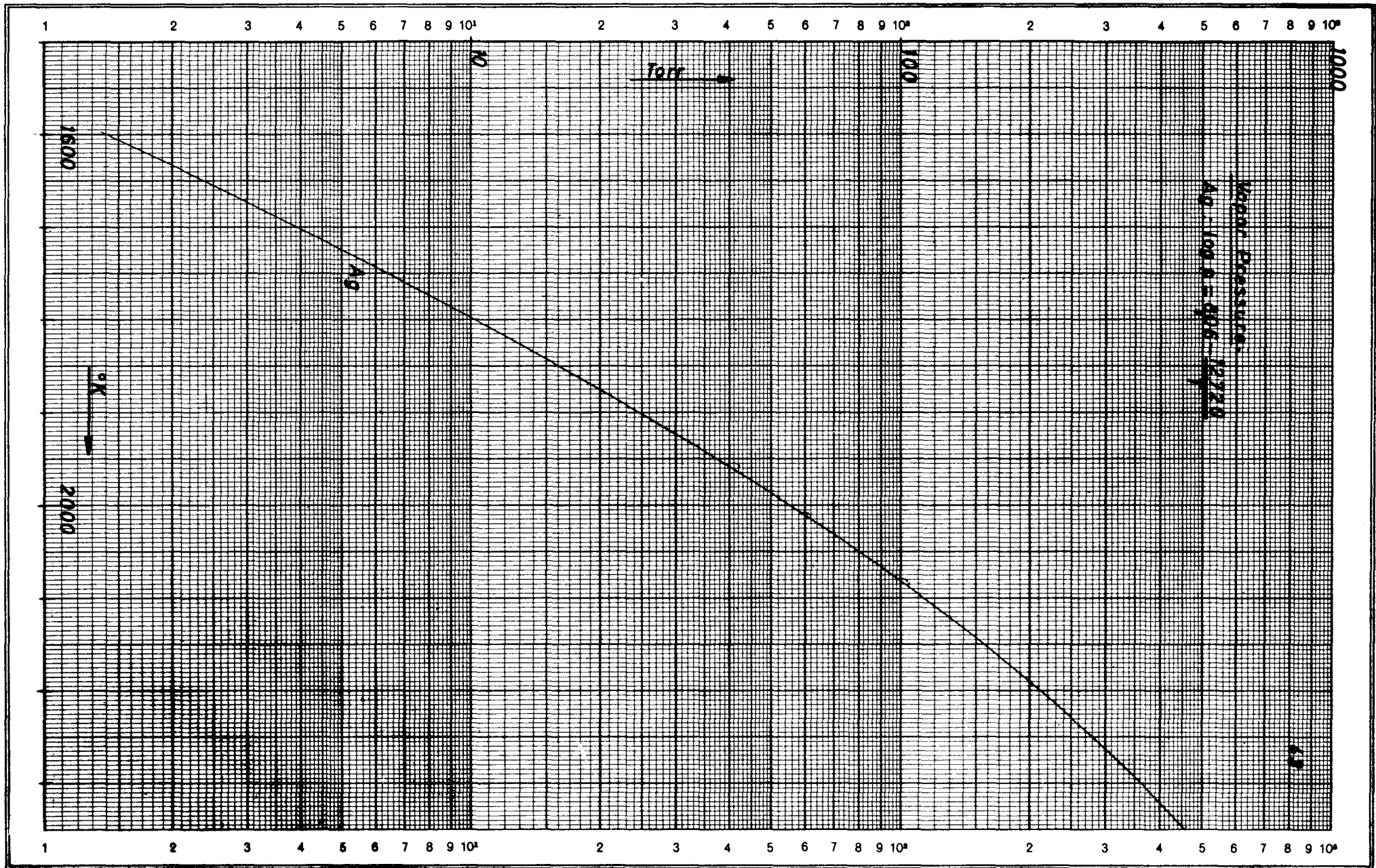
$\gamma = 10,53 - 0,56 \cdot 10^{-4} T$



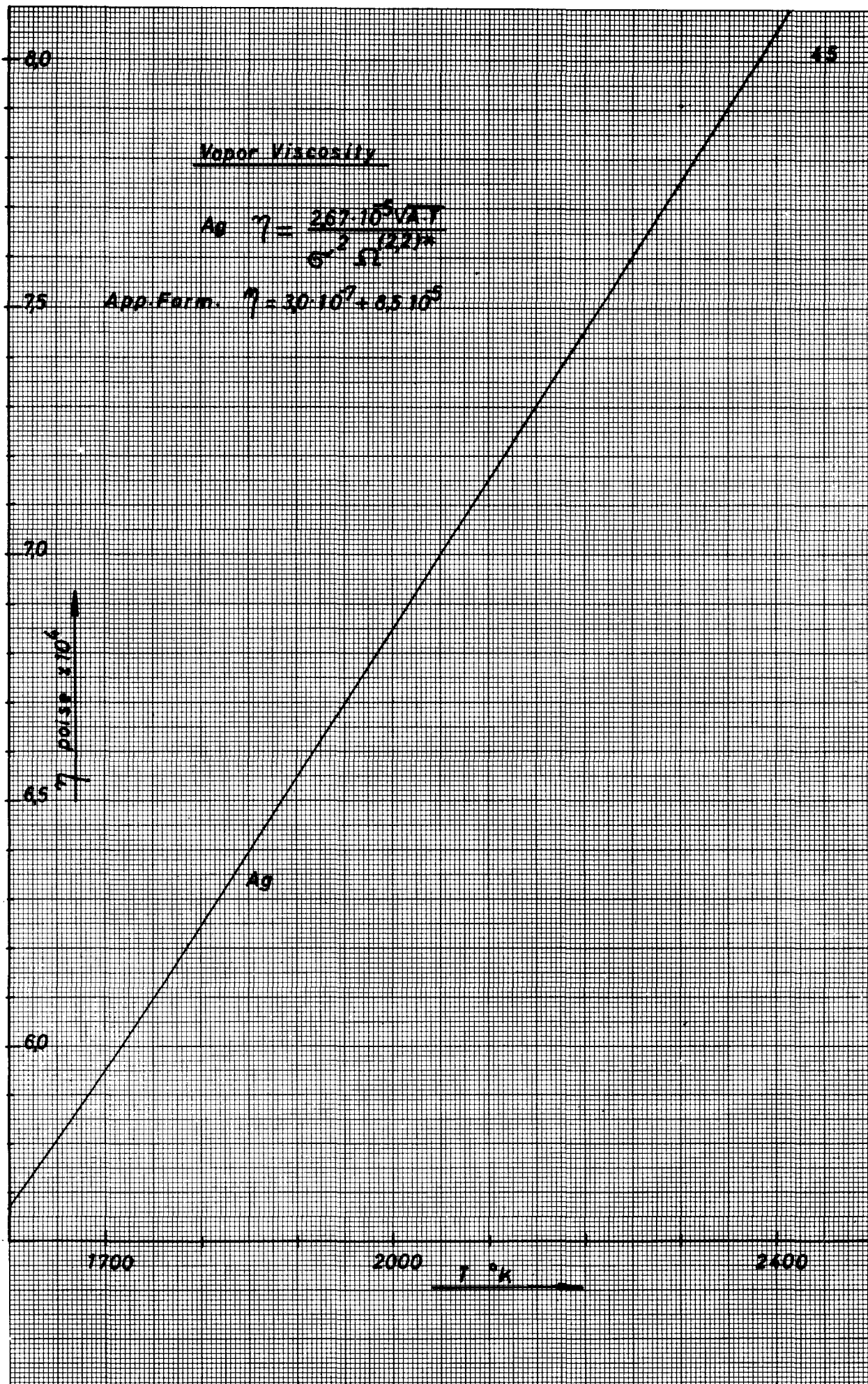
Viscosity
 $\eta = \frac{1400}{T} - 0.50$







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2.7. Calcium	Value / Formula	References
Atomic number	20	
Atomic weight	40,08	
Atomic diameter	3,50 Å	132
Melting point	1123°K	17
Boiling point	1762°K	5
Critical temp.	4700°K	this work
Heat of vapor.	37,1 Cal/mol	5
Density	$1,613-2,21 \cdot 10^{-4} T \text{ g/cm}^3$	76,144
Viscosity	$0,524 \exp 5000/RT \text{ cP}$	144
Surface tension	$472-0,100 T \text{ dyne/cm}$	76,105
Vapor pressure	$\log P = 7,48-8110/T \text{ torr}$	5
Vapor density	$6,44 \cdot 10^{-4} P/T \text{ (P in torr)}$	
approx. formula	$4,29-8110/T - \log T$	
Vapor viscosity	$1,39 \cdot 10^{-5} \sqrt{T/\Omega}^{(2,2)*} \text{ poise}$	125,61
approx. formula	$1,10 \cdot 10^{-7} \cdot T + 2,5 \cdot 10^{-5}$	

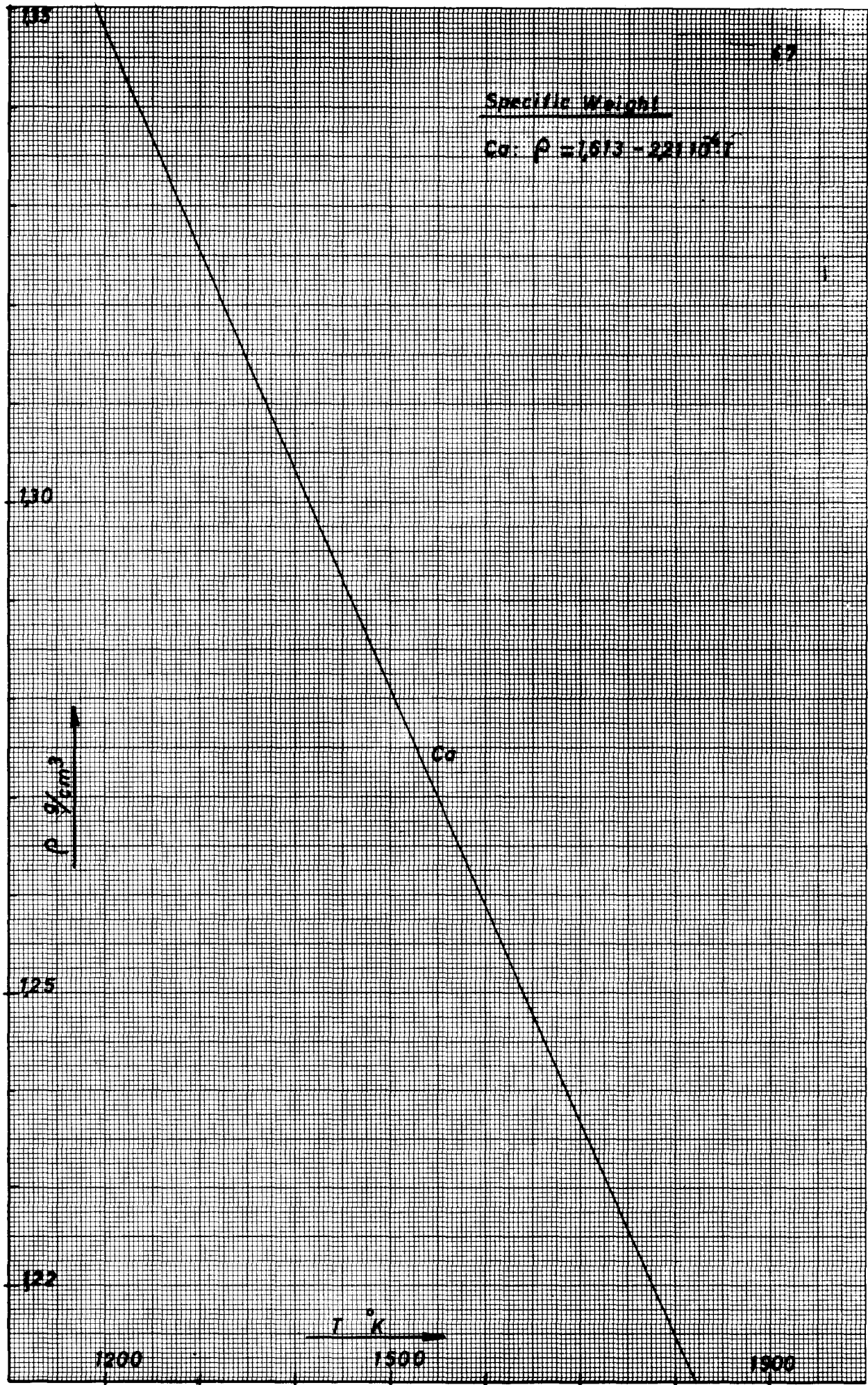
2.7.1. The viscosity of calcium

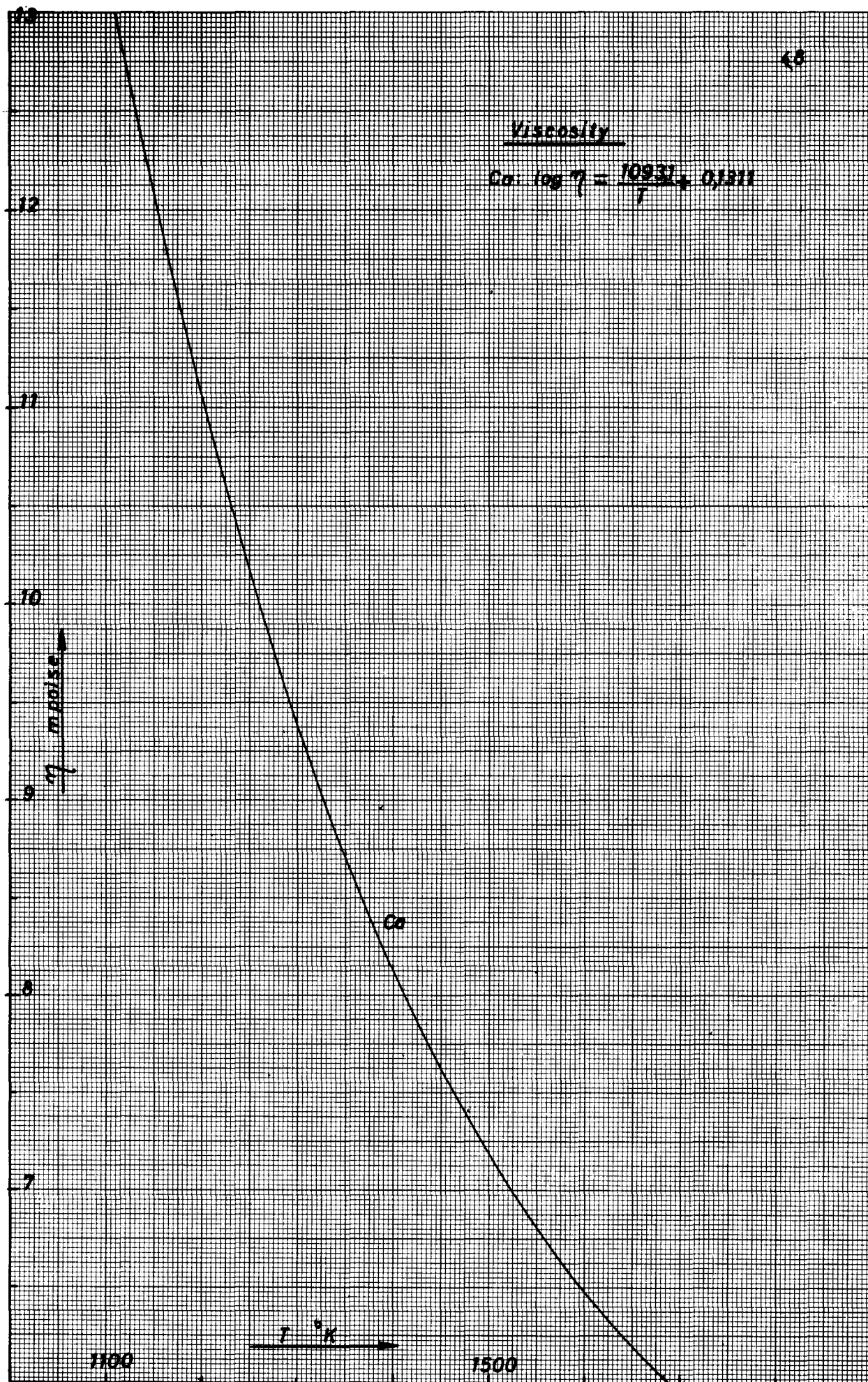
The measurements of Culpin (144) can be represented by

$\eta = 0,524 \exp 6790/RT \text{ (mP)}$. Confronted with Grosse's plot of H-values (22) it appears extremely high. Because of the fact that Culpin's density measurements are incorrect, we preferred the theoretical approach of Andrade and Grosse (12, 22).

2.7.2. The density of calcium

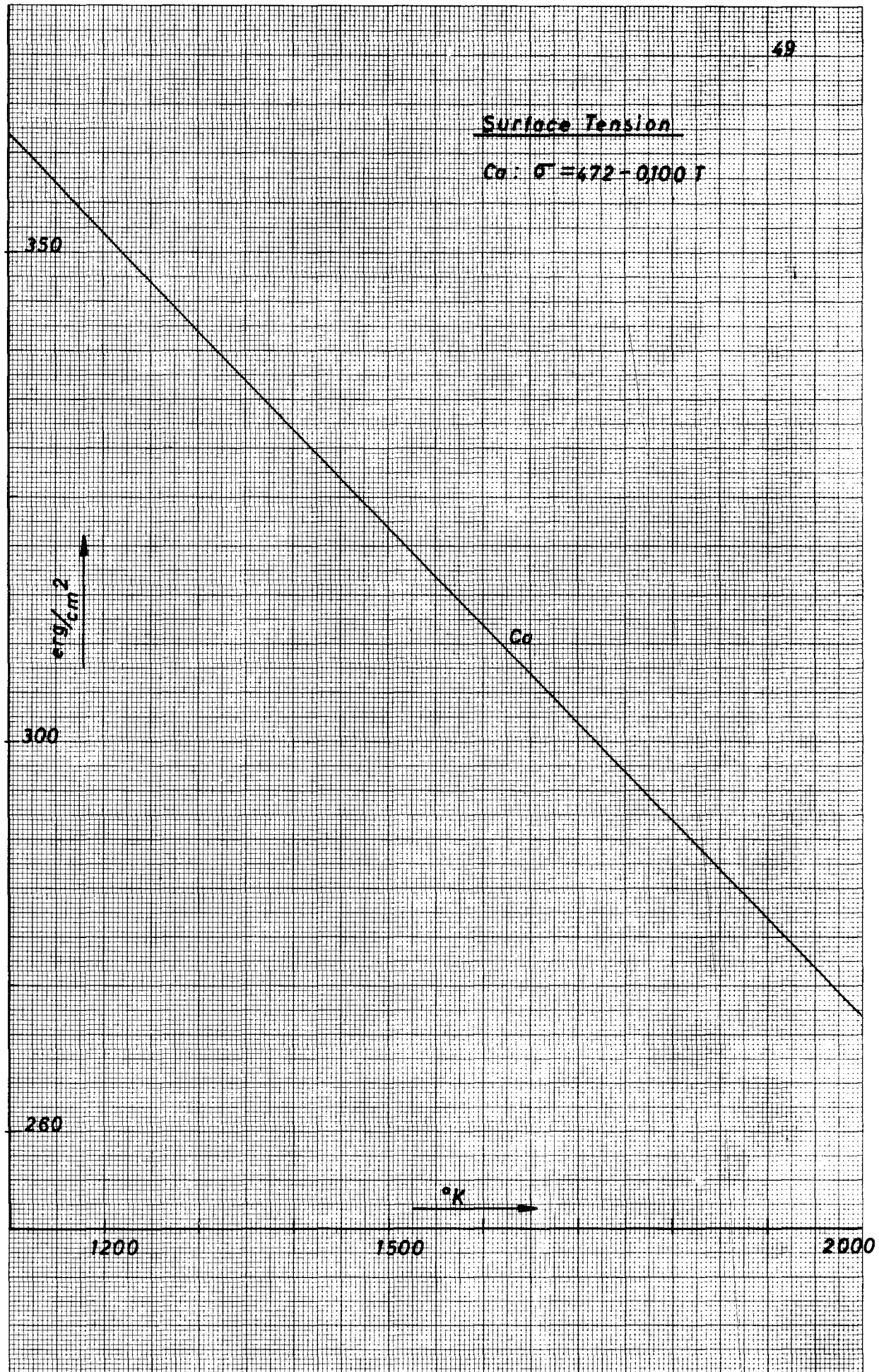
Author	Year	Method	Max. Temp.	Formula
(144)	1957	Pycnometer	880°C	$2,36-8,87 \cdot 10^{-4} T$
(76)	1967	Max. Bubble P.	1380°C	$1,613-2,21 \cdot 10^{-4} T$
see also (20, 21)				

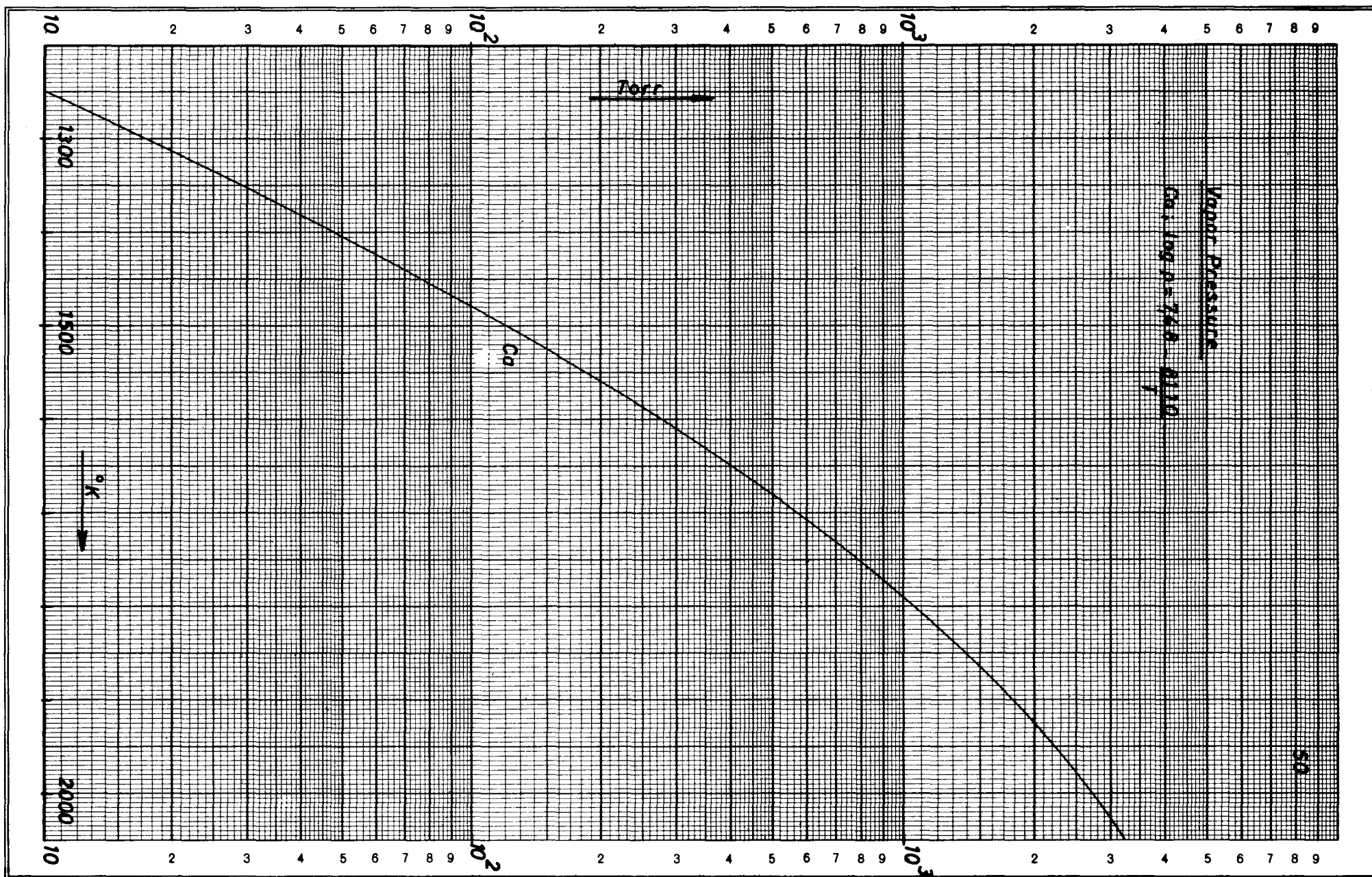




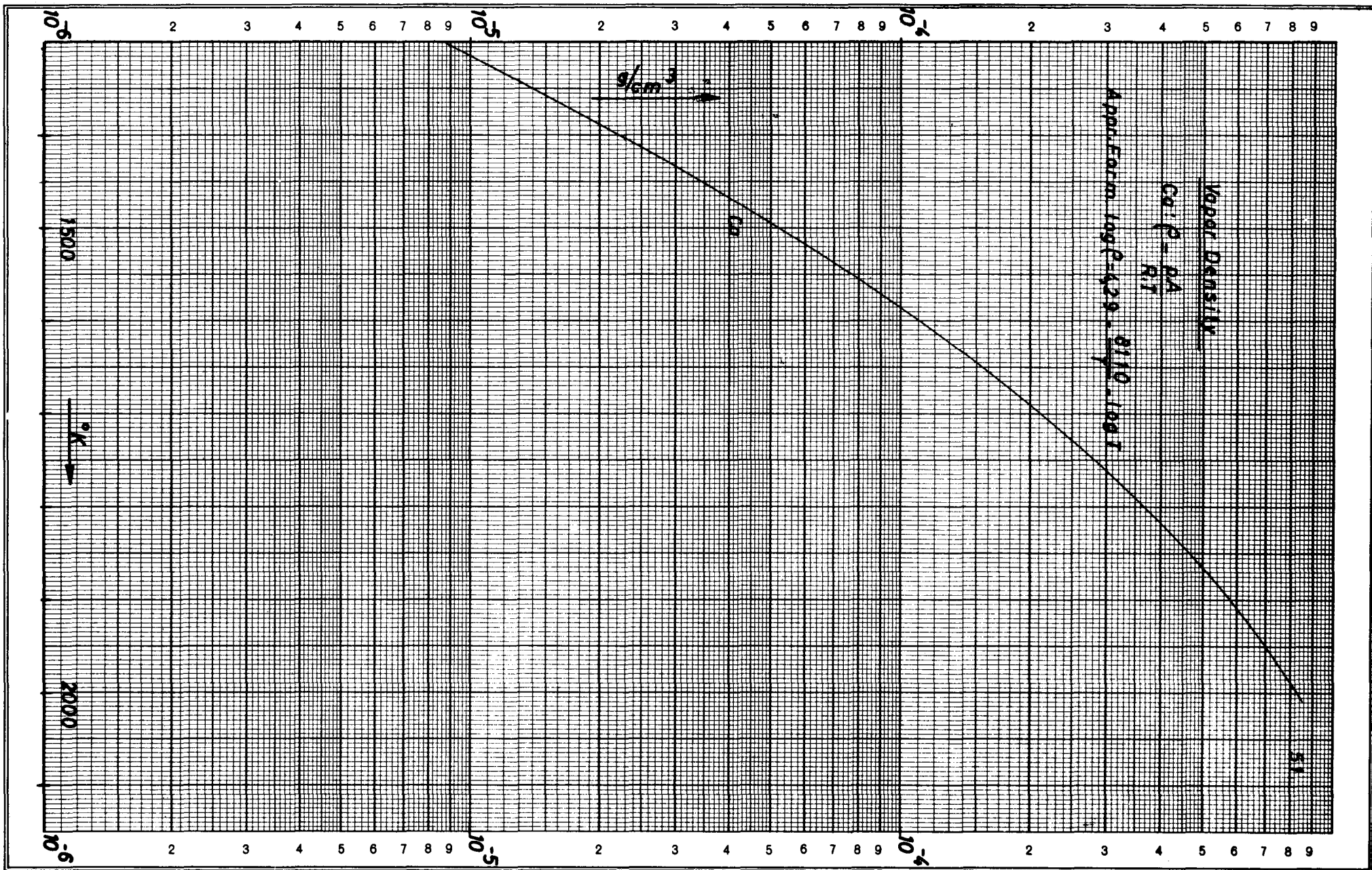
Surface Tension

Co: $\sigma = 472 - 0,100 T$

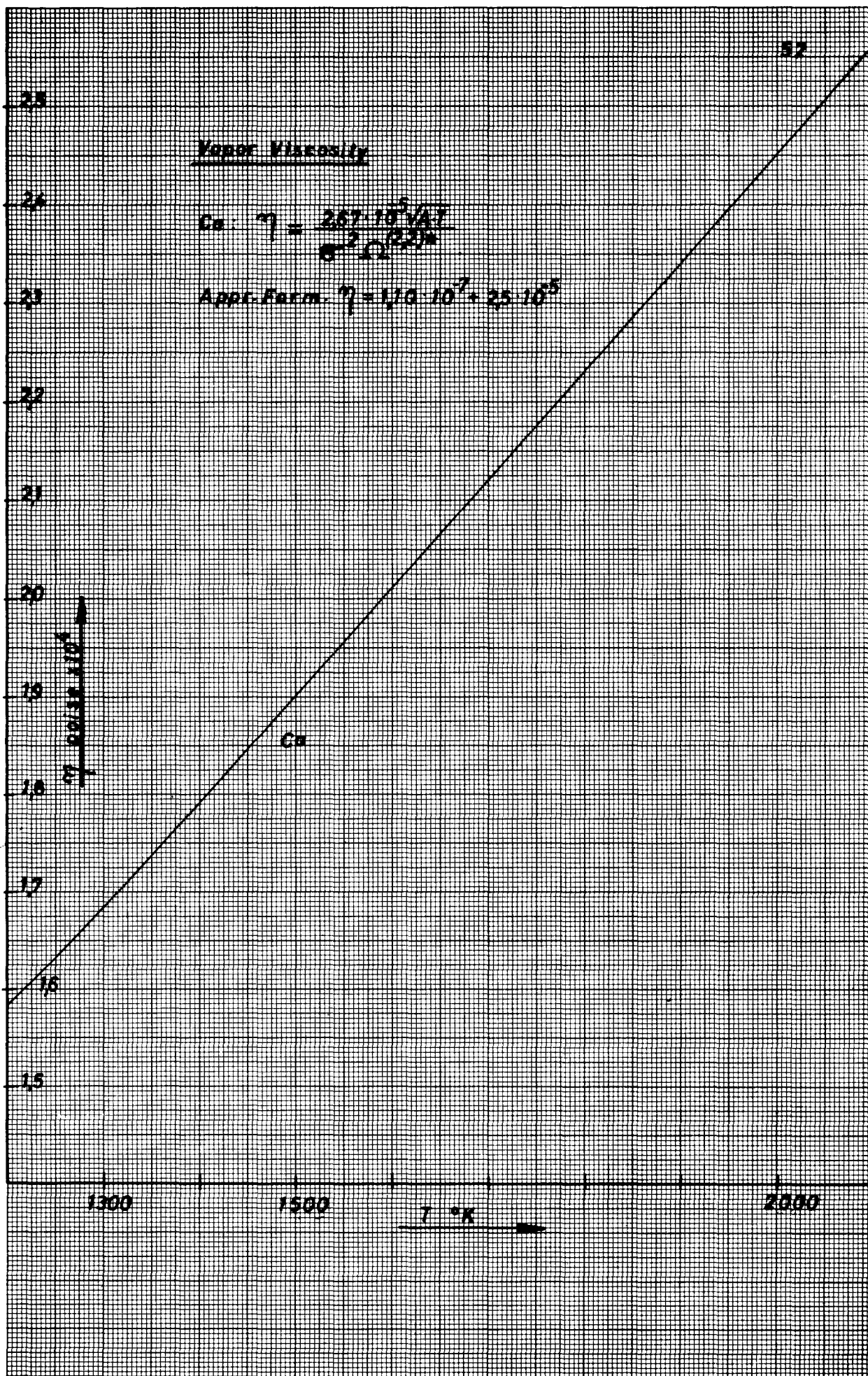




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2.8.	Strontium	Value / Formula	References
	Atomic number	38	
	Atomic weight	87, 63	
	Atomic diameter	3, 80 Å	132
	Melting point	1043°K	17
	Boiling point	1649°K	5
	Critical temp.	4300°K	this work
	Heat of vapor.	33, 7 Cal/mol	5
	Density	$2, 648-2, 62 \cdot 10^{-4} T \text{ g/cm}^3$	76
	Viscosity	$0, 145 \exp 4900 / RT \text{ cP}$	
	Surface tension	$392-0, 085 T \text{ dyne/cm}$	76, 147
	Vapor pressure	$\log P = 7, 35-7370/T \text{ torr}$	5
	Vapor density	$1, 41 \cdot 10^{-3} P/T \text{ (P in torr)}$	
	approx. formula	$4, 50-7370/T - \log T$	
	Vapor viscosity	$1, 73 \cdot 10^{-5} \sqrt{T/\Omega}^{(2, 2)*} \text{ poise}$	125, 61
	approx. formula	$1, 36 \cdot 10^{-7} \cdot T + 3, 1 \cdot 10^{-5}$	

2.8.1. The viscosity of strontium

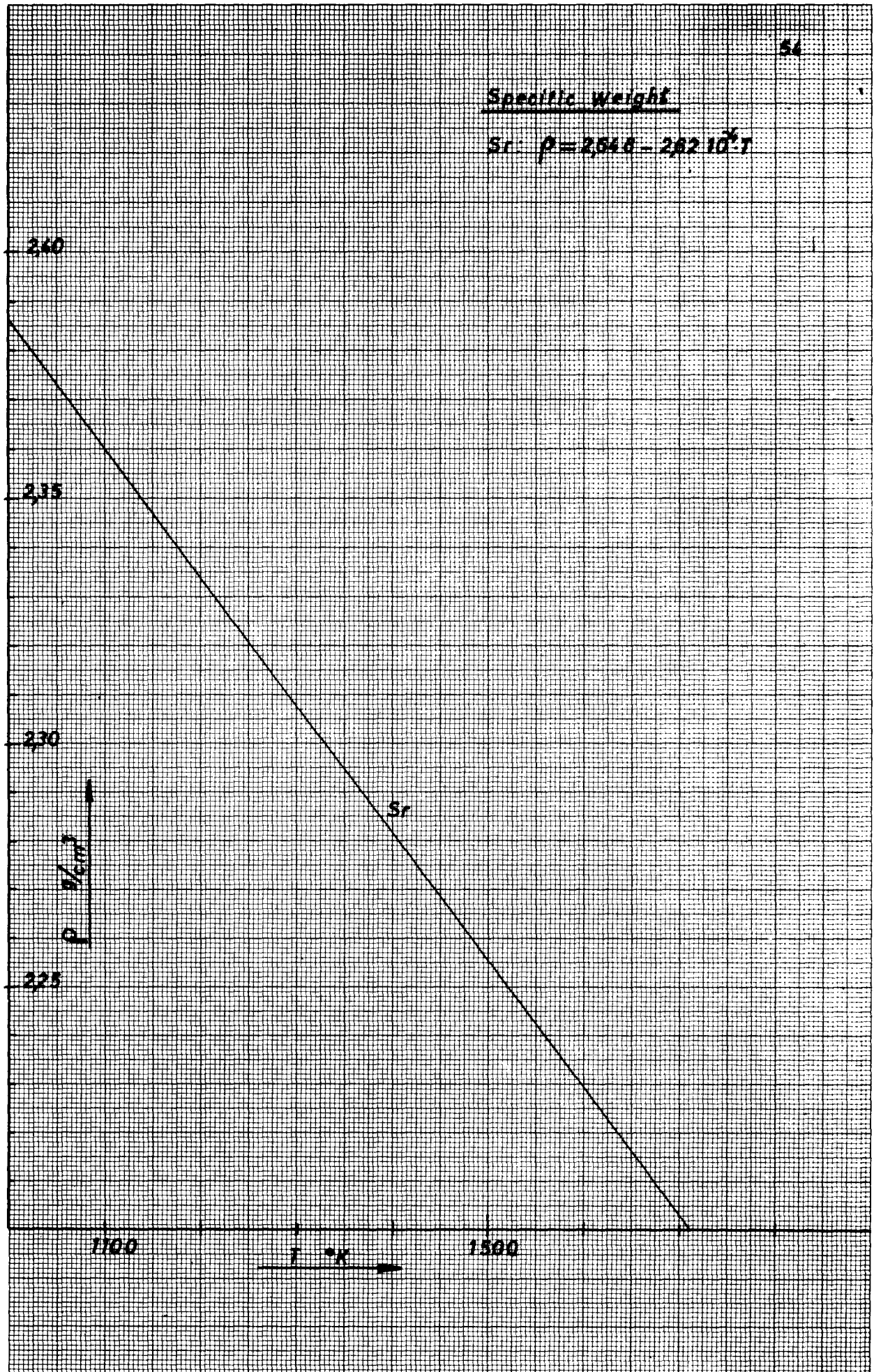
No measurements are known to the author, so we calculated the viscosity with the Andrade-Grosse approach (12, 22).

2.8.2. The density of strontium

Author	Year	Method	Max. Temp.	Formula
(76)	1967	Max Bubble P.	1330°C	$2, 648-2, 62 \cdot 10^{-4} T$

Specific weight

Sr: $\rho = 2,545 - 2,82 \cdot 10^{-3} \cdot T$



Viscosity

$$S_r: \log \eta = \frac{10.713}{T} + 0.1658$$

150

140

130

120

110

100

90

80

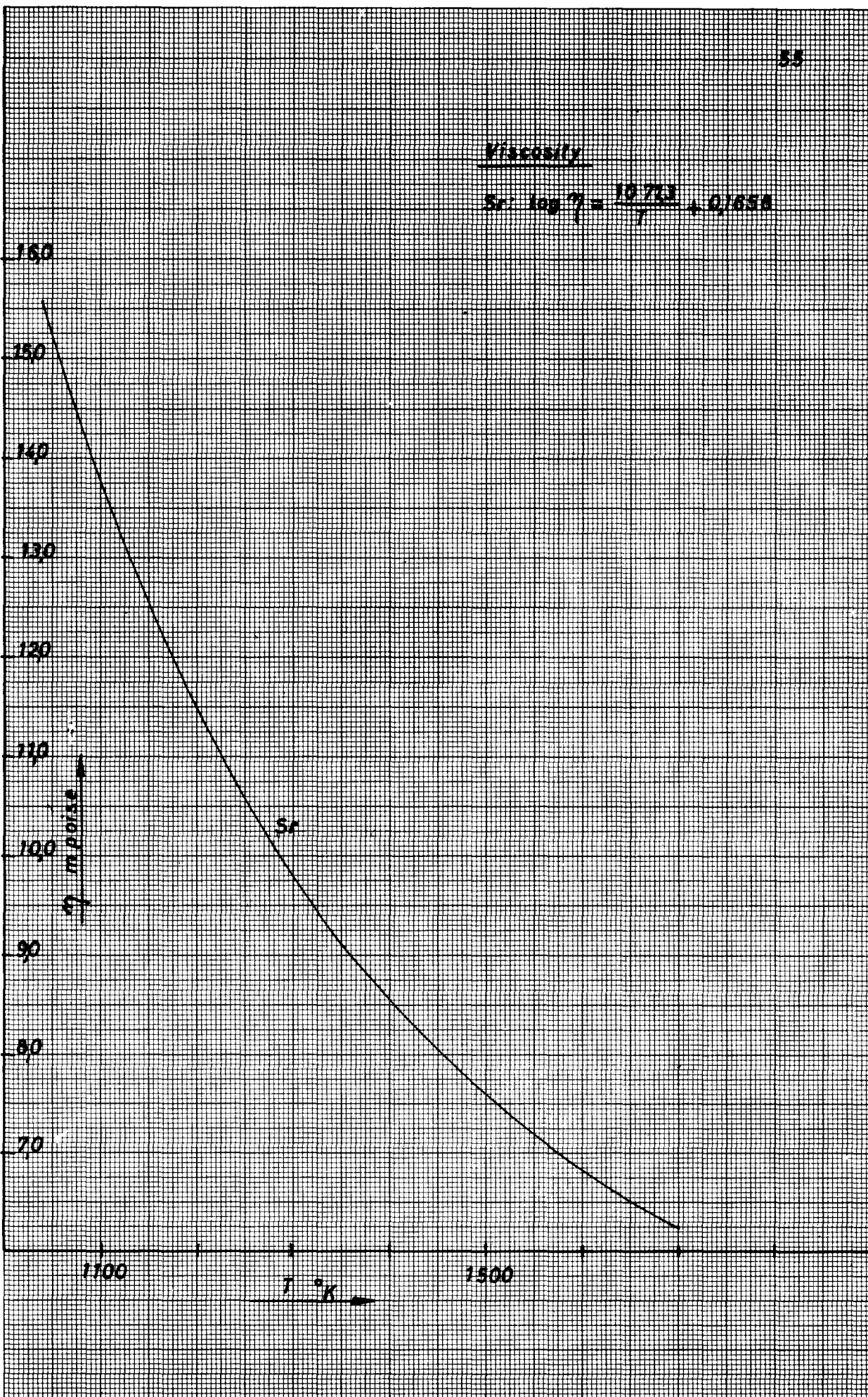
70

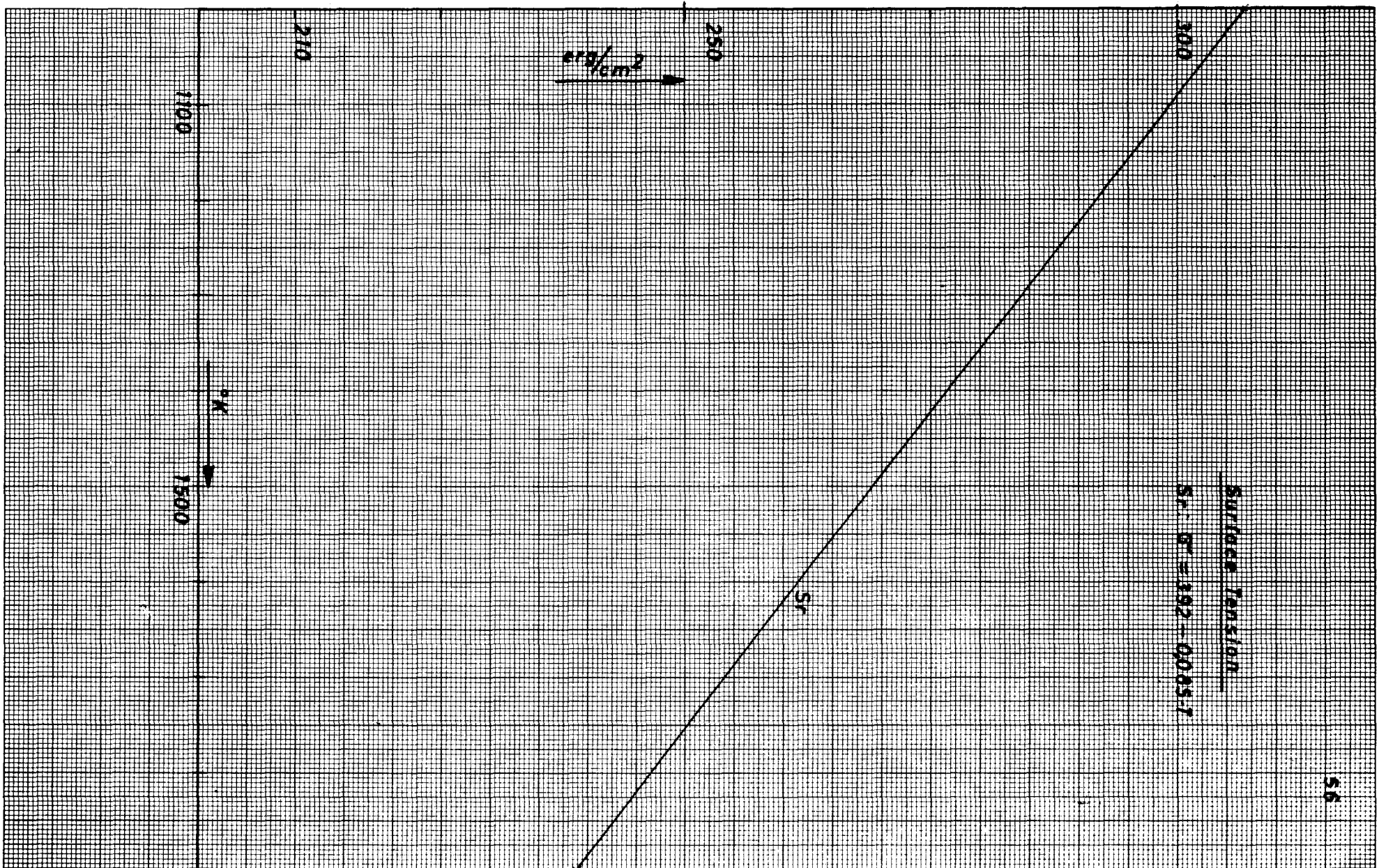
 η mpoise

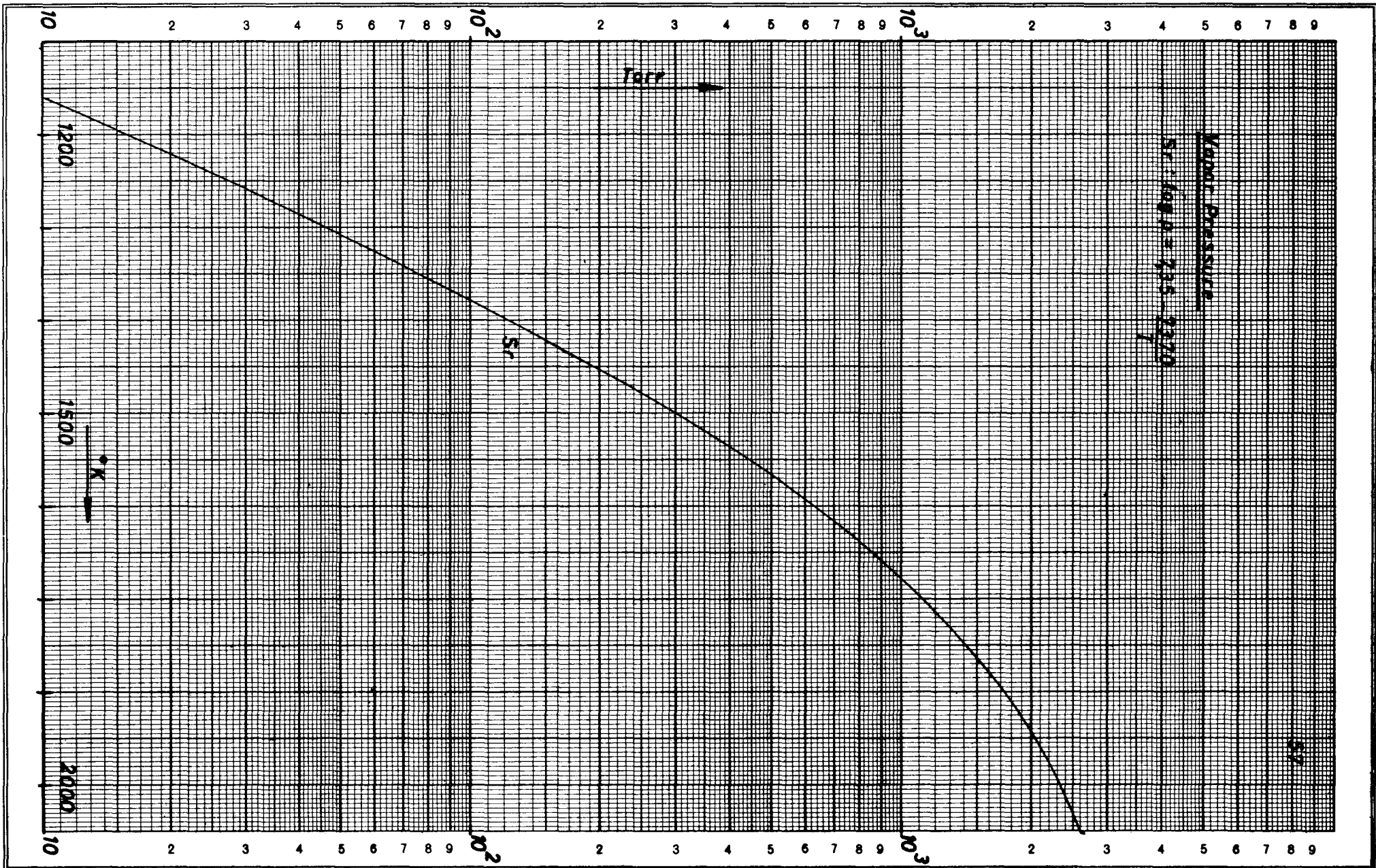
1100

 T °K

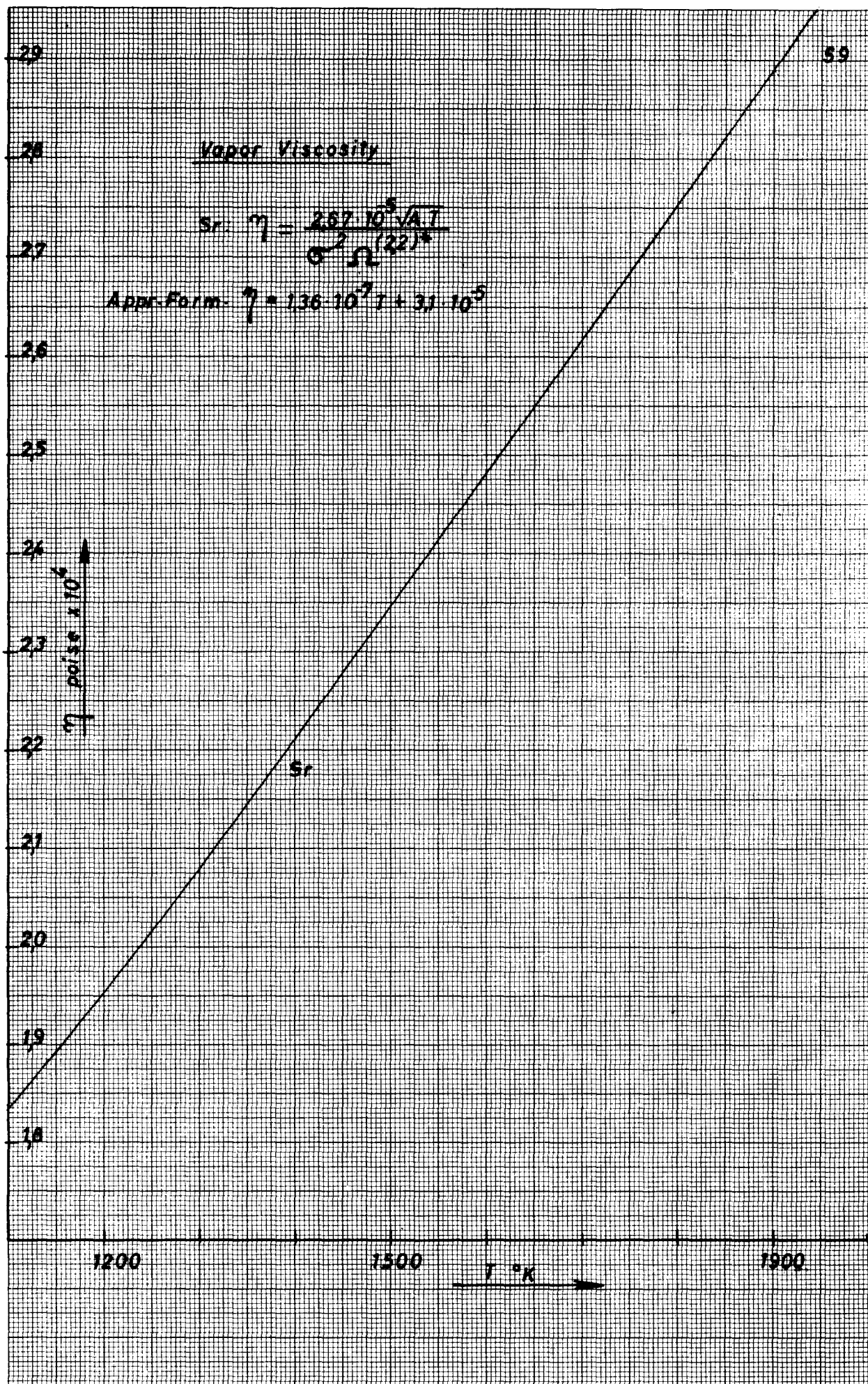
1500

S_r





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2.9.	Barium	Value / Formula	References
	Atomic number	56	
	Atomic weight	137,36	
	Atomic diameter	$3,90 \text{ \AA}$	132
	Melting point	983°K	17
	Boiling point	2063°K	5
	Critical temp.	5150°K	20,21
	Heat of vapor.	39,3 Cal/mol	5
	Density	$3,59-2,74 \cdot 10^{-4} \cdot T \text{ g/cm}^3$	20,21,63,76
	Viscosity	$0,196 \exp 4300/RT \text{ cP}$	
	Surface tension	$351-0,075 T \text{ dyne/cm}$	76,105
	Vapor pressure	$\log P = 7,04-8590/T \text{ torr}$	5
	Vapor density	$2,205 \cdot 10^{-3} P/T \text{ (P in torr)}$	
	approx. formula	$4,38-8590/T - \log T$	
	Vapor viscosity	$2,06 \cdot 10^{-5} \sqrt{T}/\Omega^{(2,2)*} \text{ poise}$	125,61
	approx. formula	$1,62 \cdot 10^{-7} \cdot T + 4,3 \cdot 10^{-5}$	

2.9.1. The viscosity of barium

No measurements are known to the author, thus the viscosity was calculated with the Andrade-Grosse approach (15, 12, 22).

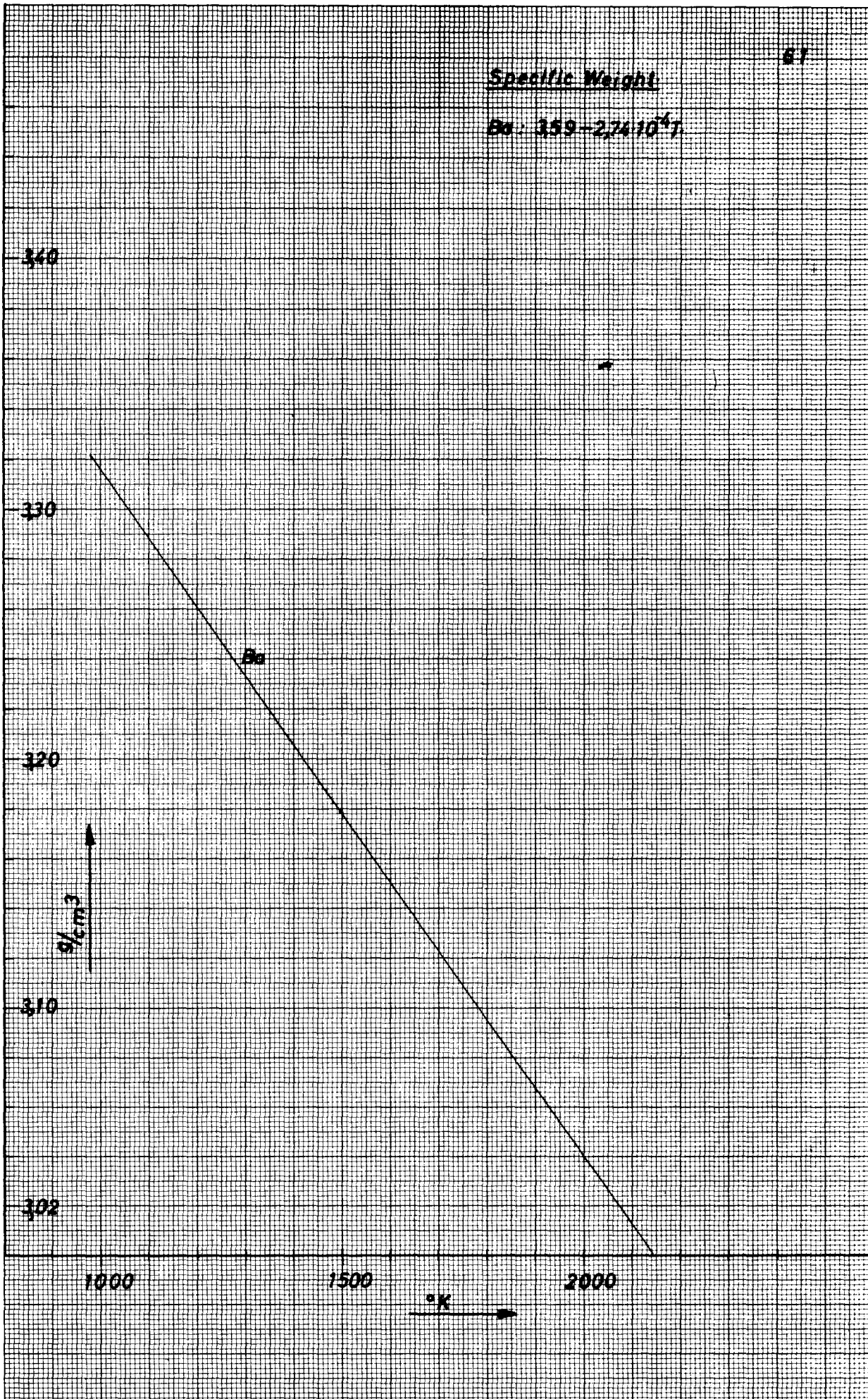
2.9.2. The density of barium

Author	Year	Method	Max. Temp.	Formula
(63)	1962	Archimedean	930°C	$3,53-2,14 \cdot 10^{-4} T$
(76)	1967	Max Bubble P.	1500°C	$3,59-2,74 \cdot 10^{-4} T$
see also (20,21)				

Specific Weight

67

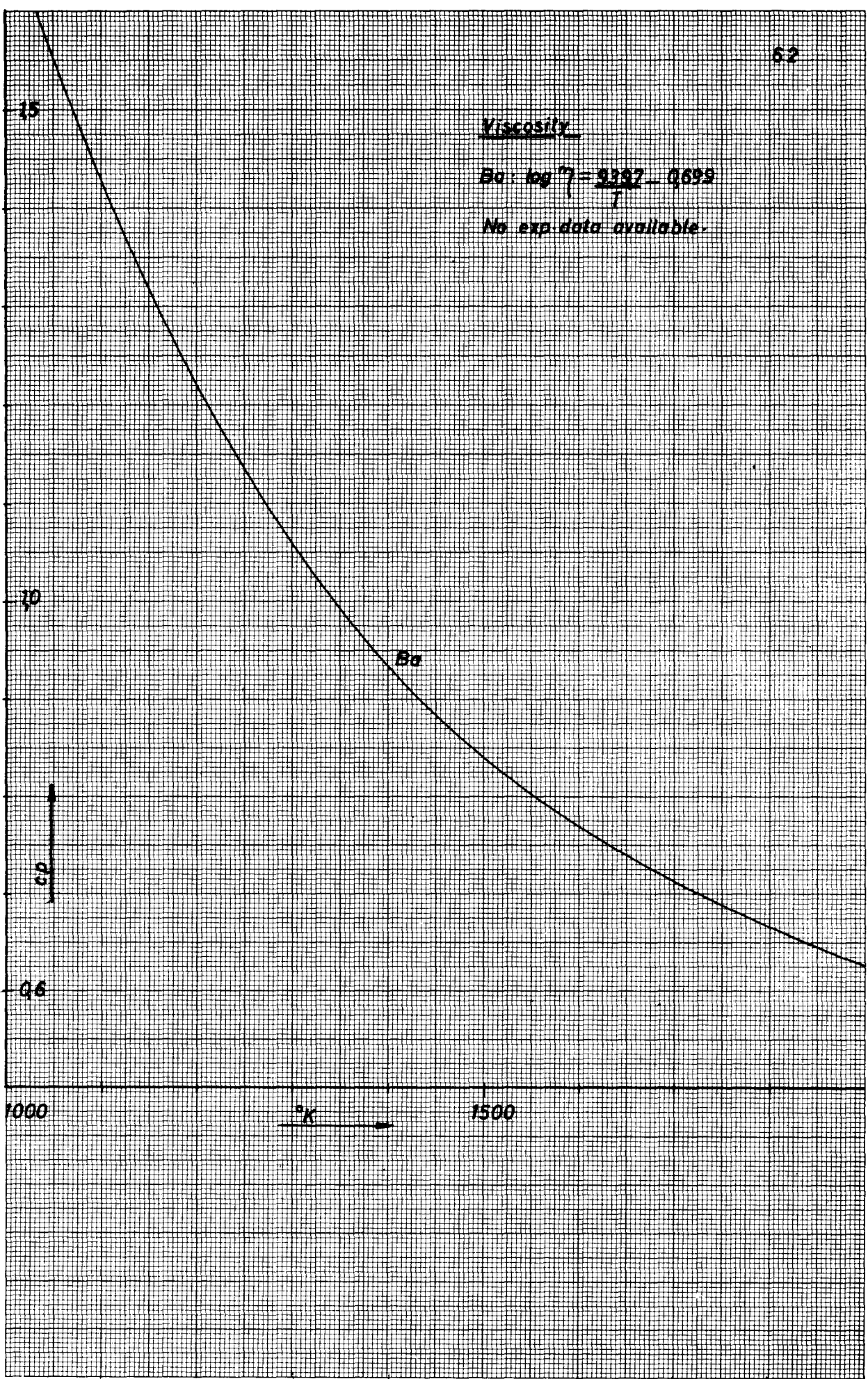
Eq. $359-27410^6$



Viscosity

$Ba: \log \eta = \frac{9397}{T} - 0.699$

No exp data available.



1.5

1.0

0.8

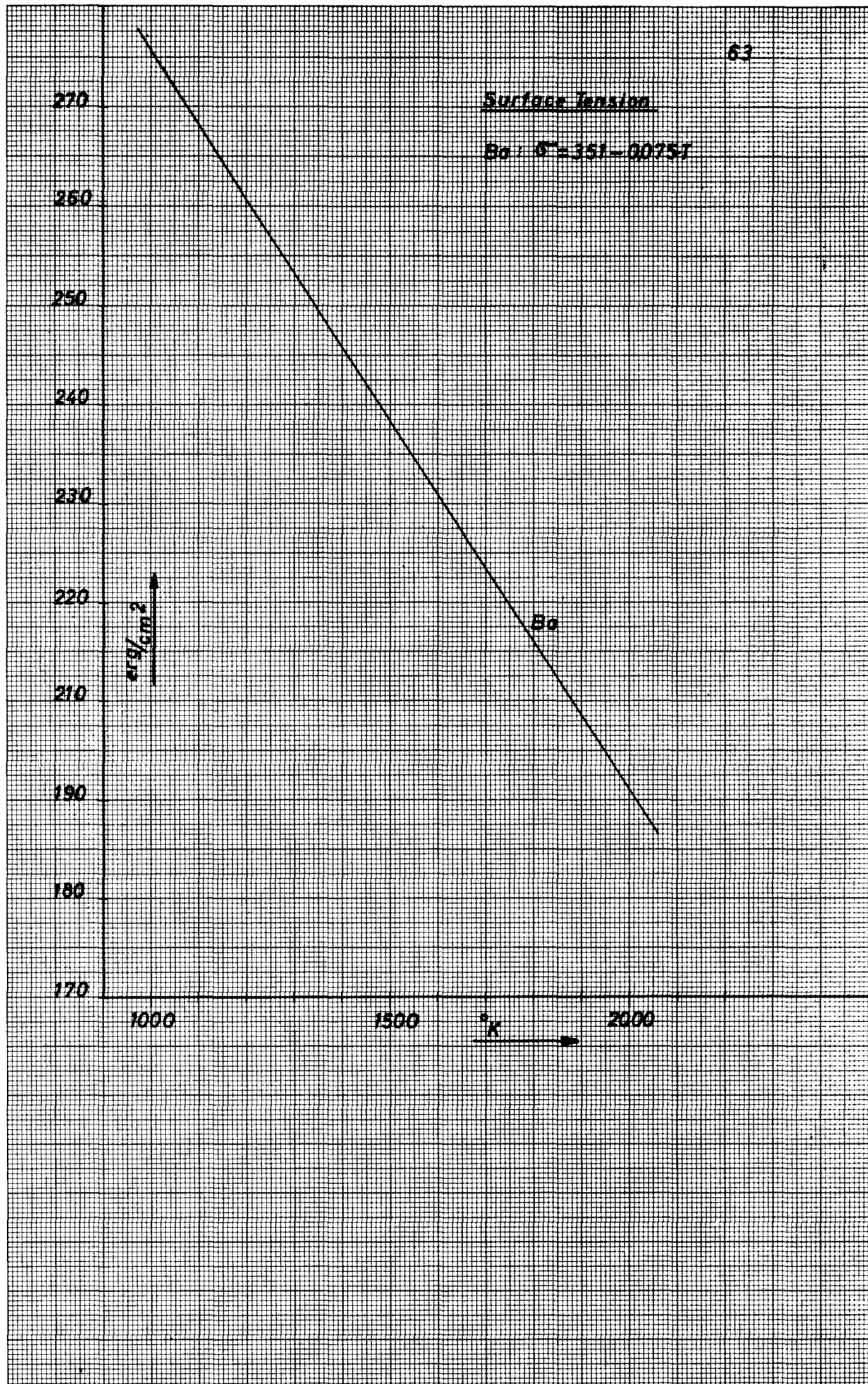
0.6

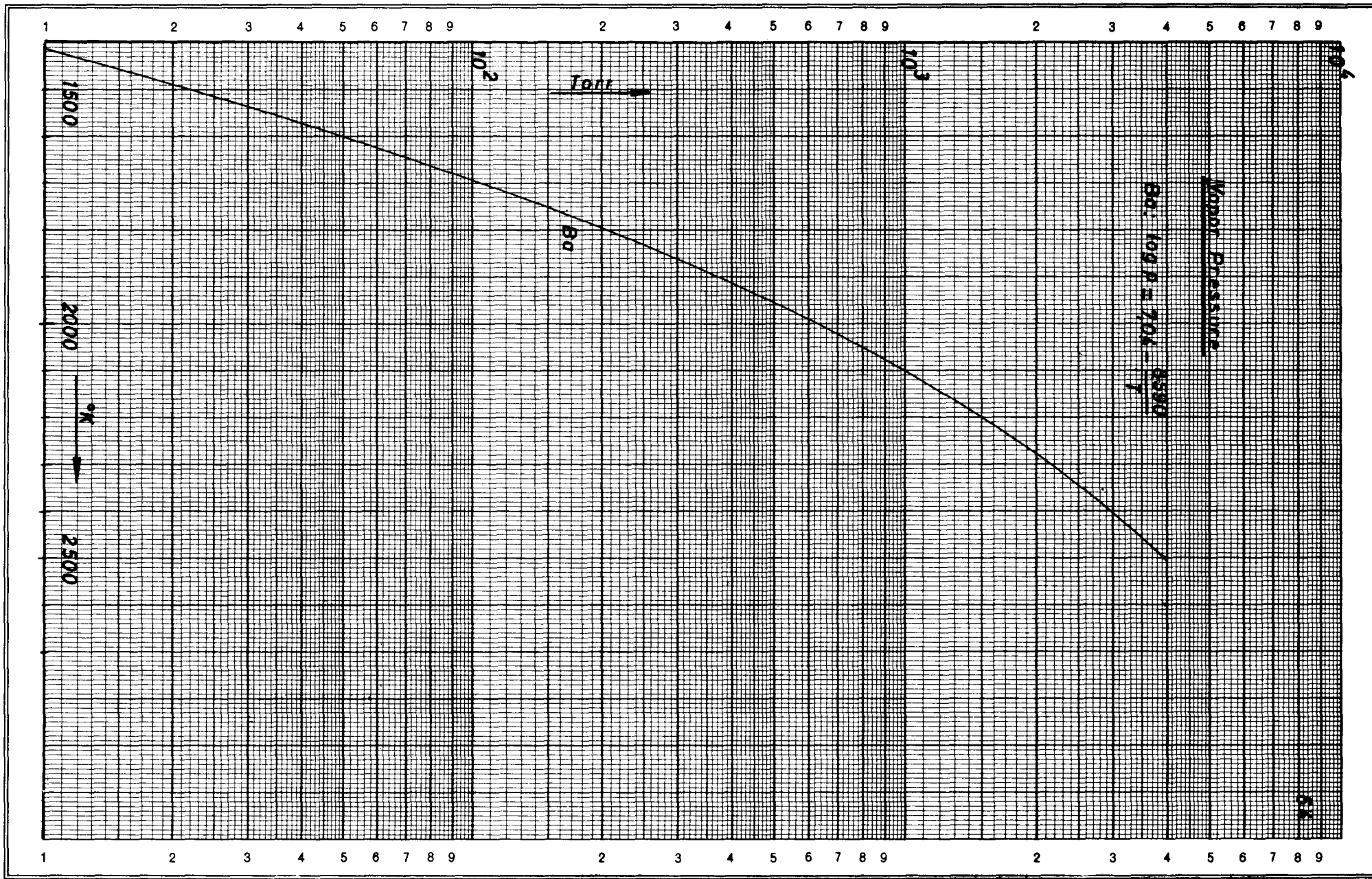
1000

°K

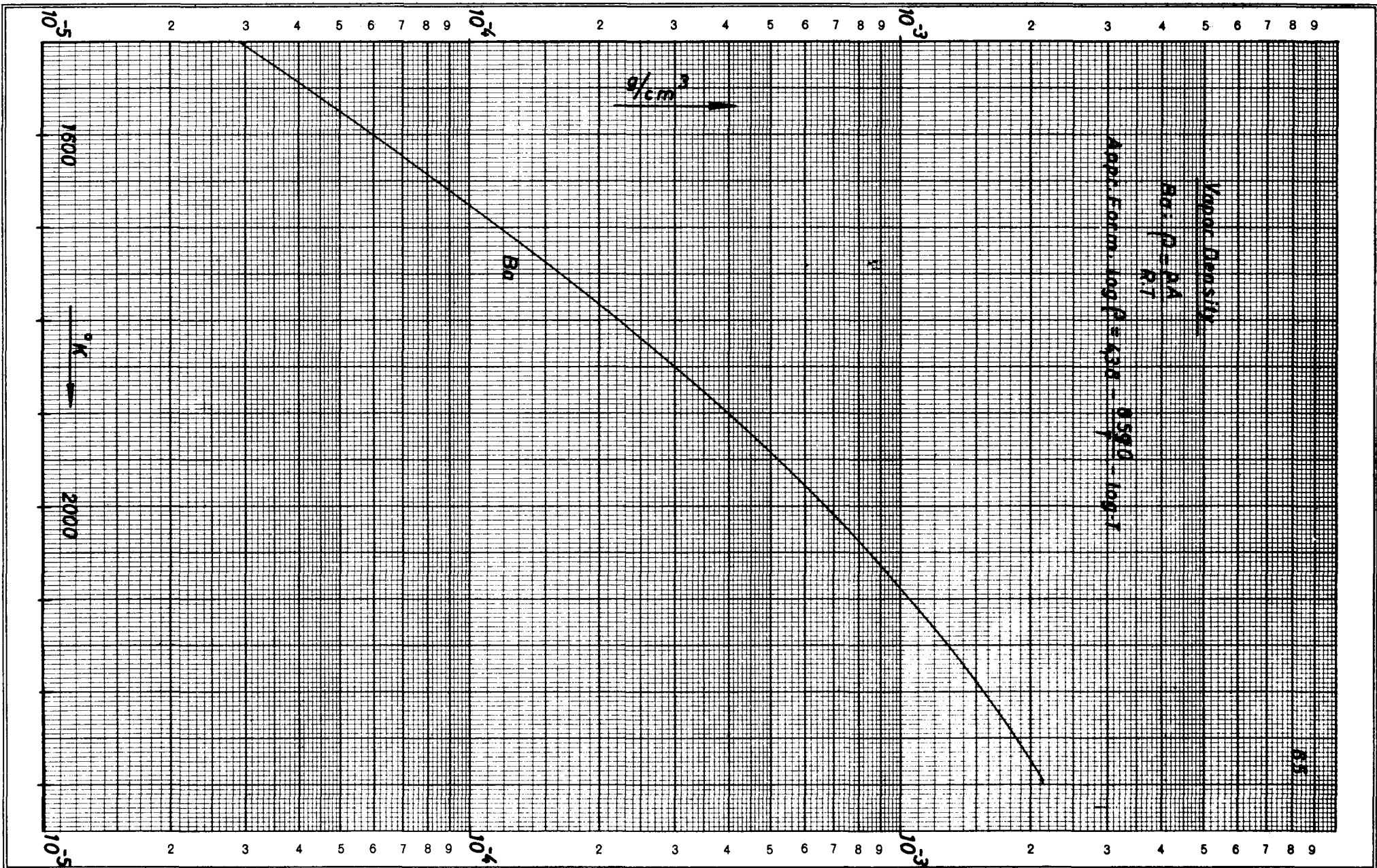
1500

Ba

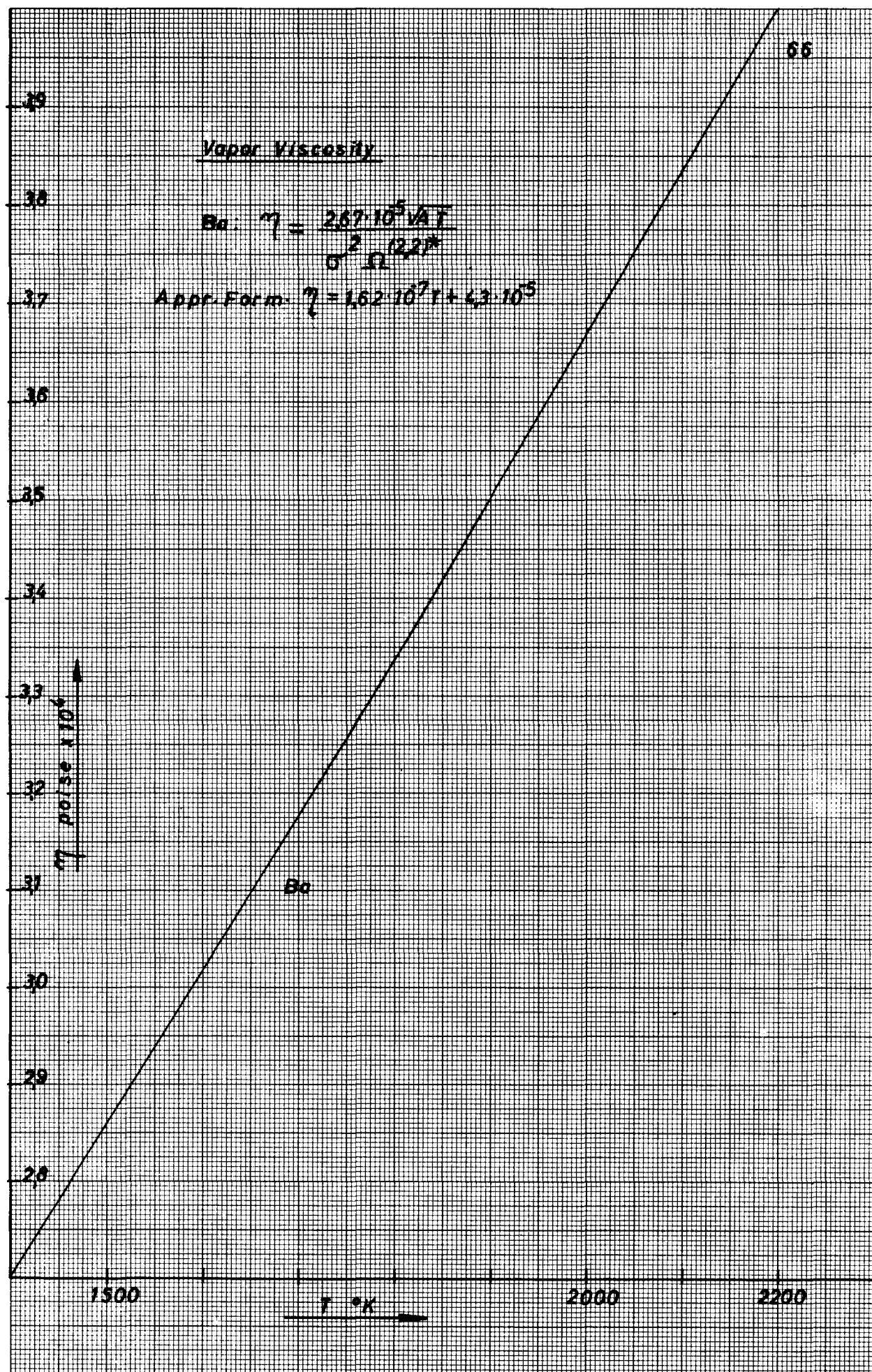




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2.10.

Thallium	Value / Formula	References
Atomic number	81	
Atomic weight	204,39	
Atomic diameter	$3,10 \text{ \AA}$	132
Melting point	577°K	17
Boiling point	1726°K	5
Critical temp.	4830°K	10
Heat of vapor.	42,4 Cal/mol	5
Density	$12,16-15,21 \cdot 10^{-4} \cdot T \text{ g/cm}^3$	41,42,58
Viscosity	$0,298 \exp 2500 / RT \text{ cP}$	60
Surface tension	$536-0,119 T \text{ dyne/cm}$	59,113,76
Vapor pressure	$\log P = 8,24 - 9260/T \text{ torr}$	5
Vapor density	$3,28 \cdot 10^{-3} P/T \text{ (P in torr)}$	
approx. formula	$5,76-9260/T - \log T$	
Vapor viscosity	$3,975 \cdot 10^{-5} \sqrt{T/\Omega}^{(2,2)*} \text{ poise}$	125,61
approx. formula	$4,5 \cdot 10^{-7} \cdot T + 1,0 \cdot 10^{-5}$	

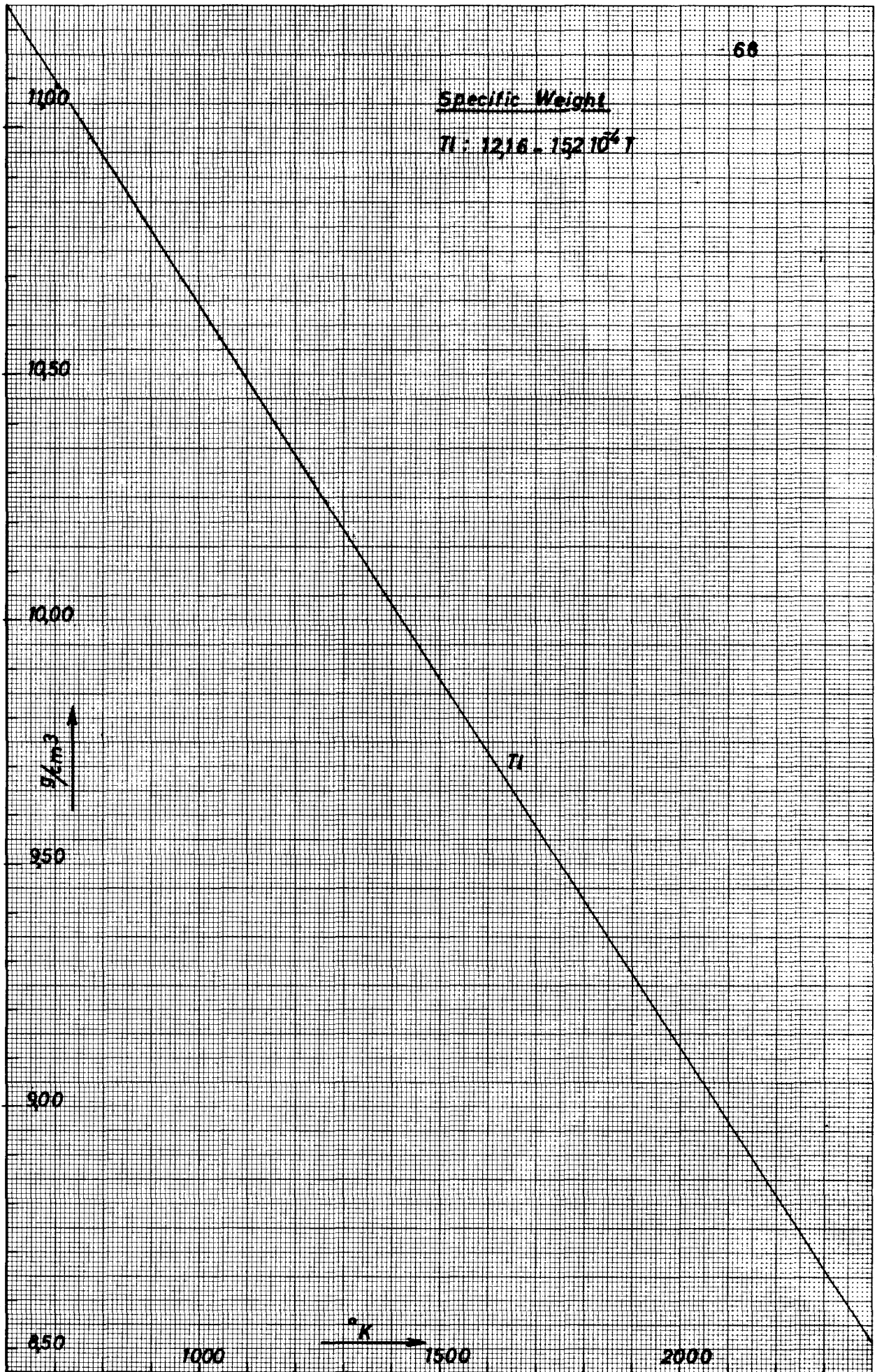
2.10.1. The viscosity of thallium

Only one measurement is reported (60). Nevertheless it seems reliable and is mentioned as fitting by Grosse (12,22) and by Chapman (61).

2.10.2. The density of thallium

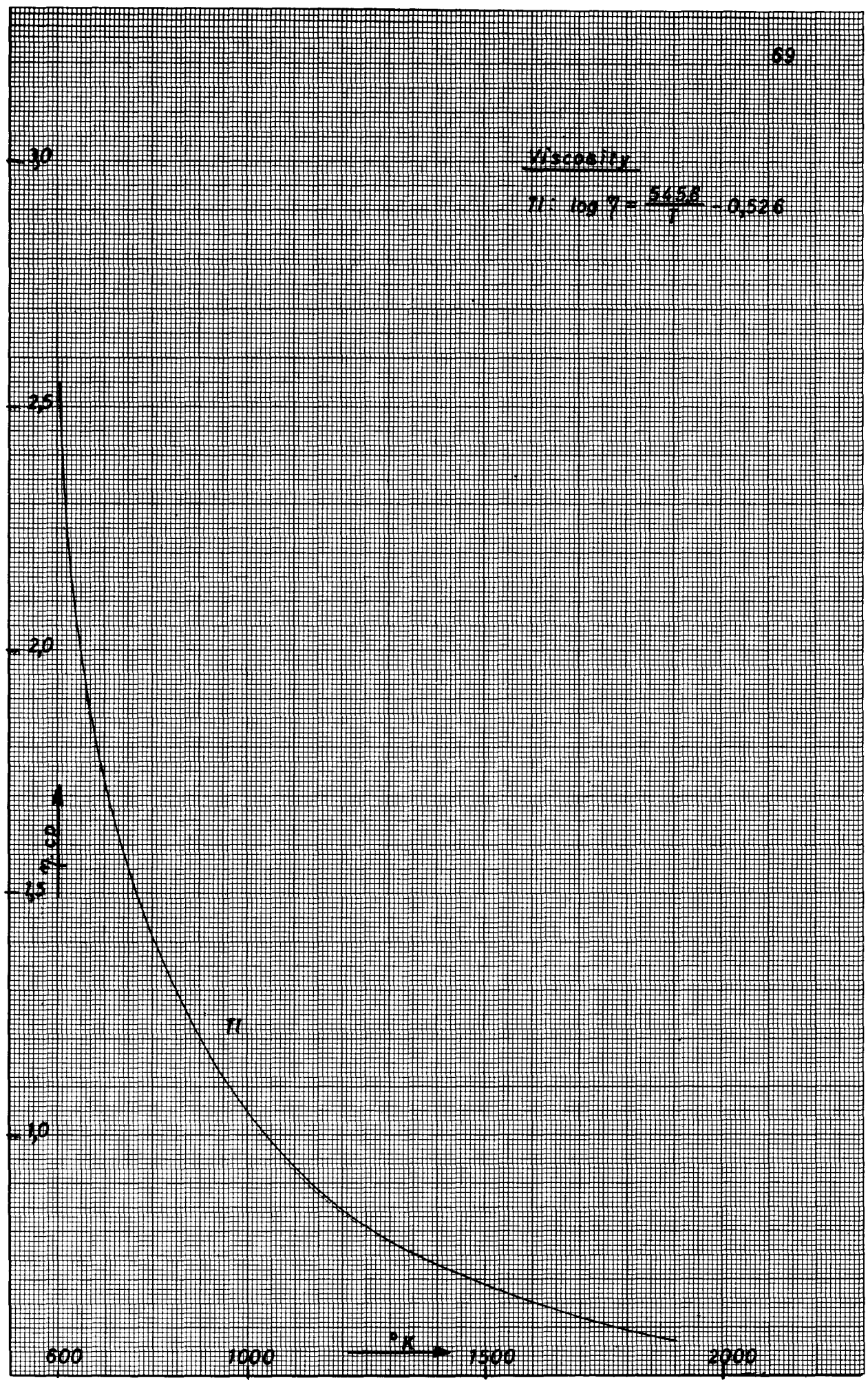
One cannot feel very happy with the two reported measurements because they correspond to the same formula though taken in another temperature range and with a different method.

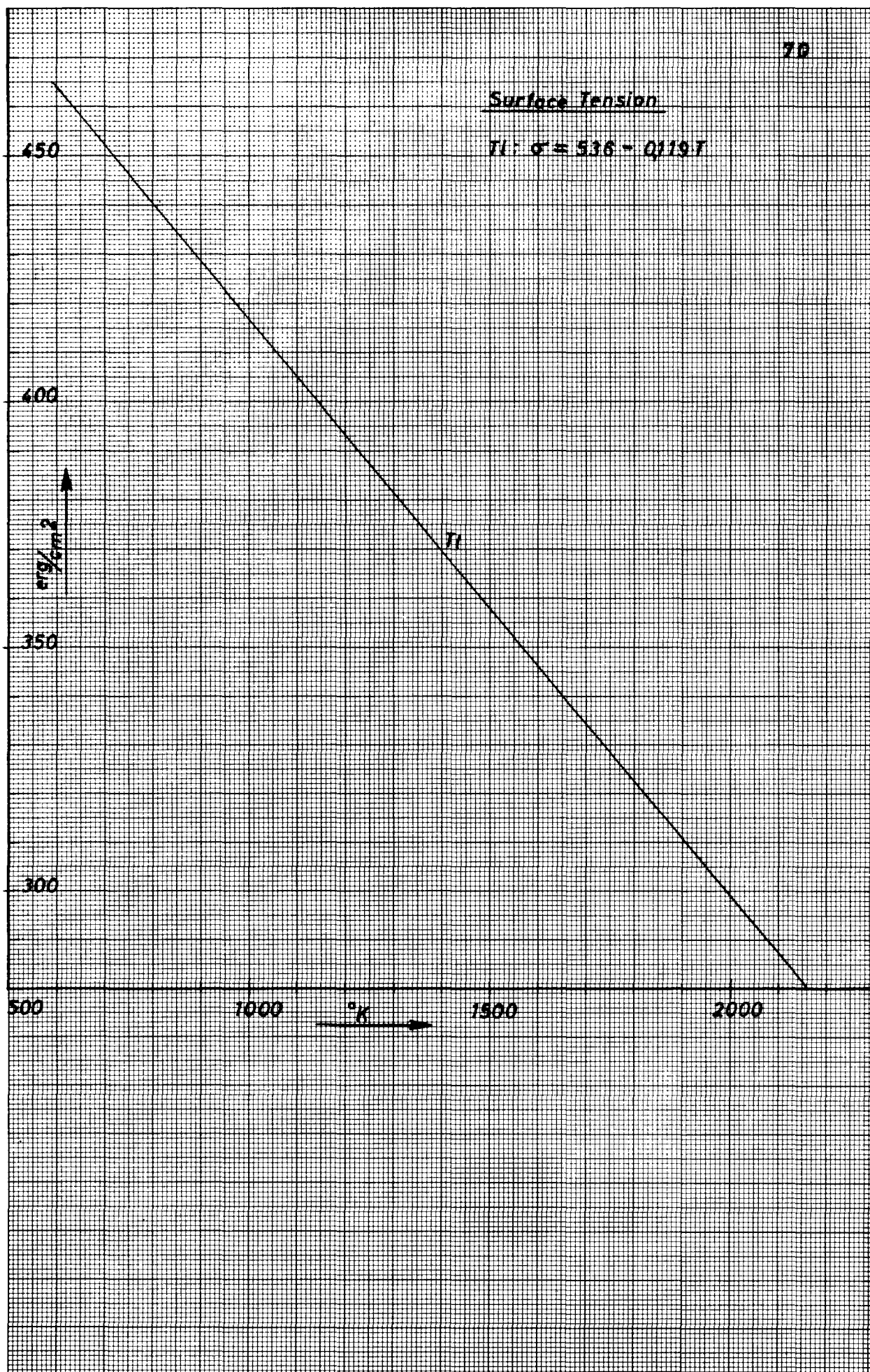
Author	Year	Method	Max. Temp.	Formula
(58)	1895	Dilatometric	326°C	$12,17-15,2 \cdot 10^{-4} T$
(42)	1954	Max Bubble P.	650°C	$12,16-15,21 \cdot 10^{-4} T$

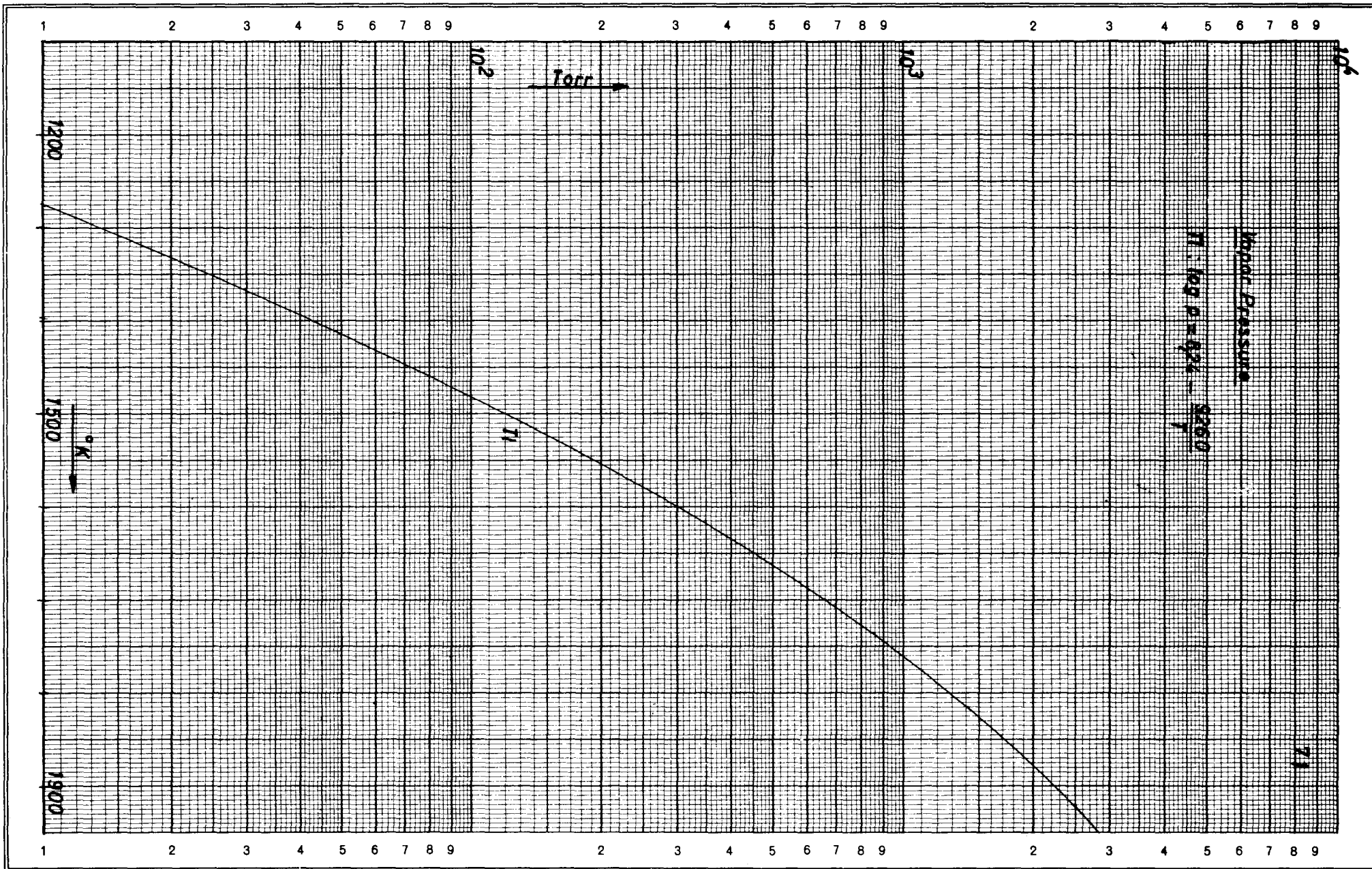


Viscosity

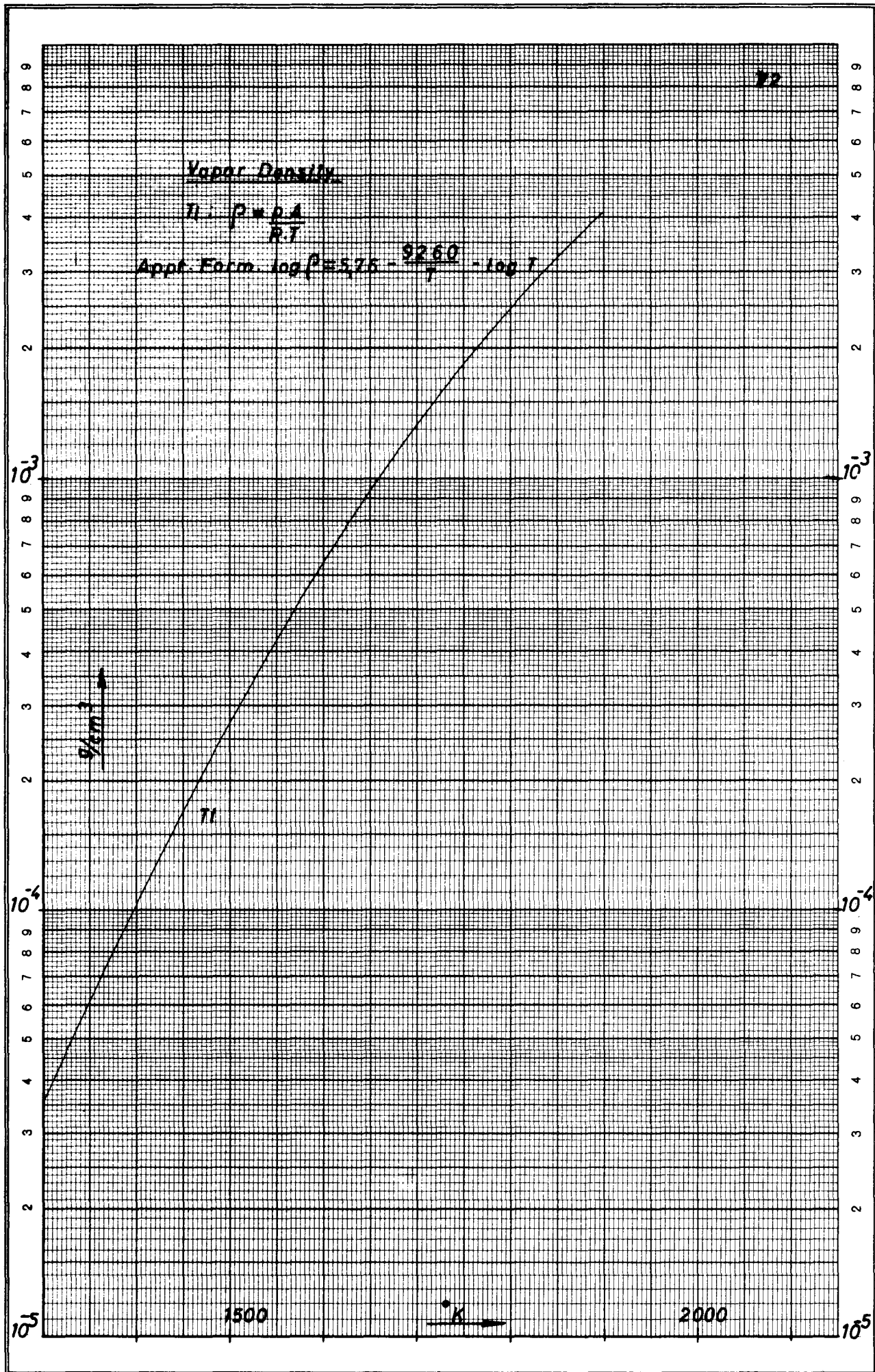
$$\eta = \log \eta = \frac{5456}{T} - 0,526$$







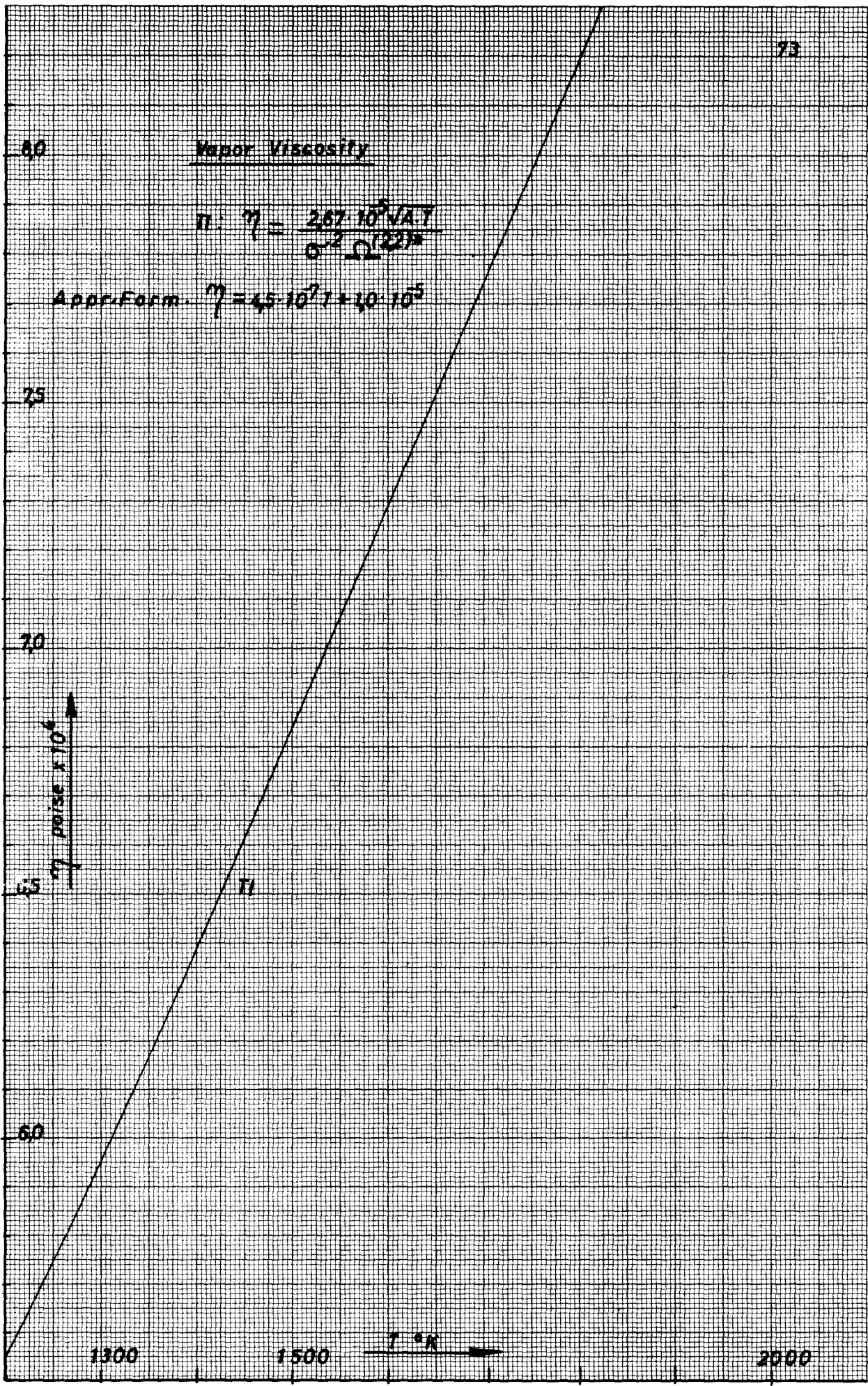
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Vapor Viscosity

$$\eta = \frac{267 \cdot 10^{-5} \sqrt{A T}}{\sigma^{0.2} \Omega (Z)^{0.8}}$$

Appr. Form. $\eta = 45 \cdot 10^{-7} T + 10 \cdot 10^{-5}$



2.11.	Lead	Value / Formula	References
	Atomic number	82	
	Atomic weight	207, 21	
	Atomic diameter	3, 20 Å	132
	Melting point	600°K	17
	Boiling point	2018°K	5
	Critical temp.	5400°K-4200°K	9, 157
	Heat of vapor.	45, 6 Cal/mol	5
	Density	11, 406-12, 82. 10 ⁻⁴ . T g/cm ³	26, 40, 42, 67, 80-82, 145, 146
	Viscosity	0, 498 exp 2019/RT cP	44, 45, 75, 87, 95, 146, 148, 149
	Surface tension	538-0, 114 T dyne/cm	26, 43, 76, 113, 150, 153, 154
	Vapor pressure	log P = 7, 83-9970/T torr	4, 5
	Vapor density	333. 10 ⁻³ P/T (P in torr)	
	approx. formula	5, 35-9970/T-log T	
	Vapor viscosity	3, 76. 10 ⁻⁵ √T/Ω ^{(2, 2)*} poise	125, 61
	approx. formula	4, 4. 10 ⁻⁷ . T + 1, 0. 10 ⁻⁵	

2.11.1. The viscosity of lead

Leaving out the measurements of Gering and Sauerwald (44, 1935), Gebhardt and Kostlin (32, 1957) and Fisher-Phillips (115, 1954), we find that all other measurements fall practically on one line in the log η to 1/T plot.

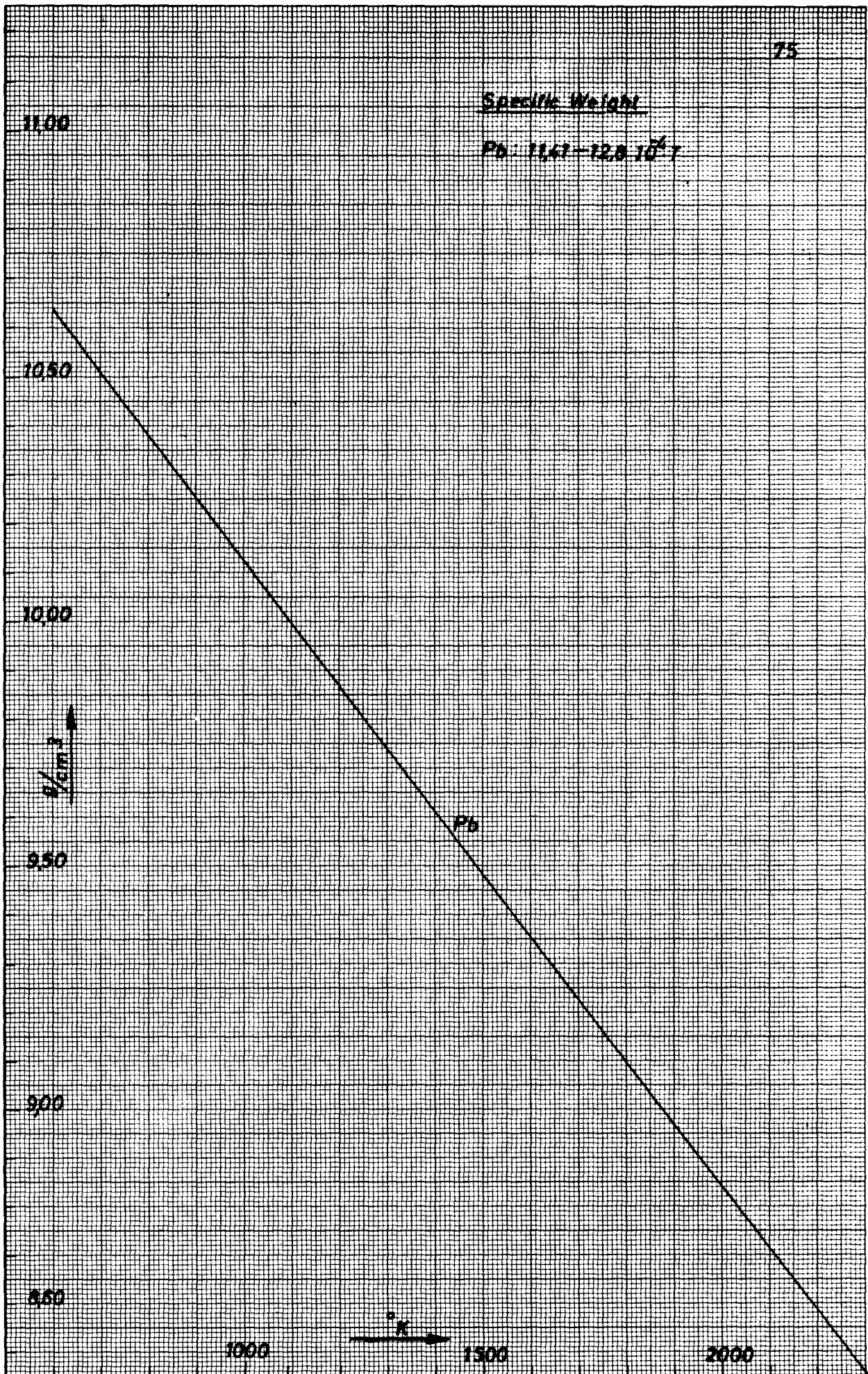
Most investigators measured up to a temperature of 750°C.

2.11.2. The density of lead

Author	Year	Method	Max. temp.	Formula
(145)	1914	Archimedean	850°C	11, 468-13, 315. 10 ⁻⁴ T
(26)	1921	U-tube	522°C	11, 544-13, 9. 10 ⁻⁴ T
(67)	1950	Max Bubble P.	1000°C	11, 287-11, 6. 10 ⁻⁴ T
(81)	1951	Archimedean	800°C	11, 43-12, 5. 10 ⁻⁴ T
(42)	1954	Max Bubble P.	700°C	11, 47-13, 2. 10 ⁻⁴ T
(80)	1960	Archimedean	800°C	11, 435-12, 53. 10 ⁻⁴ T
(40)	1961	Archimedean	1700°C	11, 469-13, 174. 10 ⁻⁴ T
(146)	1965	Pycnometer	450°C	11, 385-11, 7. 10 ⁻⁴ T

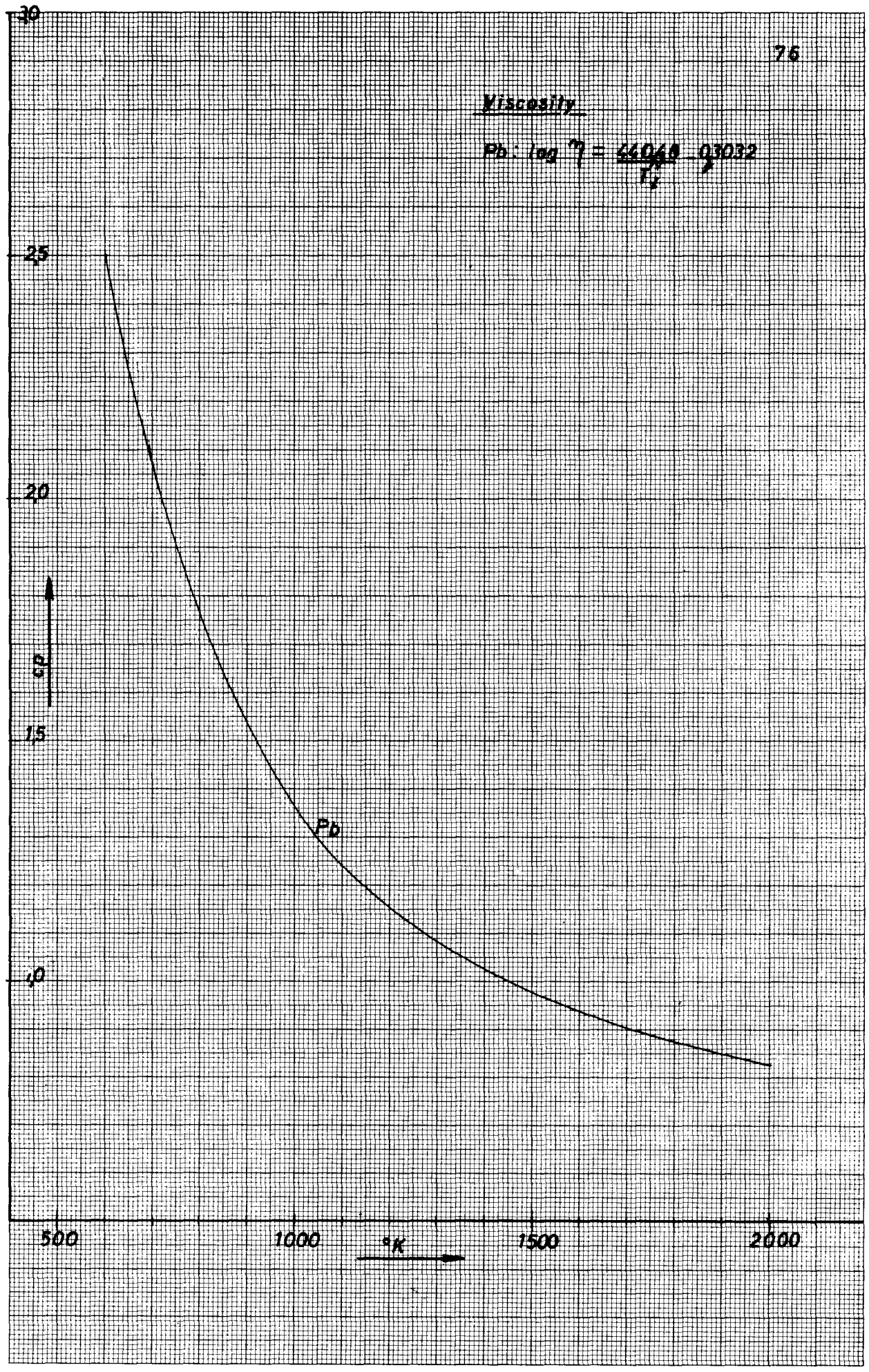
Specific Weight

Pb: $11.47 - 12.8 \cdot 10^{-4} T$



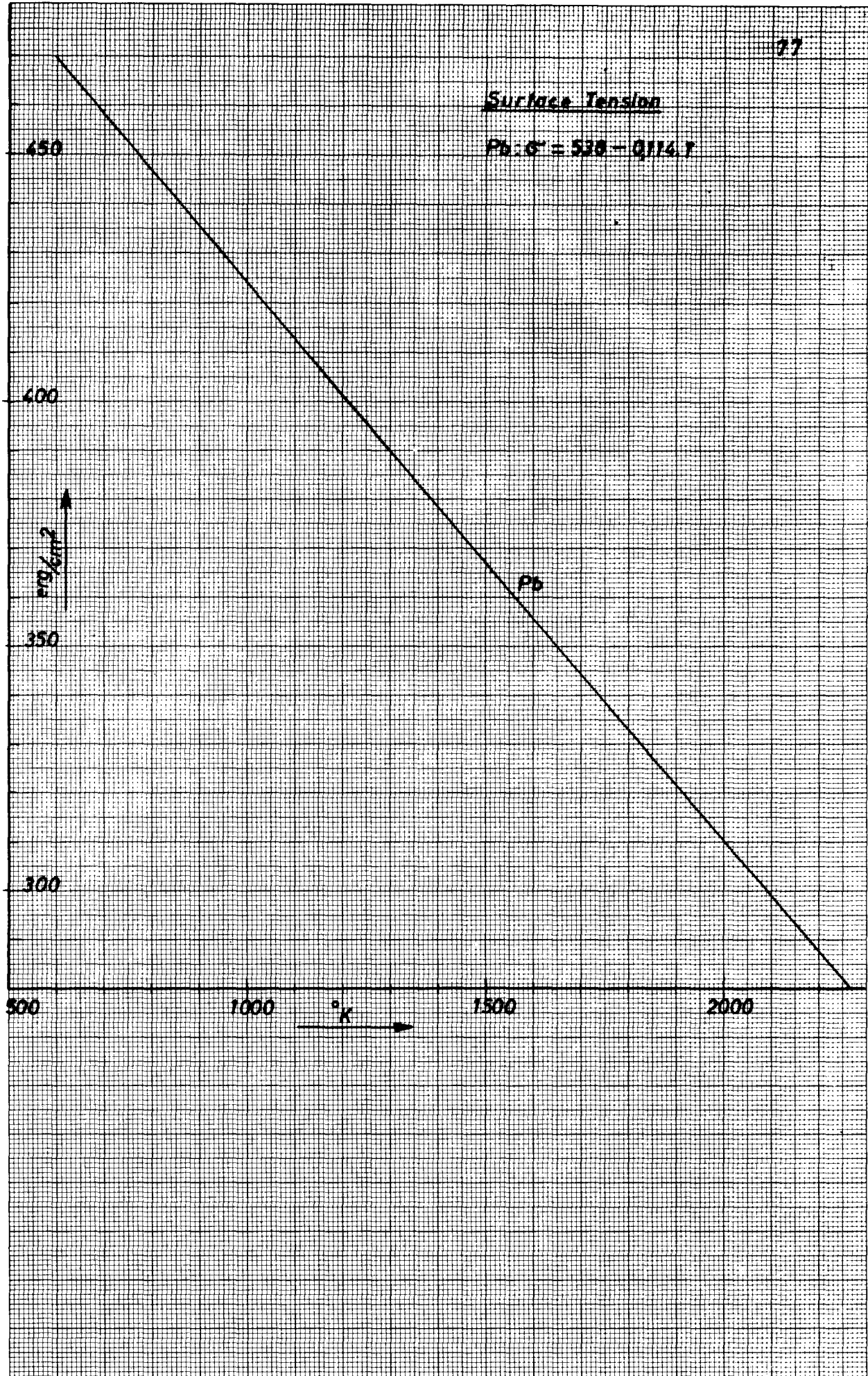
Viscosity

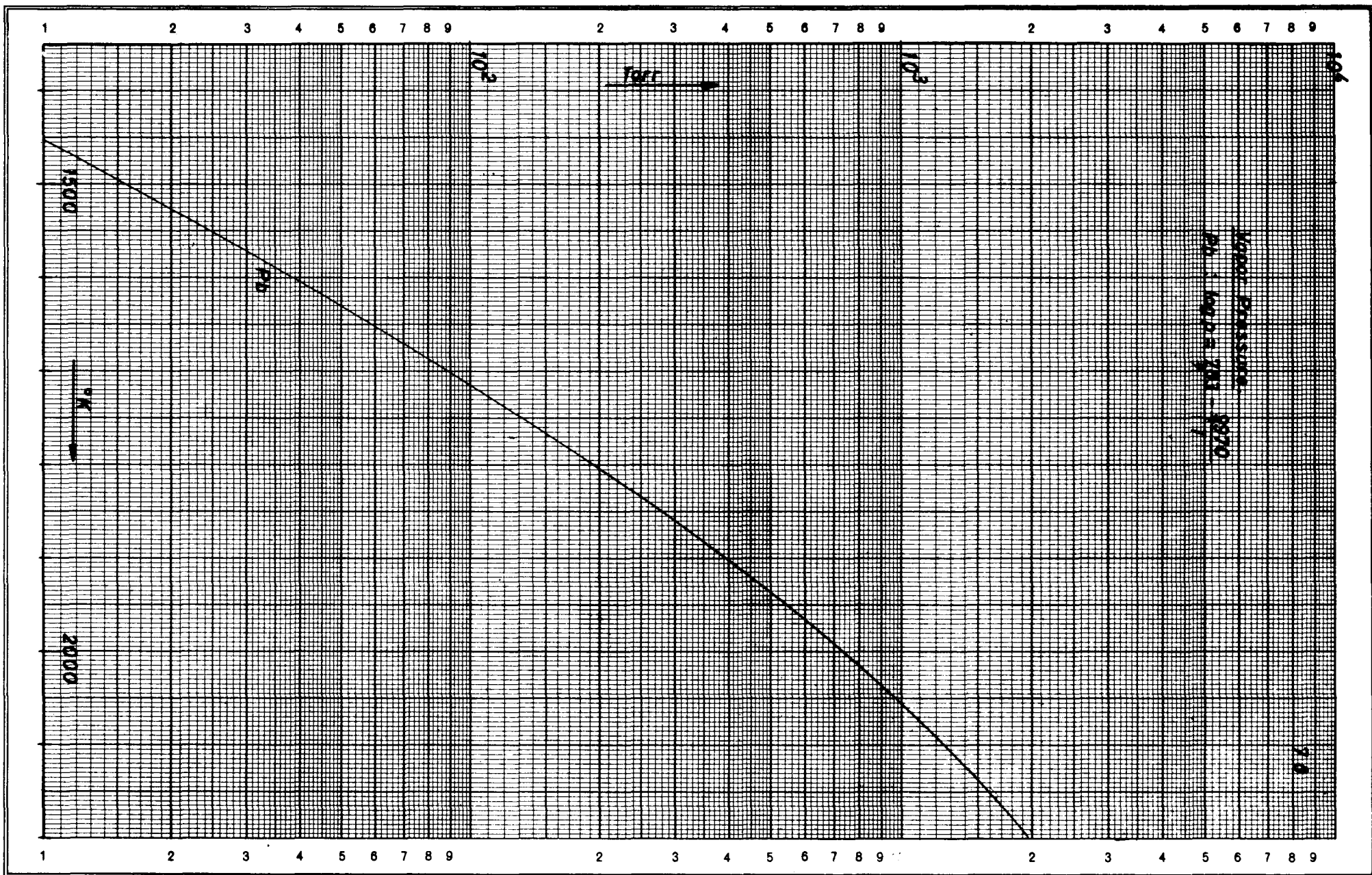
$$Pb: \log \eta = \frac{66048}{T} - 0.3032$$



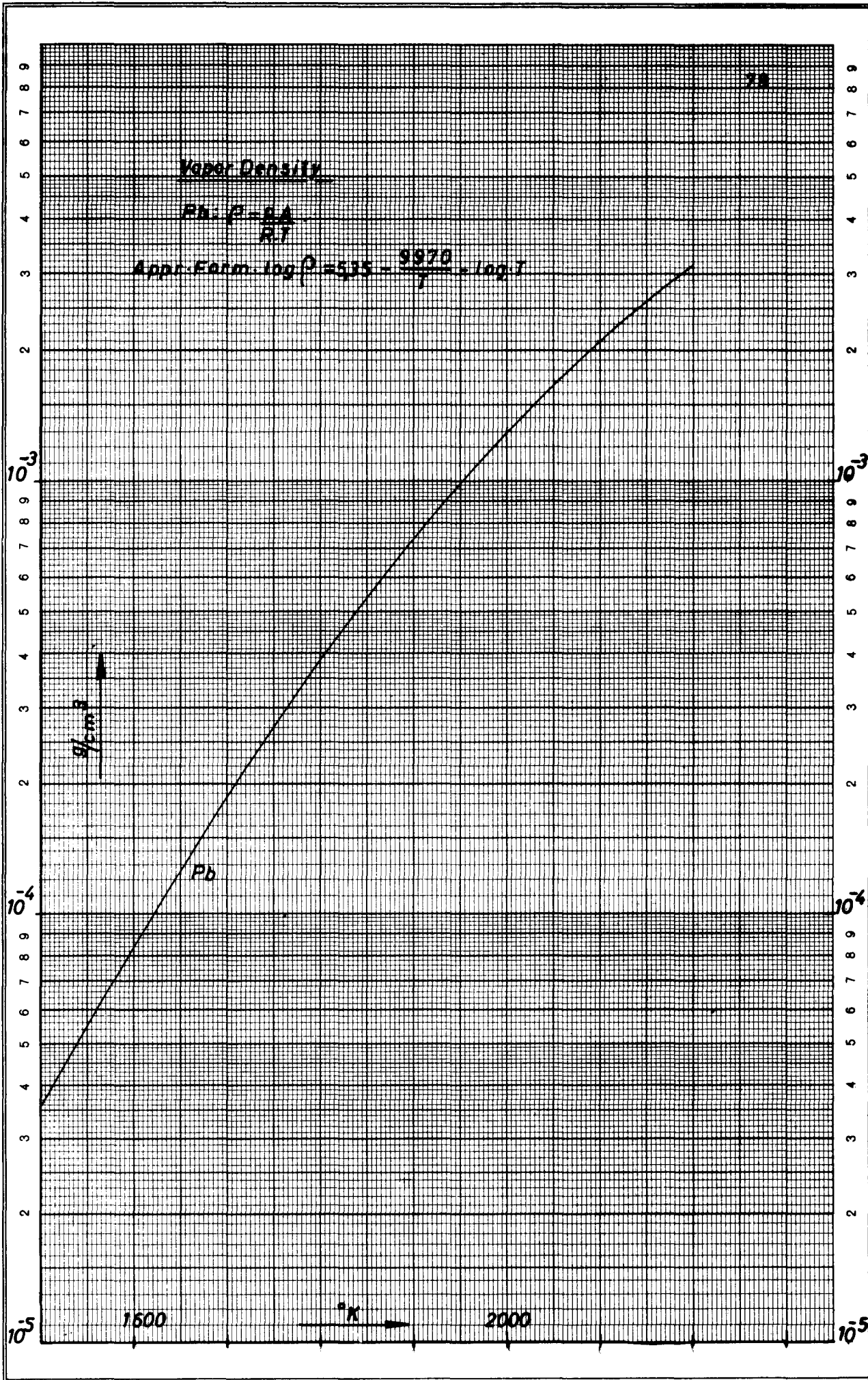
Surface Tension

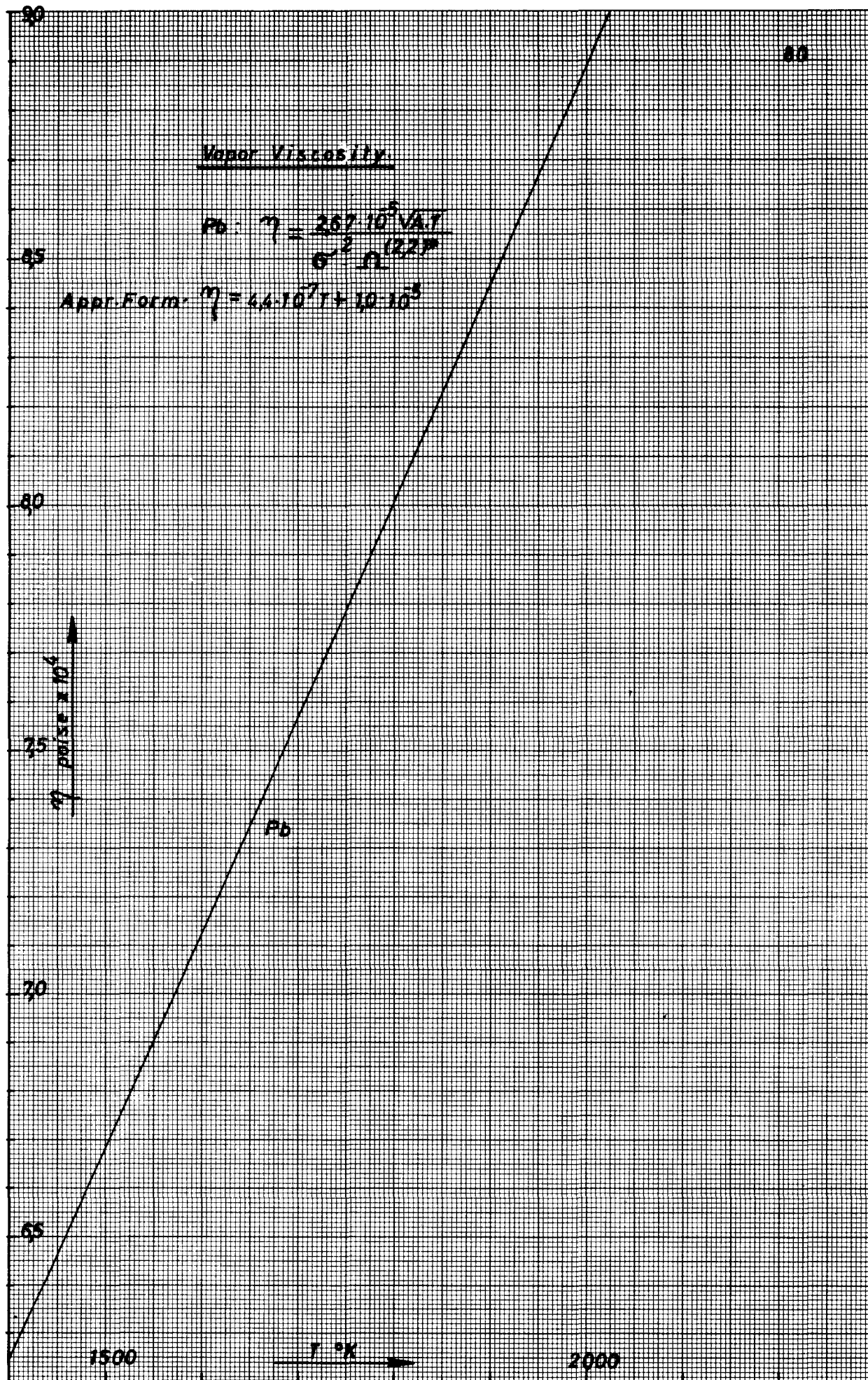
$Pb: \sigma = 530 - 0.114 T$





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2.12.

Bismuth	Value / Formula	References
Atomic number	83	
Atomic weight	209,00	
Atomic diameter	3,30 Å	132
Melting point	545°K	17
Boiling point	1838°K	5
Critical temp.	4345°K, 6500°K	9,157
Heat of vapor.	45,1 Cal/mol	5
Density	$10,710-12,2 \cdot 10^{-4} \cdot T \text{ g/cm}^3$	67-74, 31, 150
Viscosity	$0,332 \exp 1879/RT \text{ cP}$	30-32, 62, 75, 95, 148, 149
Surface tension	$428-0,088 T \text{ dyne/cm}$	26, 27, 29, 76, 106, 150, 155
Vapor pressure	$\log P = 8,25 - 9860/T \text{ torr}$	5
Vapor density	$3,36 \cdot 10^{-3} P/T \text{ (P in torr)}$	
approx. formula	$5,78-9860/T - \log T$	
Vapor viscosity	$3,54 \cdot 10^{-5} \sqrt{T/\Omega}^{(2,2)*} \text{ poise}$	125, 61
approx. formula	$4,1 \cdot 10^{-7} \cdot T + 1,0 \cdot 10^{-5}$	

2.12.1. The viscosity of bismuth

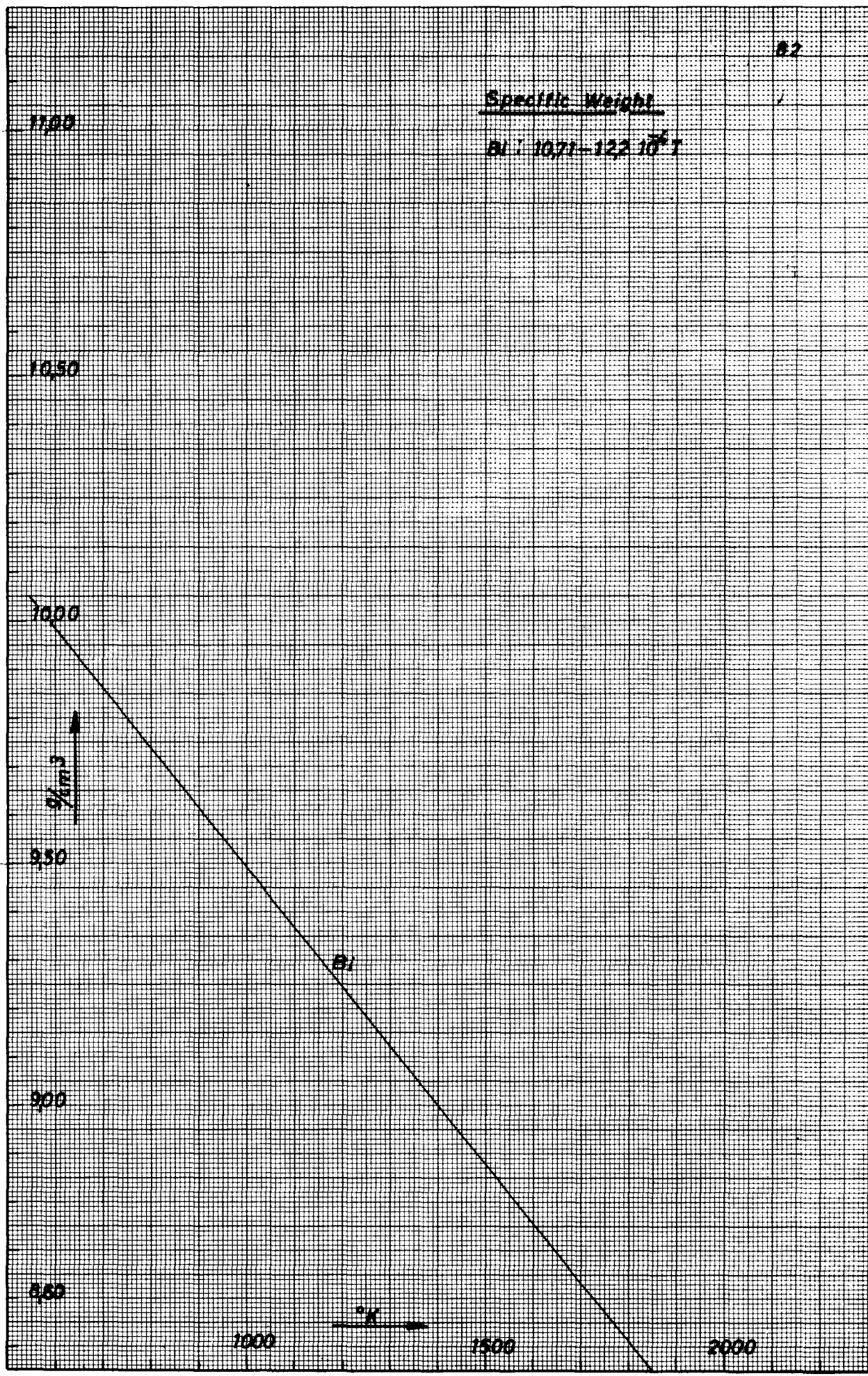
Only the measurements of Arpi (62, 1914) and Plüss (30, 1915) fall clearly away from the other series which agree satisfactorily.

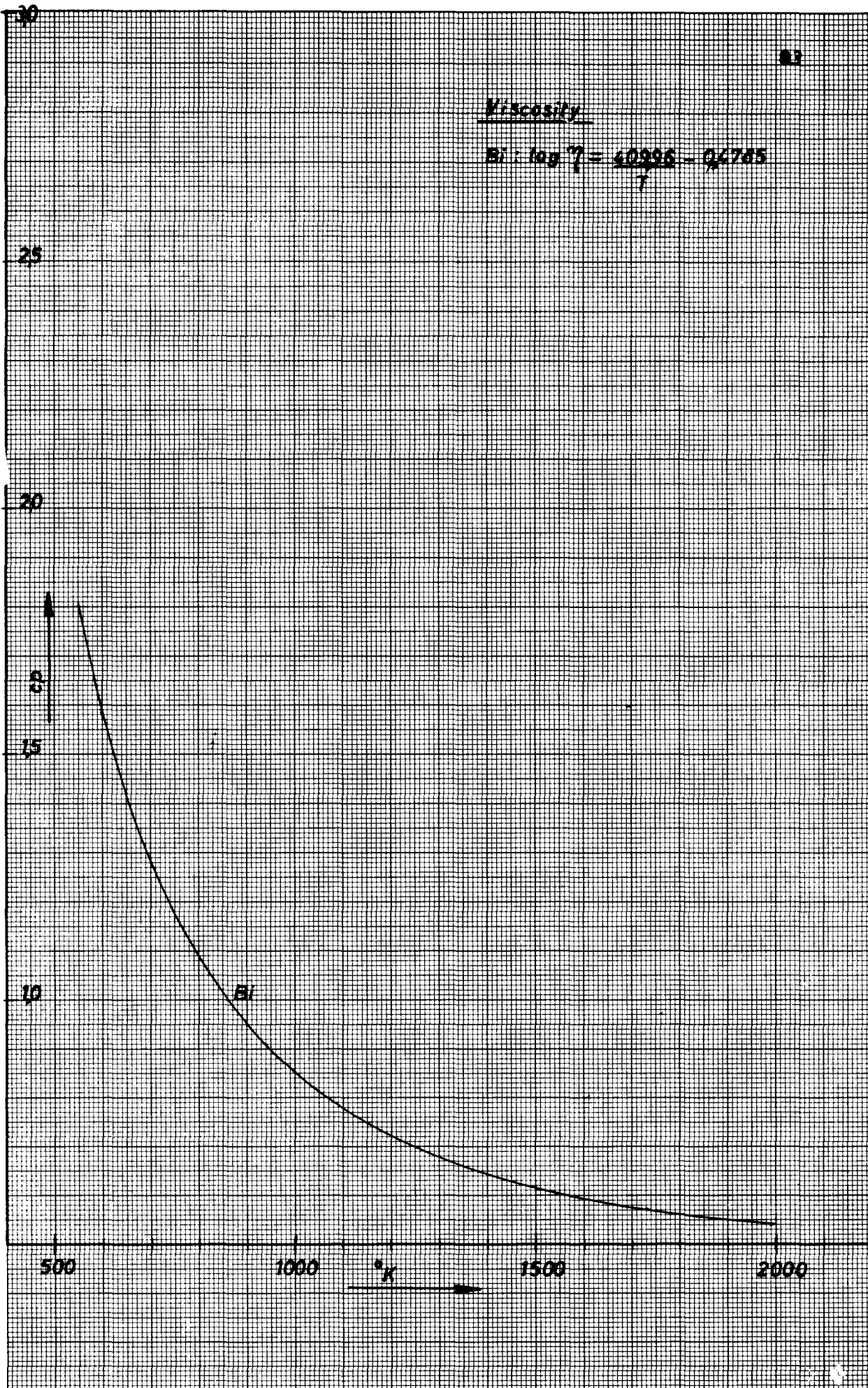
2.12.2. The density of bismuth

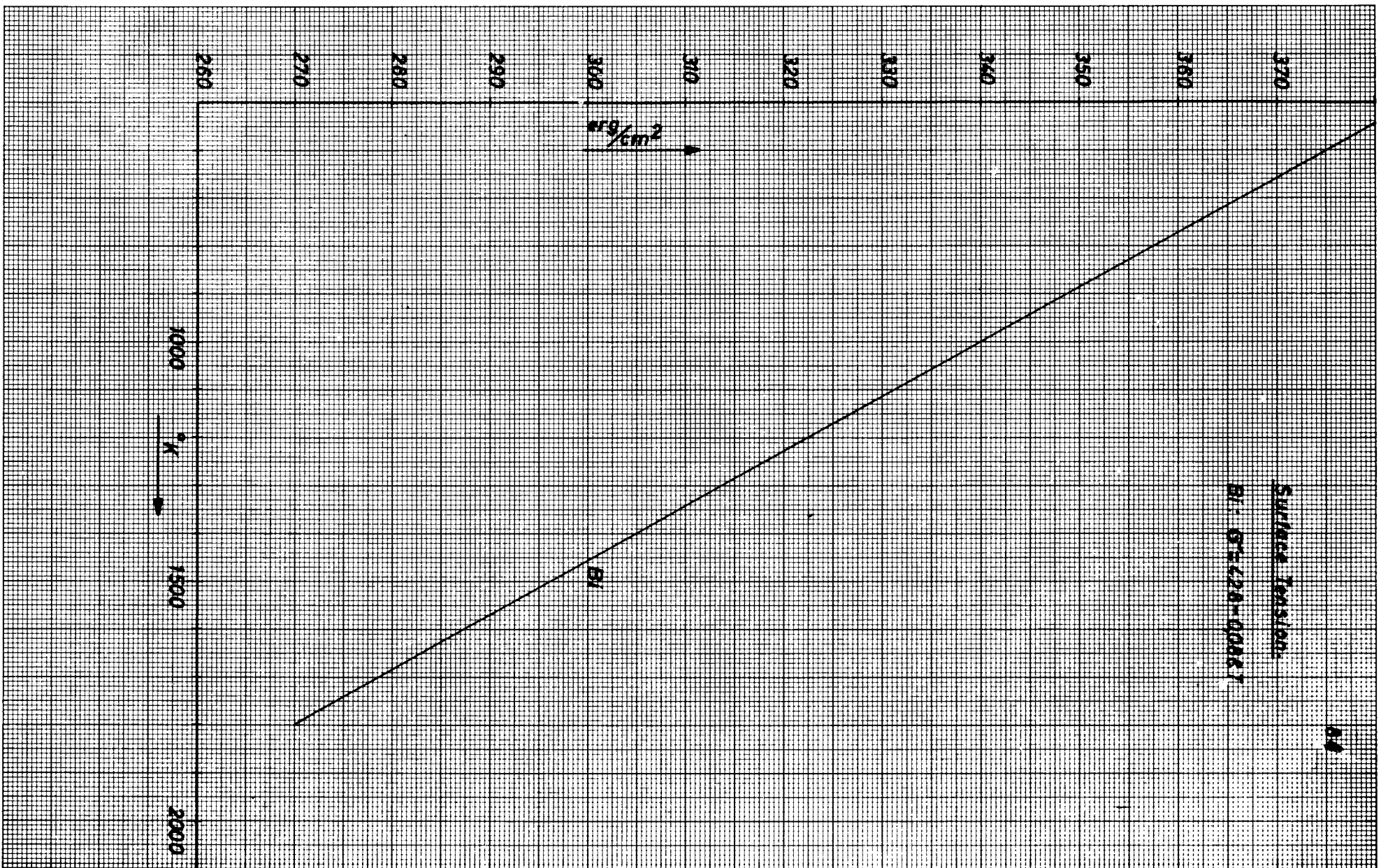
Author	Year	Method	Max. Temp.	Formula
(26) Bornemann	1921	U-tube	500°C	$10,75-12,5 \cdot 10^{-4} T$
see (67)	1922	Pycnometer	600°C	$10,674-11,95 \cdot 10^{-4} T$
(68)	1928	"	800°C	$10,698-11,8 \cdot 10^{-4} T$
(67)	1950	Max Bubble P.	1000°C	$10,81-13,2 \cdot 10^{-4} T$
(70)	1959	Archimedean	700°C	$10,687-12,14 \cdot 10^{-4} T$
(150)	1959	Max Bubble P.	1000°C	$10,93-14,0 \cdot 10^{-4} T$
(69)	1960	Pycnometer	700°C	$10,698-11,8 \cdot 10^{-4} T$
(71)	1960	U-tube	550°C	$10,67-12,25 \cdot 10^{-4} T$
(72)	1962	Archimedean	800°C	$10,7315-12,36 \cdot 10^{-4} T$
(73)	1962	Max Bubble P.	1000°C	$10,706-11,73 \cdot 10^{-4} T$
(74)	1963	Archimedean	1500°C	$10,665-11,82 \cdot 10^{-4} T$

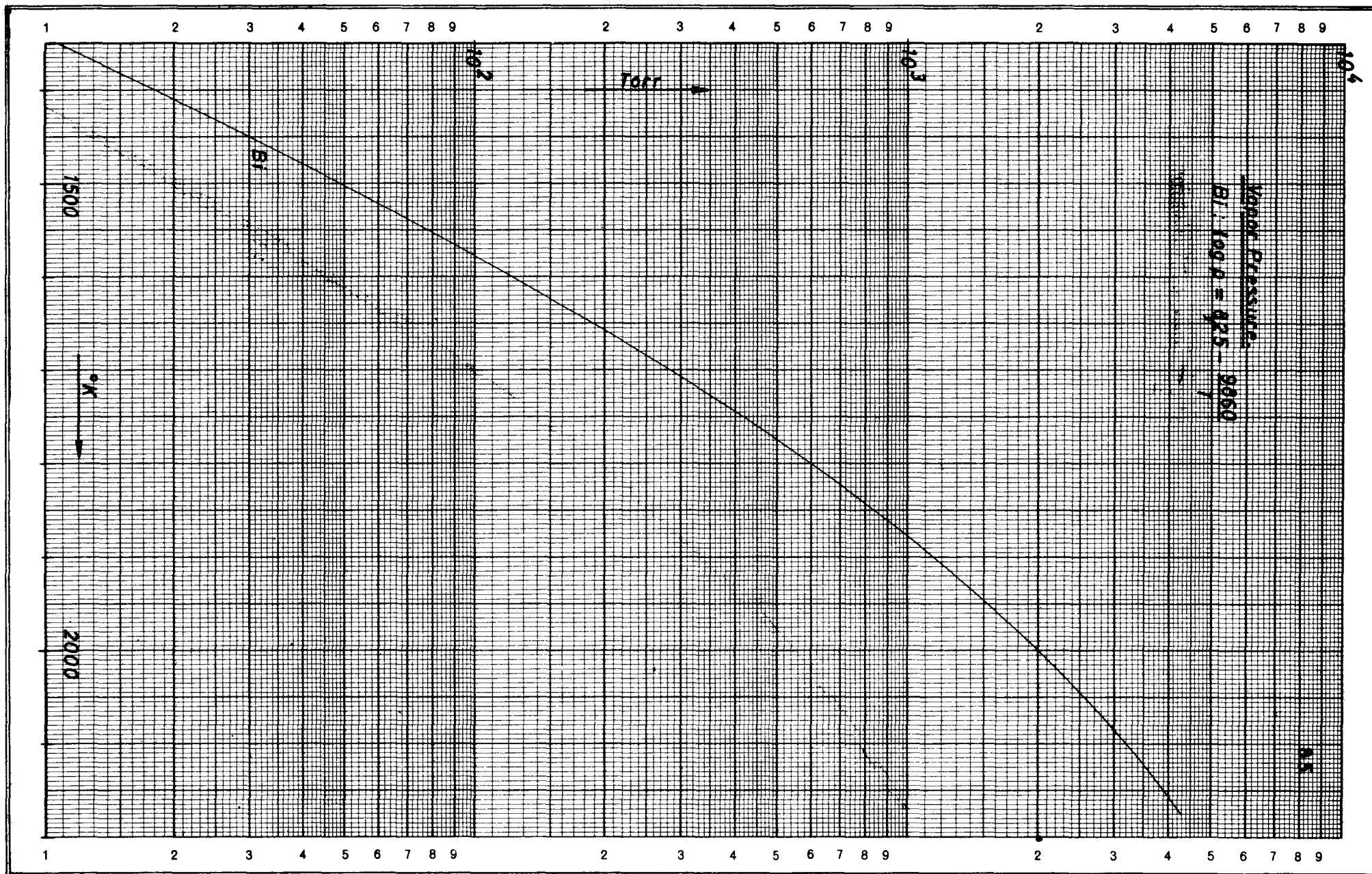
Specific Weight

BI: 10,71-122 10⁻¹



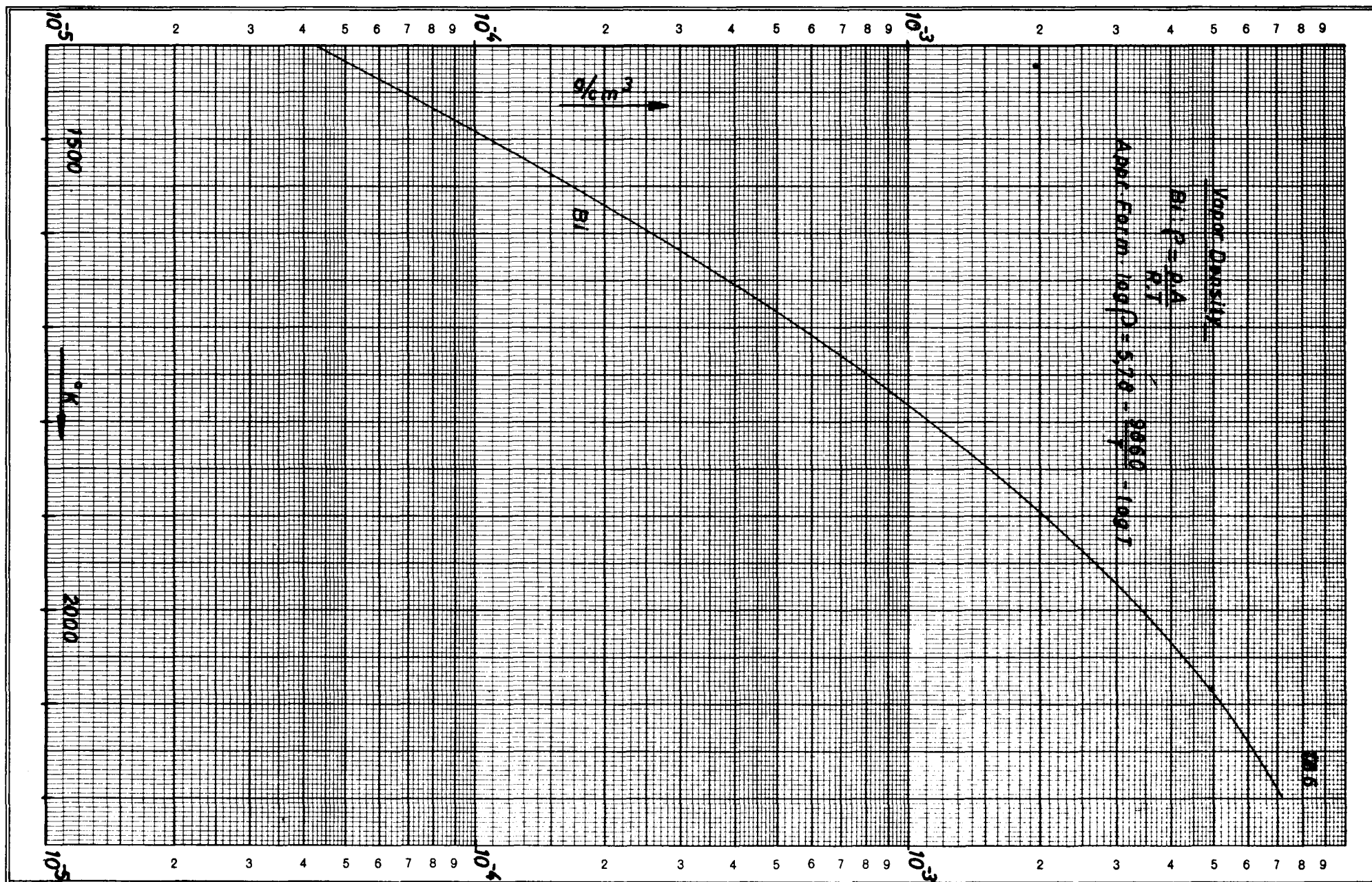




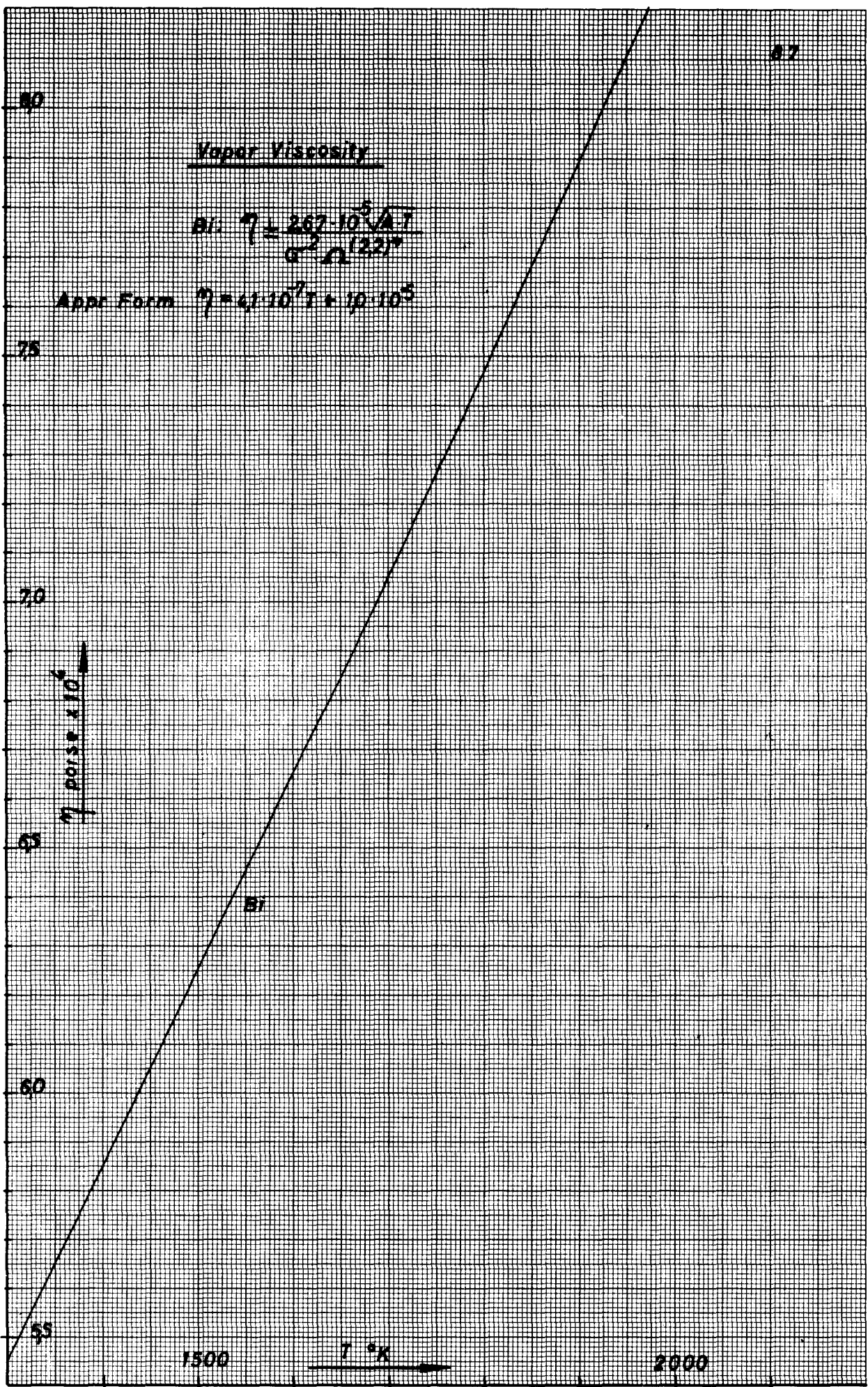


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3. GENERAL CORRELATIONS USEFUL FOR EXTRAPOLATION PURPOSES

Liquid metal data are richly scattered in literature but general relationships are difficult to obtain because of the fact that the study of pure liquid metals is still at an early stage.

Reviewing the theory and the results of experiments one can say that it is possible to account for the straight-line relationship of $\log \eta$ over $1/T$ (Chapter 6).

As to the liquid range (Chapter 4) we think that the method of A. V. Grosse to determine the critical point is rather good but is still too far from being exact. It appears also that entropy of vaporisation is not a quantity to be adapted to the theory of the corresponding states as pointed out by Dillon (93).

General formulas for the specific weight have been brought forward by Strauss and McGonigal. Strauss's formula is too empirical and must be considered as preliminary also because of his use of the critical temperatures from Gates and Thodos. McGonigals use of the critical temperatures of Grosse is more fortunate but his final form is too complicate and also inexact (Chapter 5).

As to the surface tension we find a lot of theories concerning both the zero Kelvin and the melting point temperature value and concerning the temperature coefficient but none of them is consistent (Chapter 7).

Coming to the vapor phase, the straight-line relationship of $\log P$ to $1/T$ in a restricted temperature region (e. g. near to the boiling point) is well confirmed (3, 17). The vapor density is easily calculated from ideal gas laws, and corresponds to actual values up to $0,8 T_c$ (9). The vapor viscosity is the least known property (Chapter 8).

4. THE LIQUID RANGE

For organic liquids relationships have been proposed for the critical temperature T_c by Guldberg and Meissner-Redding. After a critical re-examination of the experimental data Gates and Thodos (7) proposed the following formula:

$$T_c = 1,4732 T_b^{1,0313} \quad (1)$$

The authors were convinced that this formula can be used also for liquid metals. However, measurements from Birch in Bridgemans laboratory indicated that the critical temperature for Hg is 1733°K (8) whereas Gates and Thodos find 1140°K , which represents a major discrepancy.

Birch proceeds in the following way. A cylindrical open quartz container was filled with Hg and put in a furnace assembly. The external electrical circuit connected in series with the mercury-column consisted simply of a milliammeter, a resistance of 150Ω , and adry cell (a bridge is impracticable near T_c). When boiling occurred the current fell to zero. In this way the vapor pressure curve was traced. At 1640 atm the fall of the current was no longer abrupt: as the temperature increased to secure boiling, the current did no more fall to zero but decreased first slowly then rapidly, but still permitting current readings to be taken. Additional weight to the interpretation of this pressure to be the critical is provided by comparison of the current-temperature curves at different pressures. At 2040 atm one needs 70°C for a current drop by a factor of 100; at 1750, 40 degrees and at 1640 atm only 15°C . So $P_c = 1640$ atm at $T_c = 1733^{\circ}\text{K}$ for Hg.

Starting from these values (and new existing data for Cs and Rb) Grosse (9) has made a new approach for the theory of the liquid range of metals. As even such thermally stable molecules as CO and N_2 are atomized above 6000°K so that chemical substances cease to exist above that temperature the exception of liquid metals (T_c for W is 20.000°K) is striking. When the forces between the molecules are the weak van der Waals forces, then equation (1) is valid. This does not seem to be true for the metals; let us consider mercury more closely.

Of special assistance for the evaluation of the liquid range is the empirical law of Cailletet and Matthias or the law of the rectilinear diameter which states that half the sum of the densities of a liquid and its saturated vapor D , decreases linearly with temperature. Especially for mercury several data correspond to obtain a uniform picture. The vapor pressure is a straight line on a log plot versus $1/T$ to very high pressure-values. Up to 1200°C - that means $0,8 T_c$ - and 665 atm there is no significant deviation between the ideal gas densities calculated from this pressure and the actual

values. Moreover, all densities were measured up to 60°C from the critical temperature (1460°C) and the law of Cailletet was fulfilled. An upper limit for the critical temperature can now be set in the following way. The two legs of density (liquid and vapor, half of whose sum is D) have two envelopes, the ideal gas density and the "straight liquid density" minus ideal gas density. (These two envelopes meet for Hg at 2000°K).

We will assume now that the rectilinear diameter law holds for other metals too. In this way one can find readily, by extrapolation of the vapor pressure data, the upper limit for the critical temperature. In order to have a second and more exact estimate it is best to choose properties which equal zero at the critical temperature and apply then the principle of the corresponding states. Such properties are the latent heat of vaporization, the entropy of vaporization and surface tension. Grosse chose the entropy of vaporization (because the zero degree Kelvin value is infinite, so equal for all metals) and the principle of the corresponding states requires now that the molar entropies of vaporization should be equal at corresponding temperatures. In fig. 1 is shown the entropy curve of mercury and we could assume that this curve is valid for all metals. We then can use the curve for the calculation of the critical temperatures of other metals knowing the boiling points only. Nevertheless, data e. g. for Na were fitted over the range of 445°K to 1667°K and the closest fit was for $T_c = 2800^{\circ}\text{K}$.

That the procedure, however, is still not quite exact, is evident from fig. 2 which shows that the entropy of vaporization is not equal for all metals at the same reduced temperature (93).

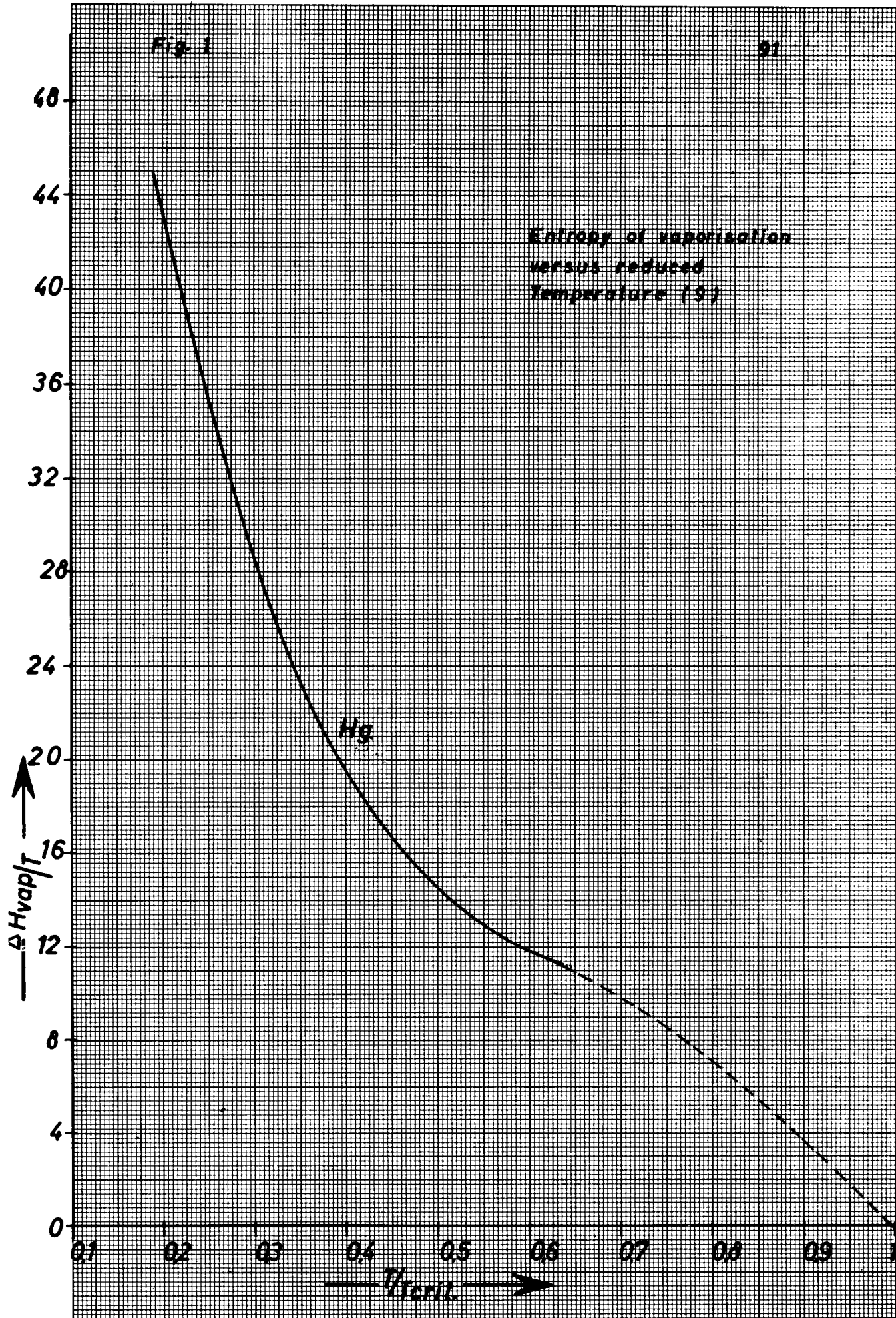
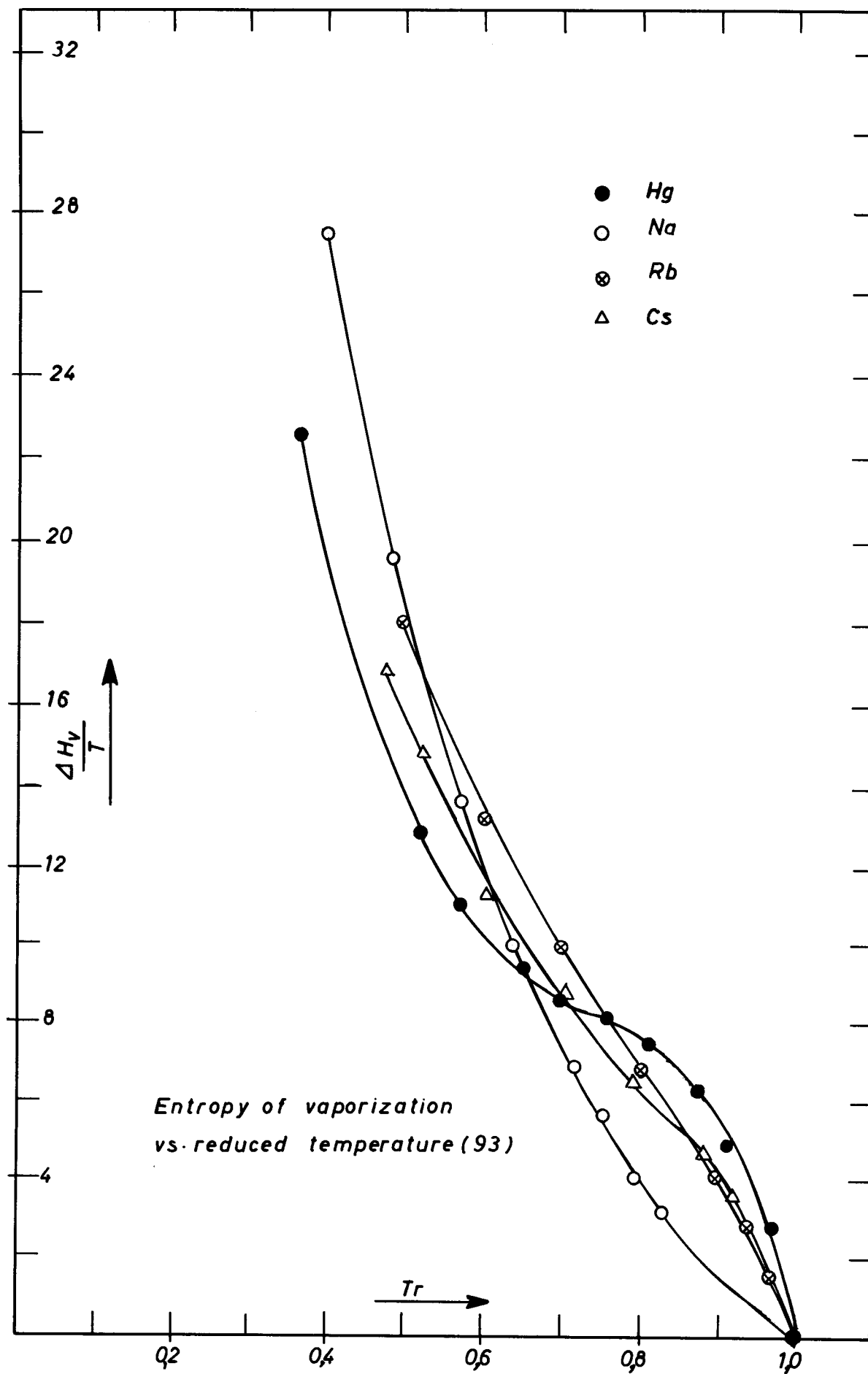


Fig 2



5. SPECIFIC WEIGHT

From the rectilinear diameter Grosse (9) derived:

$$\rho = \rho_c \frac{\rho_m / \rho_c - 2}{T_c - T_m} (T_c - T) + 2\rho_c \quad (2)$$

ρ_m, T_m values at melting point

ρ_c, T_c values at critical temperature.

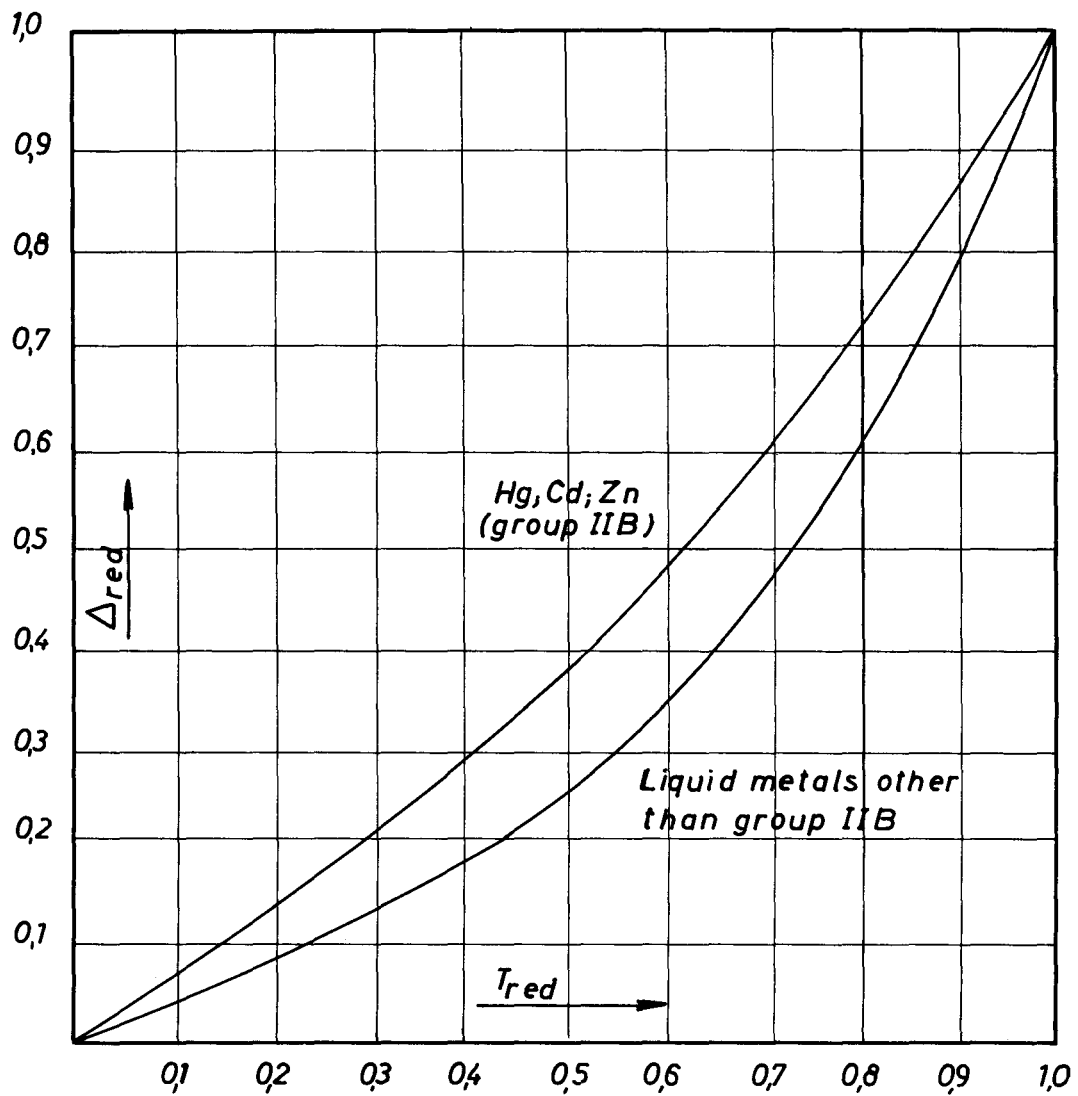
In this formula ρ_c has to be roughly estimated or it has to be derived from exact extrapolation. A universal approach to obtain an unknown value of ρ_c has been made by McGonigal (10). He uses the T_c and ρ_c from common metals as obtained from Grosses' plots and traces the values of reduced properties Δ / Δ_c versus T/T_c where Δ is:

$$\Delta = \frac{d(D) T}{d(T) D}$$

These values are plotted in fig. 3 and it appears that all metals fall on one curve. So if one knows one value of specific weight for a certain metal i. e. $2D$ at a temperature T one finds readily the corresponding D_c from fig. 3, putting $\frac{d D}{d T} = \text{constant}$. Thus one derives readily a universal density formula starting from a single measured value, or from some values which are close together. This method depends, however, too specifically on the (badly known) critical temperatures to be of efficient aid.

The generalisation of Strauss (65) is still more empirical. Furthermore, when we needed some extrapolation for the earth-alkaline metals it turned out to be fairly inexact when confronted afterwards with the measured values.

Fig. 3



Relation between Reduced Density and Temperature for liquids (10)

6. VISCOSITY

It is a wellknown experimental fact that a plot of $\log \eta$ versus $1/T$ is a straight line for all known liquids. It was first stated by de Guzman in 1913 and is commonly put in the form:

$$\eta = A \exp H/RT \quad (3)$$

where H is the energy for viscous activation. The variant form which contains a volume factor makes little or no improvement over the simple equation. That it works equally well is due to the slow variation of the volume with the temperature.

An effort for the derivation of the experimental law (3) was made by Andrade (15). He puts forward a vibration theory and states that viscosity is caused by the sharing of momentum at extreme libration. The liquid viscosity decreases with temperature because the temperature agitation interferes with the momentum transfer. The number of cases favorable for transfer will decrease as temperature rises. The temporary union which is postulated for interchange resembles that involved in association but is of far shorter duration, he calls it "instantaneous combination". He supposes that there is a local electrostatic field which tends to place the molecules in a condition favorable for interchange of momentum, this condition being disturbed by the heat agitation. So for transfer of momentum the atoms considered must possess a minimum mutual potential energy ϵ . If the vibration frequency ν is taken to be constant, the variation of viscosity with temperature will be governed by the fraction of molecules possessing this energy ϵ at extreme libration. So according to Boltzmann

$$\frac{\eta}{\eta_m} = \frac{\exp \epsilon/k T}{\exp \epsilon/k T_m} \quad (4)$$

and from equation (7) equation (3) is readily derived.

Andrade succeeded also in deriving a formula for the viscosity at the melting point. He states that the essential difference between the liquid and the solid state is not the magnitude of the intermolecular force, but the amplitude of the vibration. The assumption is that there is for a liquid atom a vibration about a slowly displaced equilibrium position with a frequency which is the same as that of the solid state. Making a calculation for lead, one can assume with Lindeman $\nu = 2.10^{12} \text{ sec}^{-1}$, $T/2 = 2.5.10^{-13} \text{ sec}$.

The time τ necessary for the diffusion of an atom over an atomic distance δ is calculated from the diffusion formula of Einstein:

$$2 D = \frac{\overline{x^2}}{\tau} \quad (5)$$

We substitute $2 D = 5,1 \cdot 10^{-5} \text{ cm}^2/\text{sec}$ $x = 3,1 \cdot 10^{-8} \text{ cm}$ and find $\tau = 1,9 \cdot 10^{-11} \text{ sec}$, so we see that $2 \tau/T = 76$ and one must conclude that gas kinetic transfer must be far less than transfer by instantaneous combination. If then viscosity is due to sharing of momentum at extreme libration, then at every extreme libration the contact of the atom of one layer with those of an adjacent layer is of sufficient duration to provoke a common velocity of translation. When there are ν vibrations per second, $2 \nu/3$ passages in 2 directions, the transfer of momentum per second per square cm is:

$$\frac{2 \nu}{3} m \frac{d v}{d z} 2 \delta \frac{1}{\delta^2} = \eta \frac{d v}{d z} \quad (6)$$

From Lindemann we have:

$$\nu = 2,8 \cdot 10^{-12} \frac{T_m^{1/2}}{M^{1/2} v^{1/3}} \quad (7)$$

And further we have $\delta^2 = n^{-2/3}$; $m = M/N$; $n = N/v$

So,

$$\eta_m = 5,7 \cdot 10^{-4} \frac{(M T_m)^{1/2}}{v^{2/3}} \quad (8)$$

If we combine finally equation (8) and (3), we find

$$\eta = \frac{5,7 \cdot 10^{-4} (M T_m)^{1/2}}{v^{2/3} \exp(H/R T_m)} \exp(H/RT) = A \exp(H/RT) \quad (9)$$

a general relationship which should receive more justification from experiment. For a discussion on the constant A see e. g. (148).

Ewell (14) relates according to Eyring the H to the energy of vaporization. They put forward a reaction rate theory of viscosity. The general features

of the theory apply to flow when gas type of viscous drag (diffusion) is negligibly small. They consider flow as a unimolecular chemical reaction in which the elementary process is the passing of a molecule (or "unit of flow") from one equilibrium position to another over a potential barrier. H represents the activation energy for this process. This activation energy will be some fraction of the vaporization energy and in fact for normal liquids and non-associated molecules $E_{\text{vap}} = 3 H$. The most striking fact regarding the metals is that $E_{\text{vap}} = 8$ to $25 H$. This can be understood if the unit of flow is taken to be the small ion:

$$E_{\text{vap}} = 3 H \frac{\text{atomic volume}}{\text{ion volume}} \quad (10)$$

A test of this relation for nine liquids showed it to be true within the experimental error of the quantities involved. Nevertheless the relation between the energy of viscous activation and the energy of vaporization is more complicated and is not very adapted for extrapolation uses.

Grosse (12, 22) and Strauss (13) plot the H of the metals over the melting points. They find a relative simple relationship, low melting metals have low energies of activation, the highest values are to be expected for the refractory metals. According to Grosse the formula for this curve is:

$$H = 0,431 T_m^{1,348} \quad (11)$$

H in cal/gr at and η in poise. Grosse's relationship is shown in fig. 4.

A method for estimating the viscosity up to the critical point is given by Grosse, using the hyperbolic diameter, see (129).

The evaluation of the viscosity of liquid metals by Chapman (61) seems to be rather good, nevertheless the correlation between his \mathcal{E}/K values and Grosse's T_c are very bad.

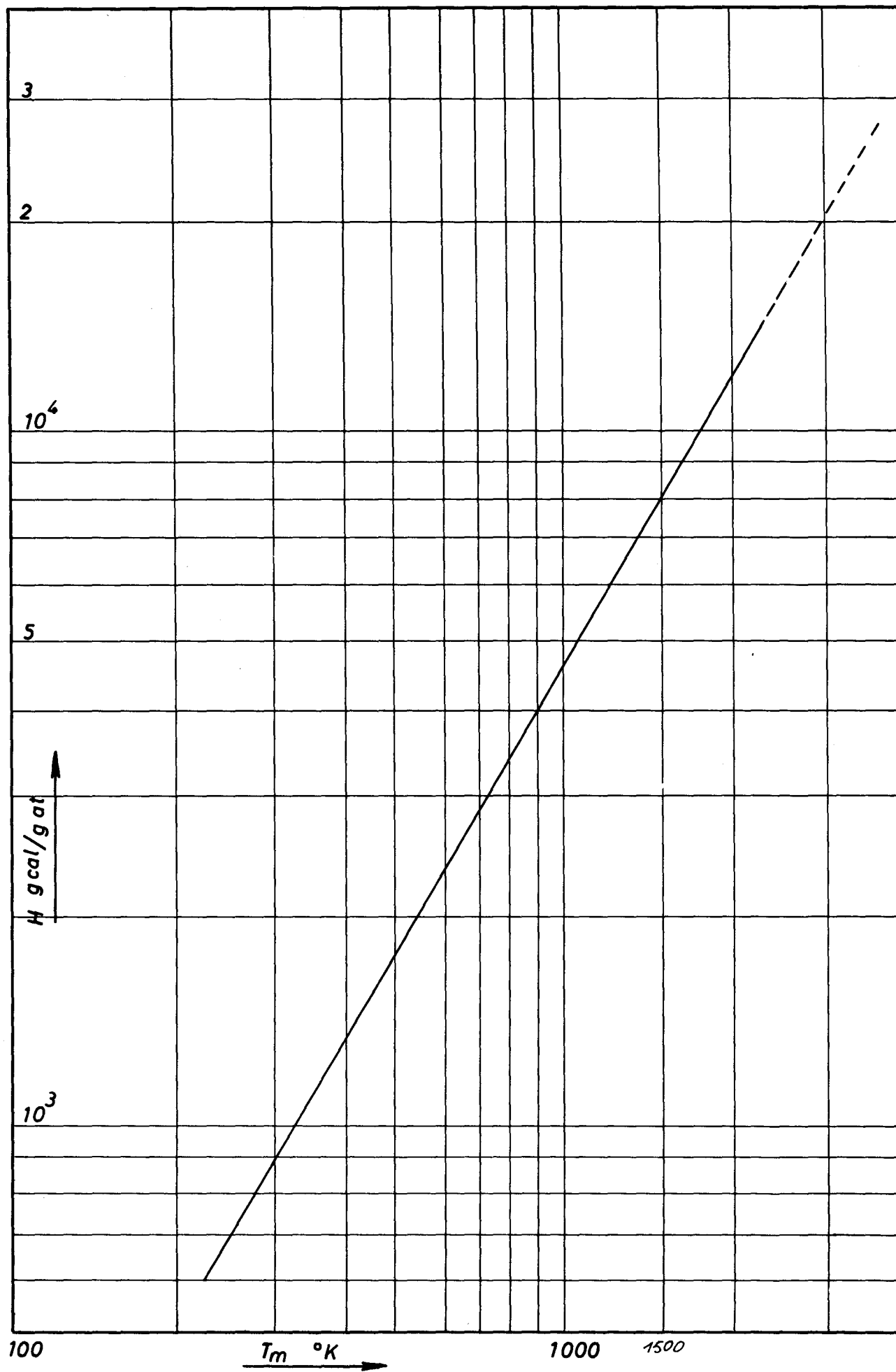


Fig. 4 Empirical relationship between H and T_m (22).

7. SURFACE TENSION

Eötvös derived the following law for molar free surface energy γ :

$$\gamma = \sigma v^{2/3} = k (T_c - T) \quad (12)$$

This can be written also:

$$\sigma = \sigma_0 \left(1 - \frac{T}{T_c}\right) \quad (13)$$

where σ_0 is the surface tension extrapolated to absolute zero.

Van der Waals used as a closer approximation:

$$\sigma = \sigma_0 \left(1 - \frac{T}{T_c}\right)^n \quad (14)$$

Grosse (11) using his critical temperatures from (9) found that the same law does apply to liquid metals but that the constant k is different namely $0,64 \text{ erg/mol}^{2/3} \text{ K}$. He extrapolated first the van der Waals σ_0 and found various n , e. g. $0,25$ for Cd and $1,28$ for Na. This is caused probably by an error effect, where all measurements are made within some 5% of the liquid range. Nevertheless he found only slightly different σ_0 values if he traced the σ as straight lines to zero values at T_c which means putting $n = 1$. Regardless of what the final accurate temperature function of σ will be (118) he chose for the present purposes the simple straight line relationship to obtain σ_0 . In order to obtain

$$\gamma_0 = \sigma_0 v^{2/3} \quad (15)$$

he uses with small error the atomic volume of the solid at room temperature.

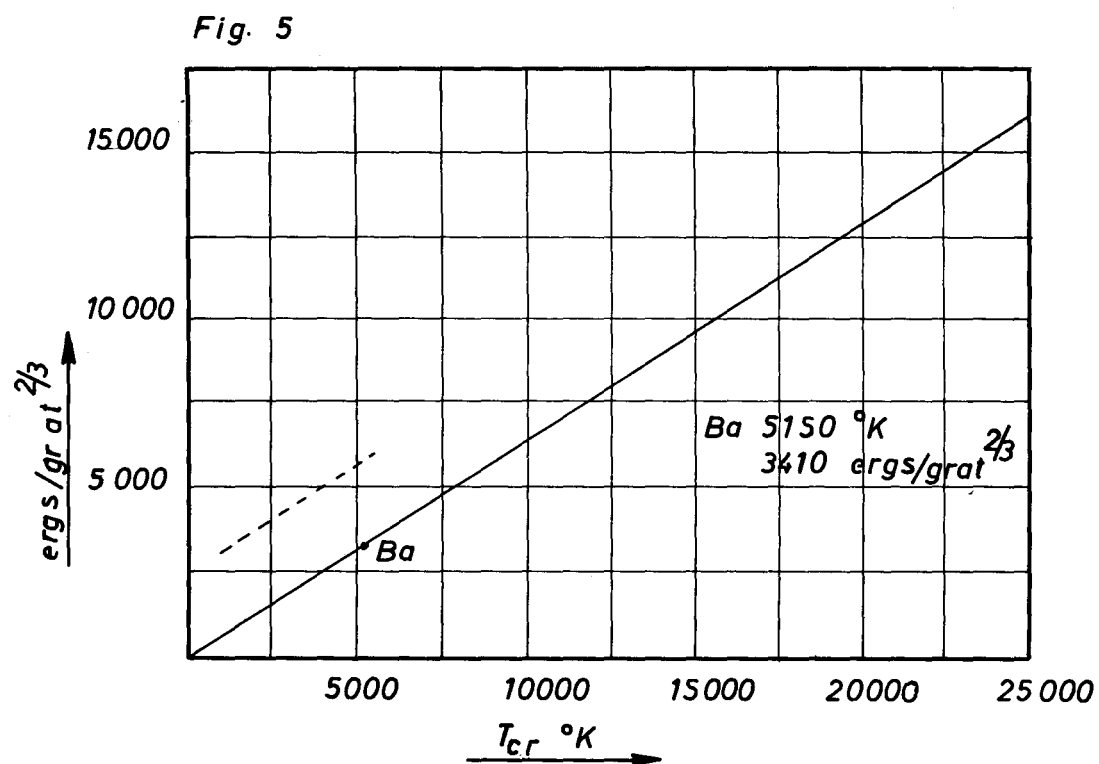
Now he can proceed to make a plot of free atomic surface energy versus the critical temperature. With the exception of Hg, Cd and Zn all the metals fall essentially on a straight line as shown in fig. 5. The general relationship permits to estimate the σ of any metal at any temperature. The slope $d\sigma/dT$ can be calculated from

$$\frac{d\sigma}{dT} = - \frac{\sigma_0}{T_c} \quad (16)$$

According to this method we calculate for Barium $\sigma = 312 - 0,0606 T$ but we measure $\sigma = 356 - 0,078 T$.

Our own measurements (6, 26) prompted Bohdansky thus to doubt about this simple supposition of the surface tension becoming zero at the critical temperature, following a regression line. In (118) he puts forward a new, i. e. thermodynamical theory about the temperature coefficient of surface tension in the regression line part of the curve at low temperatures.

The number of surface tension theories since the summary of J. W. Taylor (18, 28) has been completed by an electronic theory of Zadumkin (119, 19) a physico-chemical theory of Glauberman (120) and Spitkovsky (121); further by a correlation to work-functions by Kunin (122); to the entropy of evaporation per unit volume by Strauss (66, 123) and to the above mentioned relation with critical temperature by Grosse (9, 11).



Atomic surface energy γ and T_{cr} (11)

8. VAPOR PRESSURE, VAPOR DENSITY, VAPOR VISCOSITY

From theoretical considerations relative to a system consisting of a single substance in two phases (see e. g. 3 and 17) it is clear that in a first approximation $\log P$ is a straight line over $1/T$:

$$\frac{d \log P}{d 1/T} = - \frac{\Delta H_v}{2,303 R} \quad (17)$$

Also it is evident (see Grosse 9) that in a wide range the experimental vapor density is equal to the ideal gas density with a fair precision (fig. 6, 7).

Difficulties arise as to the value of the vapor viscosity. Many formulas have been put forward (Weatherford 130, Dunning 55, Grosse 129), which originate from normal gas considerations and are difficultly applied to metal vapors. Our procedure was to insert in the formula of Hirschfelder (125) the characteristic temperature of Chapman and to compare the results with the measured values of mercury (126, 127) and Caesium (128). We found these results very satisfactory and as they diverged evidently from other approaches, we adopted this method for the calculation of all other vapor viscosities (fig. 8, 9).

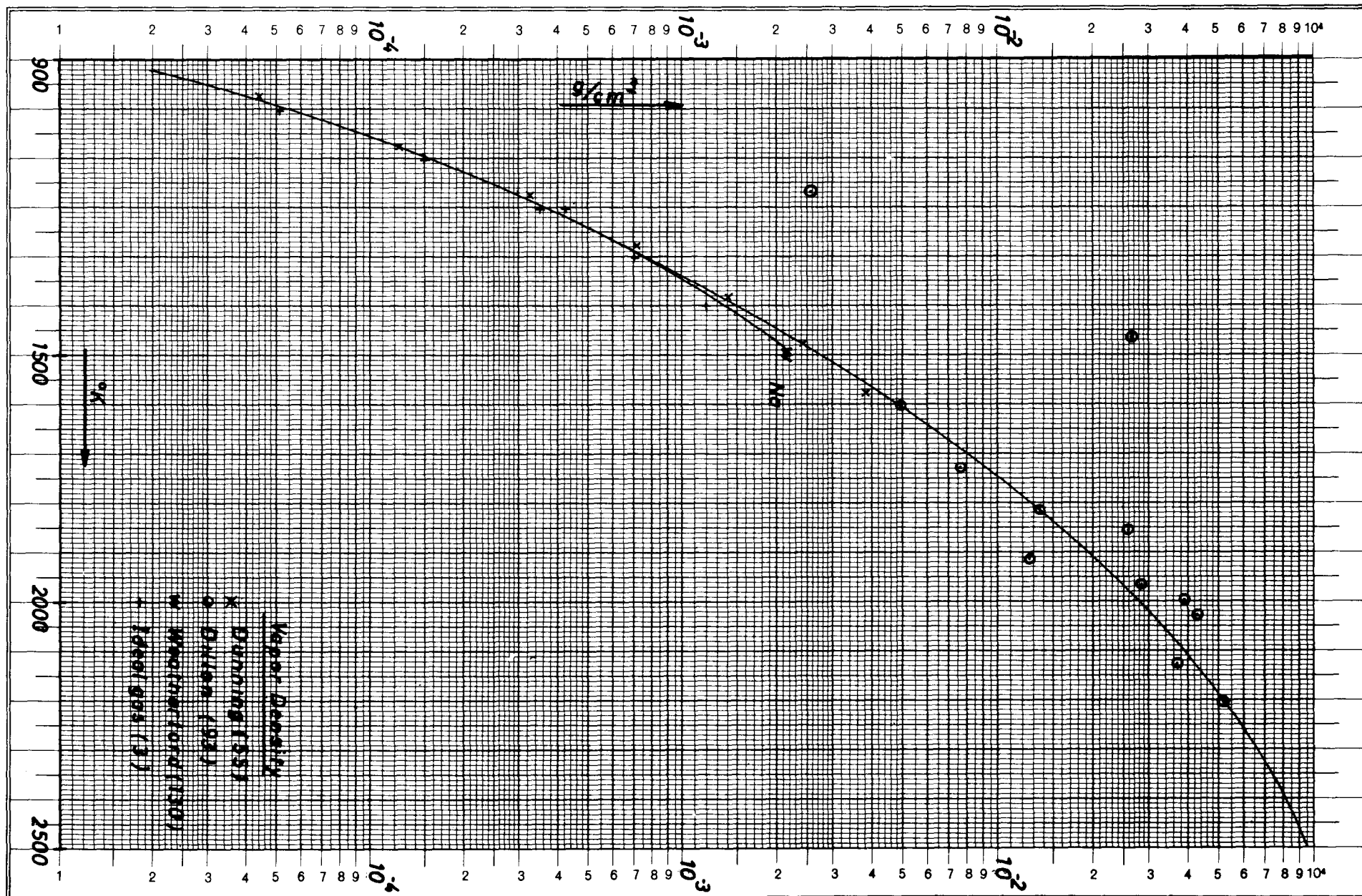
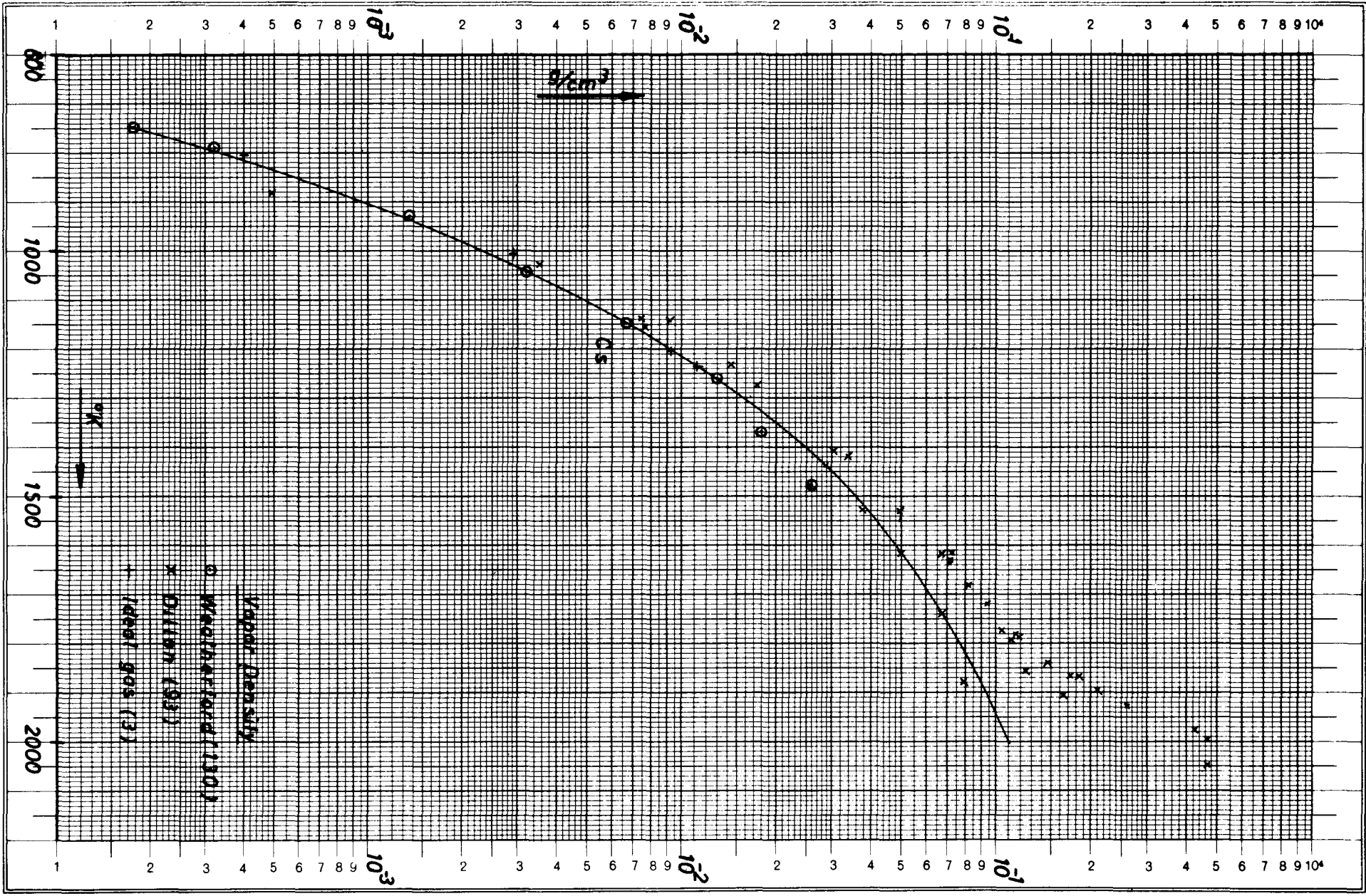


Fig. 6

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Fig. 7



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Fig. 8

Vapor Viscosity Hg

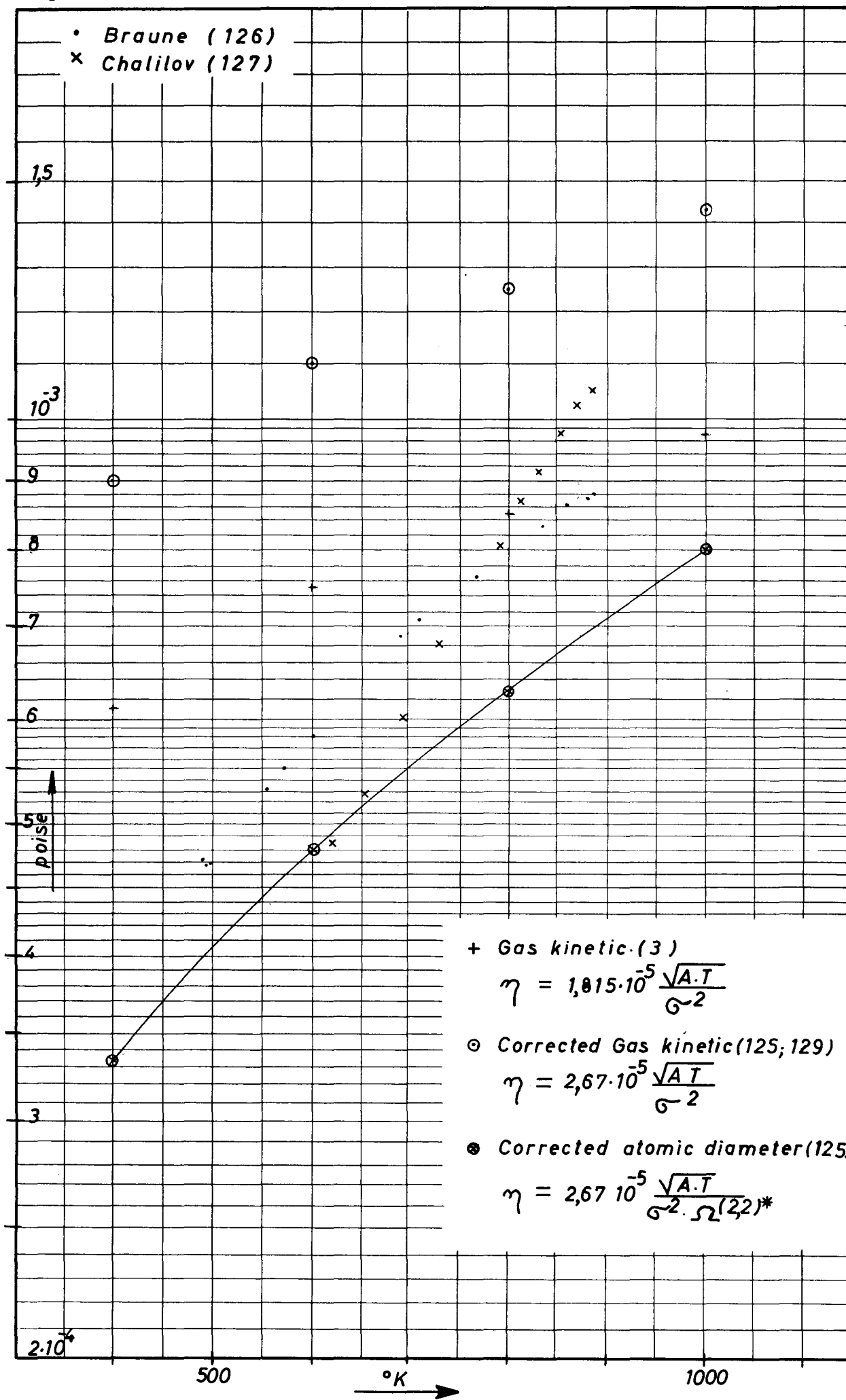
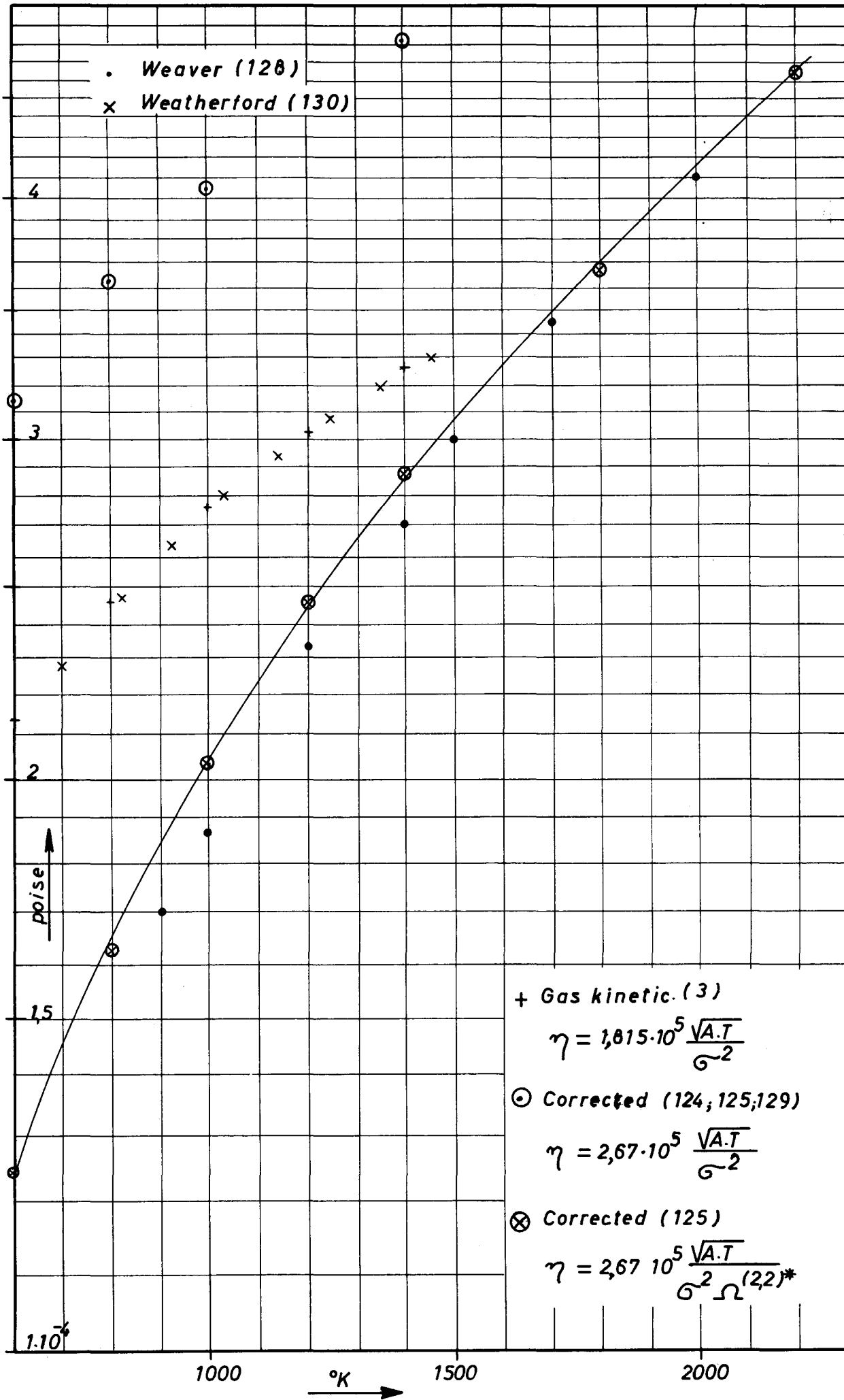
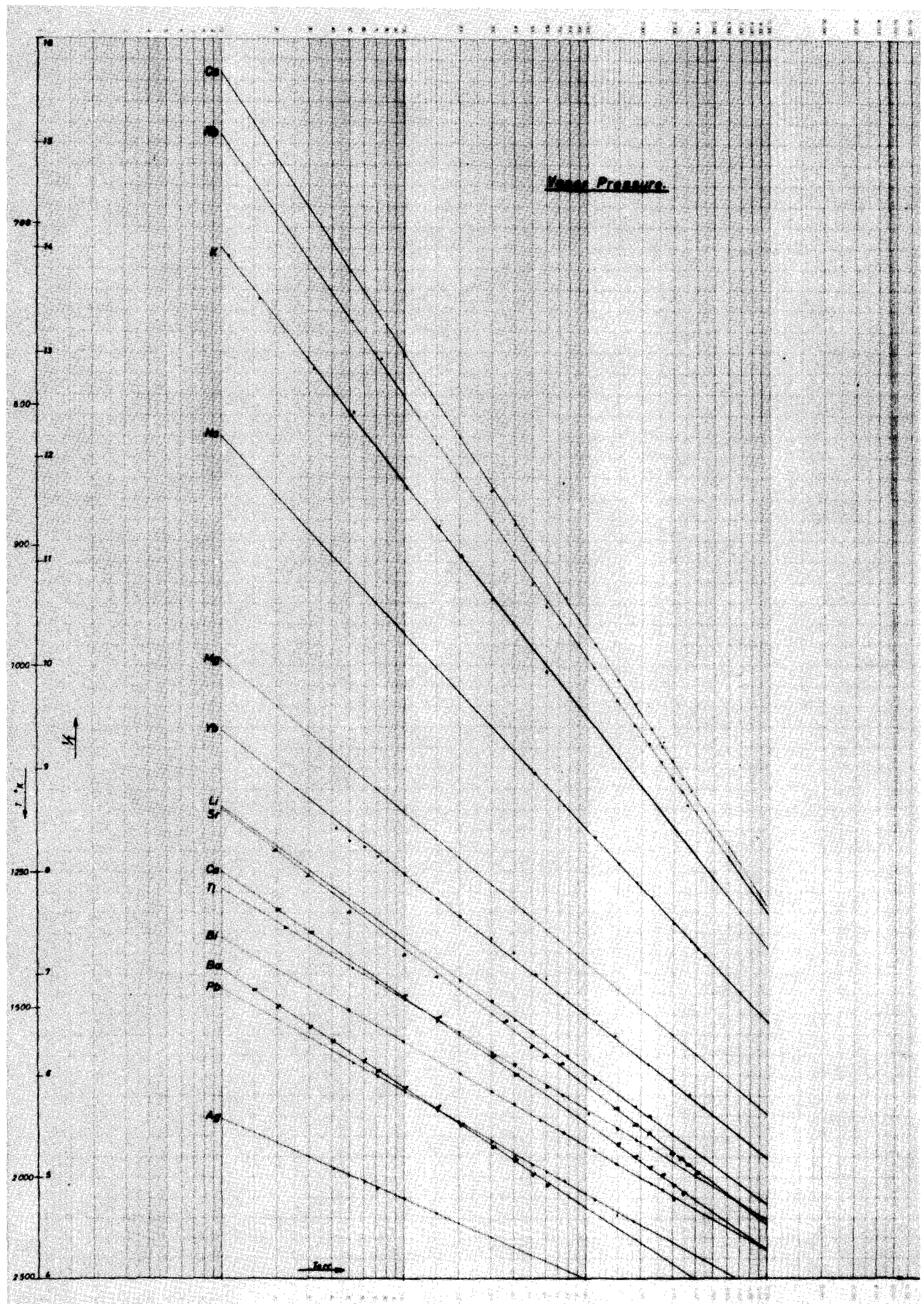
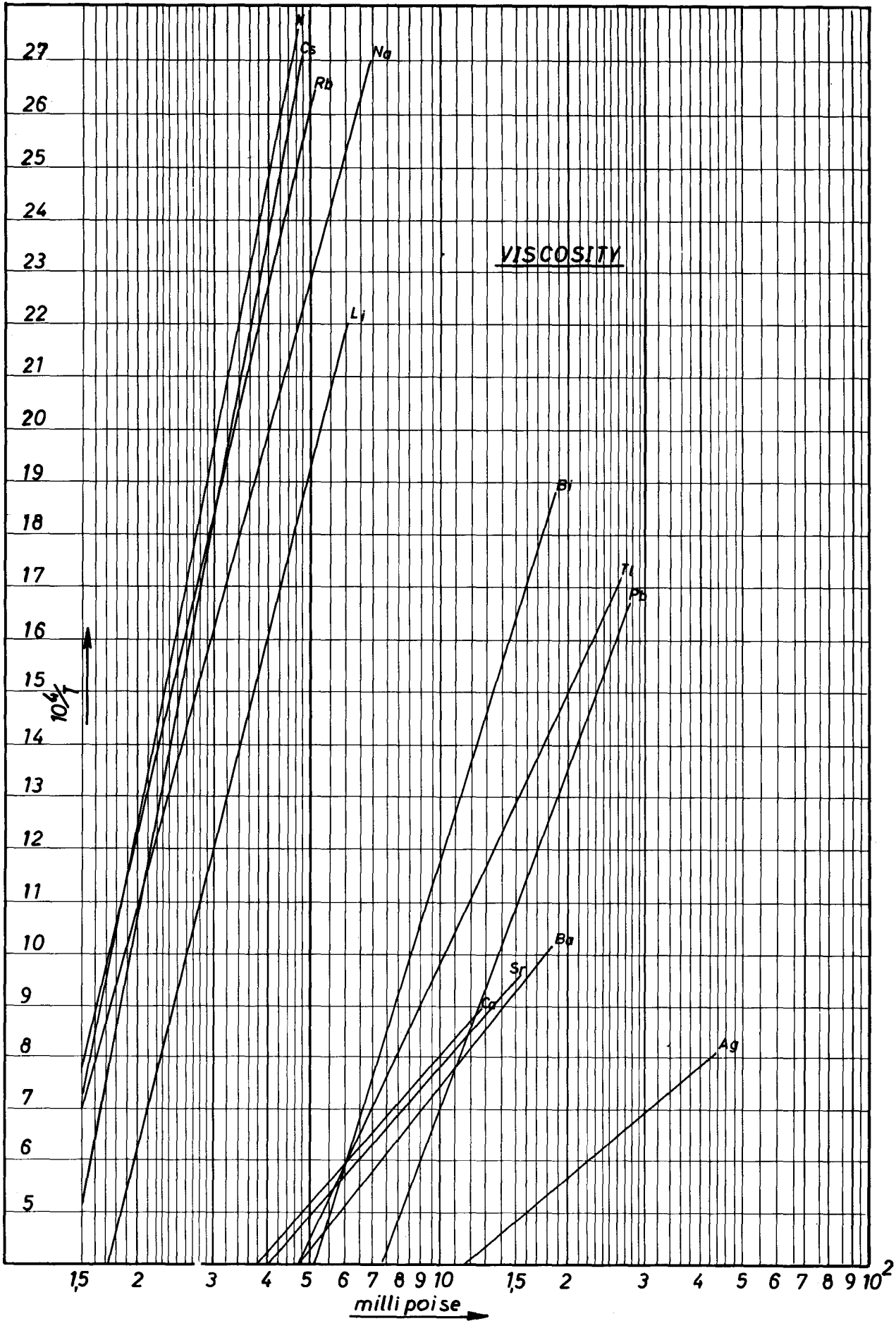


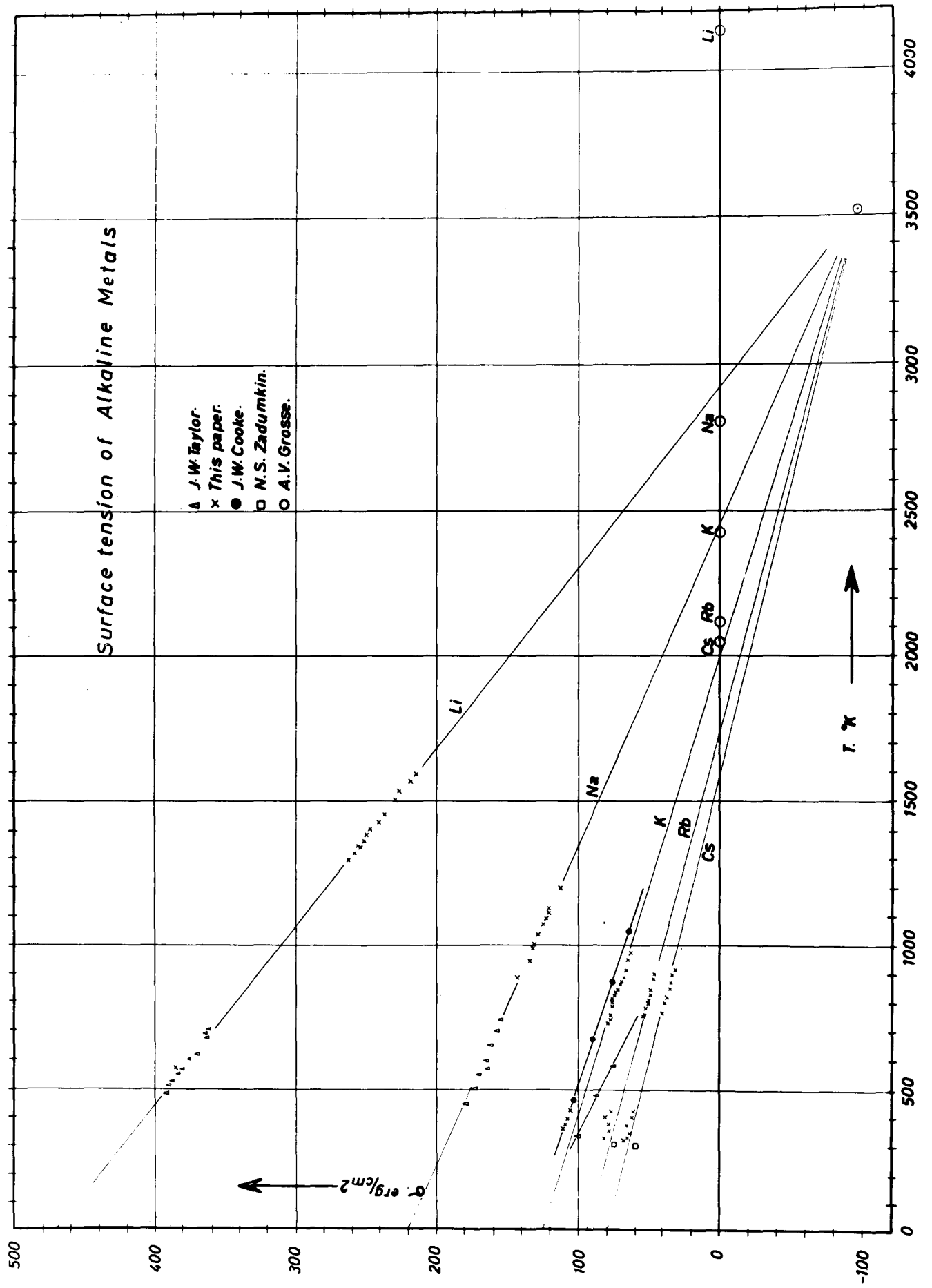
Fig. 9

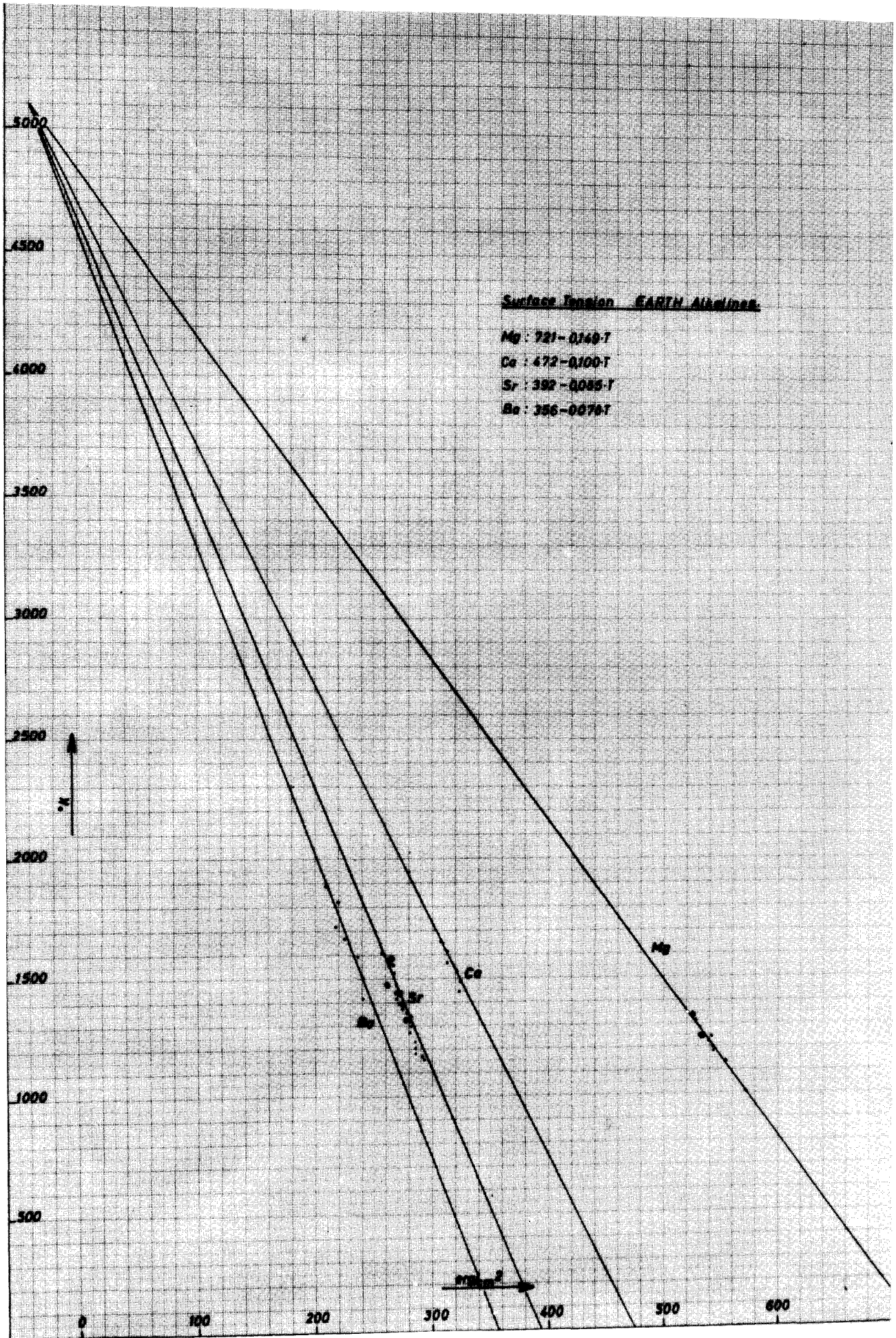
Vapor Viscosity Cs.

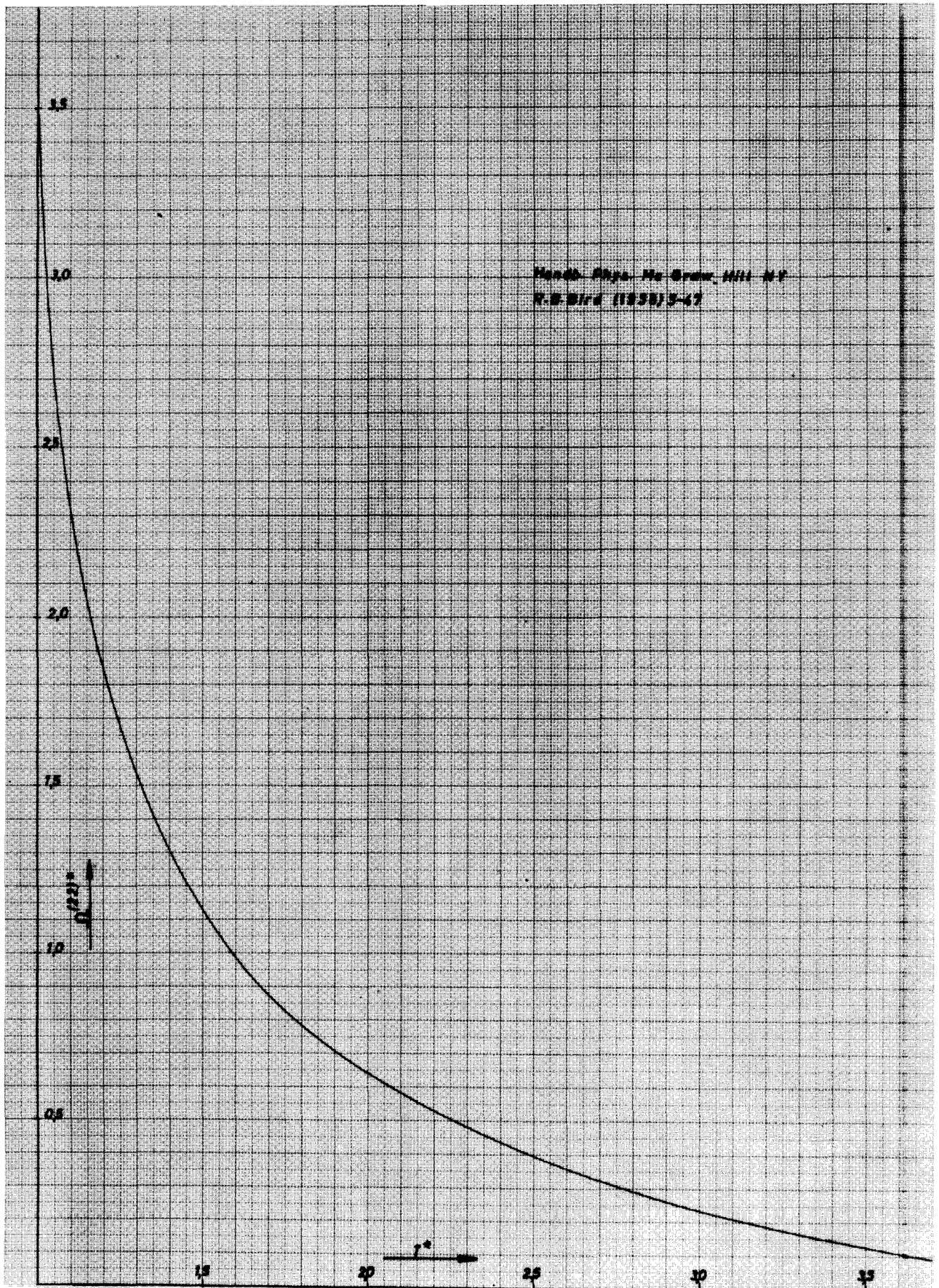












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Mr. R. van Wijk did much of the construction- and experimental work and made all the plots of this report.



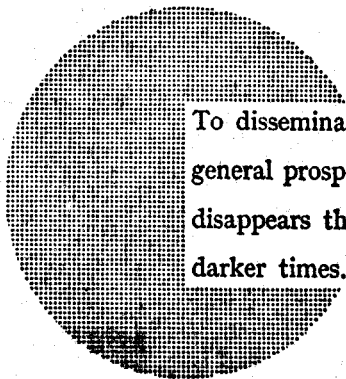
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