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January 1996

CMMT(A)9

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ABSTRACT

This report presents the results of an intercomparison of extrudate swell measurements of a polyethylene melt using capillary extrusion rheometers. The measurements have been made using a variety of makes of capillary extrusion rheometer and methods for measuring the dimensions of the extrudate. The measurements were carried out as part of a larger round robin on the measurement of shear viscosity and entrance pressure drop values based on ISO 11443 Plastics - Determination of the fluidity of plastics using capillary and slit die rheometers. The various techniques that were used are described and the extrudate swell results obtained are compared. Recommendations are made for the improved specification of the measurement of extrudate swell presented in ISO 11443.

ISSN 1361-4061

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CONTENTS

1. INTRODUCTION

This report presents the results of an intercomparison of extrudate swell measurements of a polyethylene melt using capillary extrusion rheometers, based on the international standard ISO 11443 Plastics - Determination of the fluidity of plastics using capillary and slit die rheometers (1). This work has been undertaken as part of the MTS programme on processability, 'Project PMP5, Polymer Moulding Models: The Determination of the elasticity of polymer melts'.

An earlier report by Rides (2), prepared as part of this research programme, indicated that industrialists were keen to employ the method of extrudate swell measurement using a capillary rheometer. The extrudate swell technique is particularly appropriate for characterising materials as it closely mimics the process of extrusion. In rheological testing it is desirable to use a test method that mimics as closely as possible the process for which the data are required. However, the repeatability and reproducibility of this technique has been questioned. This intercomparison has been carried out to improve the understanding of this measurement technique and to improve the specification for extrudate swell measurement that is currently in ISO 11443.

The measurements were made using a variety of makes of capillary extrusion rheometer and various techniques and procedures for measuring the dimensions of the extrudate. The measurements were carried out as part of a larger round robin on the measurement of shear viscosity and entrance pressure drop values based on ISO 11443. The various techniques that were used in this intercomparison are described and the extrudate swell results obtained are compared. Recommendations are made for the improved specification of the measurement of extrudate swell for ISO 11443.

2. EXTRUDATE SWELL MEASUREMENTS

All testing was based on the ISO standard 11443 Plastics - Determination of the fluidity of plastics using capillary and slit-die rheometers. Results are presented for measurements made on a high density polyethylene (NPL reference HFU000, round robin reference material A) at two temperatures, 190 °C and 210 °C. Five laboratories took part using four different capillary rheometer makes, namely: C.B.E. RACER, Gottfert 2002, Rosand RH7-2C and Toyo Seiki Seisaku-sho Caprio Graph 1C. Details of the instruments and the conditions used are presented in Table 1. The standard ISO 11443 defines two principle methods for performing extrudate swell measurements. The broad principles of the two methods are summarised as follows:

Method 1

The extrudate sample is removed from the rheometer after extrusion and allowed to cool in a suspended state in air to room temperature. Its diameter is then measured using a micrometer.

Method 2

The extrudate diameter is measured below the die exit during extrusion by use of a non-contacting photographic or optical measuring instrument. Measurements are made preferably at a distance of 10 mm below the die exit, although other distances may be used.

Full specification of the methods is given in ISO 11443(1). In this study two laboratories employed method 1. The remaining three laboratories employed method 2, each taking

measurements of the extrudate diameter at a different distance from the die exit. Extrudate swell measurement method 2 is considered to be more suitable for automation.

3. RESULTS AND DISCUSSION

Extrudate swell results obtained using method 1 with dies of different length but of the same diameter of 1 mm and at 190 °C are compared in Figure 1. The extrudate swell increased with increasing apparent shear rate where the apparent shear rate is defined as $4Q/\pi R^3$ and were Q is the volume flow rate and R is the radius of the capillary die. The effect of die length on extrudate swell is clearly demonstrated despite the relatively high scatter in results. The use of a longer die resulted in less extrudate swell. The same trends were also exhibited using method 1 at 210 °C, Figure 2. Equivalent results obtained using method 2 at 190 °C and 210 °C are shown in Figures 3 and 4 respectively and include results obtained using 2 mm diameter dies. The behaviour as measured using method 2 was qualitatively broadly similar to that using method 1. However, significant differences are apparent between the two sets of results, particularly in their magnitudes, and these are discussed further below.

It was noted that for the extrudate swell measurements at 210 °C made by laboratory 18 the values of extrudate swell decreased above an apparent shear rate of 100 s^{-1} whereas values obtained by other laboratories exhibited an increasing trend, Figure 4. However, the results obtained by all the laboratories at 190 °C all exhibited the same trend of increasing extrudate swell with increasing apparent shear rate, Figure 3. The main difference in the measurement techniques between laboratories 7, 8 and 18 is that laboratories 7 and 8 made measurements 70 mm and 60 mm below the die exit respectively, whereas laboratory 18 made measurements at 10 mm below the die exit. A possible explanation of this difference in behaviour is that the melt extrudate undergoes a delayed swell effect on exiting the die, this effect being associated with flows in which the inertia is significant - that is in cases where the viscosity is low and extrusion rate is high. As a consequence of the delayed extrudate swell the diameter of the extrudate may reach a maximum at more than 10 mm from the exit of the die, the distance from the die at which the extrudate reaches its maximum diameter being, in part, a function of the extrusion rate.

The effect on results obtained using method 2 of the distance below the die exit at which the measurements are made has not been systematically evaluated in this work. However, comparison of the results obtained by laboratory 18 with those of laboratories 7 and 8 indicate that the extrudate swell was smaller for laboratory 18, for which the measurement distance was 10 mm, than for laboratories 7 and 8 for which the distances were 60 and 70 mm respectively, figures 5 and 6. For laboratory number 8 is was not possible, using existing equipment, to carry out measurements at 10 mm below the exit of the die due to the design of the rheometer. More importantly, it may be undesirable to measure at 10 mm below the die exit as this may have a detrimental effect on the ability to control the temperature along the length of the die and up to the die exit. The contraction ratio from the barrel to the die was also smallest for laboratory 18 (Table 1) and this might also contribute to the lower die swell.

To aid comparison of the results obtained by the various laboratories the values of extrudate swell at an apparent shear rate of 300 s^{-1} have been determined by interpolation of the data presented in Figures 1 - 4. These values are presented in Table 2. A value of 300 s^{-1} was chosen because all laboratories had results that either were at or spanned this apparent shear rate value. Extrudate swell results at an apparent shear rate of 300 s^{-1} and at 190 °C and 210 °C have been plotted as functions of die length and diameter, Figures 5 - 8. There is a consistent difference in extrudate swell values between the two method with method 1 giving higher values than method 2, Figure 5 and 6. This difference appears to be greater when using short length dies. The trend of decreasing extrudate swell value with increasing die

length is clearly exhibited at both test temperatures and using both methods, Figure 5 and 6. However, the results indicate that the effect of die diameter, for a given apparent shear rate, is not so significant on extrudate swell values, Figures 7 and 8. For method 2, the values obtained using 2 mm diameter dies were similar to those obtained using 1 mm diameter dies. This may be because the area contraction ratios from the barrel to the die were not significantly different as the 2 mm diameter die was used in conjunction with a 25 mm diameter barrel.

The results presented in Figures 5 and 6 at an apparent shear rate of 300 s^{-1} have been used to compile Table 3 that summarises the effects of method, die length/diameter and temperature on extrudate swell values. Due to the variations in measurements observed and as reported above, only general trends can be derived from the data. Allowing for this then the following trends were deduced on the basis of all the results presented.

- The percentage extrudate swell values measured in method 1 tests were in the range 65 - 135% at 190 °C and 55 - 135% at 210 °C where the values varied as functions of apparent shear rate and die dimensions. For method 2 the values were in the range 30 - 90% at 190 °C and 45 - 95% at 210 °C.

- For both methods 1 and 2, extrudate swell values were approximately 5 - 15% greater at 210 °C than at 190 °C.

- At 190 °C extrudate swell values increased with shear rate by approximately 10% over the apparent shear rate range 10 - 100 s^{-1}, and by approximately 10% over the range 100 - 1000 s^{-1}. At 210 °C extrudate swell values increased with shear rate by approximately 5 - 10% over the range 10 - 100 s^{-1}, and by approximately 5 - 10% over the range 100 - 1000 s^{-1}.

- The die length to diameter ratio had a significant effect on extrudate swell values. There was an increase in extrudate swell of approximately 50% in changing from a die of length to diameter ratio of 20/1 to 5/1.

- At an apparent shear rate of 300 s^{-1} method 1 extrudate swell results were approximately 25 - 30% greater than method 2 results.

The differences in results between methods 1 and 2 are considered to be consistent with the additional time for relaxation and thus increased swelling that occurs when testing in accordance with method 1.

In earlier work by Rides and Allen (3) using method 2 it was concluded that the use of entry cones reduced the scatter in swell results, enabled measurements to be made at higher shear rates as it delayed the onset of melt instabilities, and had no significant effect on the magnitude of the swell. Also, scatter was reduced by using longer dies and higher shear rates. However, these last two factors also affect the magnitude of the swelling as indicated above.

4. CONCLUSIONS

The following conclusions are made based on the above reported work and indicate how improved and hence more valuable measurements of extrudate swell can be made:

- Using the specification given in ISO 11443 for the measurement of extrudate swell a wide range in values for extrudate swell can be obtained depending on the technique and testing conditions used.

- The most significant test variables in determining extrudate swell for a typical range of testing conditions are extrusion rate and die length.

- The results presented above indicate that data generated using method 2 may have less scatter than those using method 1. Scatter can be reduced by using longer dies, smaller diameter dies (for a given extrusion rate) and entry cones although the first two have a significant effect on the magnitude of the swelling.

- The distance from the die exit at which measurements are made using method 2 and the contraction ratio from the barrel to die may have significant affects on results. Their effects needs to be investigated further.

- For valid comparison of results obtained on different materials it is necessary that tests are carried out using the same procedure, geometries and testing conditions.

5. RECOMMENDATIONS FOR REVISION OF ISO 11443 FOR EXTRUDATE SWELL MEASUREMENTS

It is recommended that the standard ISO 11443 should be revised to take into account these conclusions. In particular, it is proposed that the standard should be amended to include the following:

a) specification of the die geometry, ie single values of length and diameter that shall be used,

b) specification of the apparent shear rate(s) at which measurements shall be made;

c) improved specification of the distance between the die exit and the measurement position.

6. ACKNOWLEDGEMENTS

The research reported in this paper was carried out as part of the "Materials Measurement Programme", a programme of underpinning research financed by the United Kingdom Department of Trade and Industry.

The NPL would like to thank the participants in this intercomparison: CarnaudMetalbox plc, Solvay, Montell and Toyo Seiki Seisaku-sho.

7. REFERENCES

1 ISO 11443:1995(E) Plastics - Determination of the fluidity of plastics using capillary and slit-die rheometers.

2. Industrial needs for polymer melt elasticity measurements, Rides, M., NPL Report DMM(A)131, October 1993.

3. Measurements of the extrudate swell behaviour of polymer melts, M. Rides and C.R.G. Allen, NPL Report CMMT(D)20, March 1996.

Table 1 Specification of the instrument and method used by each of the laboratories

Lab No:	Rheometer	Barrel diameter, mm	Capillary diameter, mm	Die length, mm	Die entry geometry, degree	Measurement technique
7	Rosand RH7-2C	15	1	16	180	method 2, IR optical gauge, 70 mm below die
8	CBE Racer special	25	2	10 30 40	180	method 2, laser micrometer, 60 mm below die
9	Gottfert 2002	12	1	5 10 20	180	method 1, vernier calliper
11	Gottfert 2002	12	1	5	180	method 1, micrometer
18	TSS Capiro Graph 1C	9.55	1	5 10 20	180	method 2, laser micrometer, 10 mm below die

Table 2 Extrudate swell results at an apparent wall shear rate of 300 s^{-1}.

Lab No (and test method M1 or M2)	Barrel diameter, mm	Die length, mm	Die diameter, mm	Extrudate swell % at 190 °C	Extrudate swell % at 210 °C
7 (M2)	15	16	1	62	75
8 (M2)	25	10	2	74	85
8 (M2)	25	30	2	56	57
8 (M2)	25	40	2	-	61
18 (M2)	9.55	5	1	77	89
18 (M2)	9.55	10	1	58	65
18 (M2)	9.55	20	1	52	57
9 (M1)	12	5	1	105	115
9 (M1)	12	10	1	85	-
9 (M1)	12	20	1	70	75
11 (M1)	12	5	1	115	-

Table 3 Approximate extrudate swell values taken from curves drawn for data obtained at 300 s^{-1}.

Method 1		
Temperature	Die ratio, length / capillary diameter	Extrudate swell, %
190 °C	20/1	70
	5/1	105
	difference	35
210 °C	20/1	75
	5/1	115
	difference	40
Method 2		
Temperature	Die ratio, length / capillary diameter	Extrudate swell, %
190 °C	20/1	52
	5/1	77
	difference	25
210 °C	20/1	57
	5/1	89
	difference	32
Difference between Methods 1 and 2		
Temperature	Die ratio, length / capillary diameter	Difference in extrudate swell, method 1 - method 2
190 °C	20/1	18
	5/1	28
210 °C	20/1	18
	5/1	26

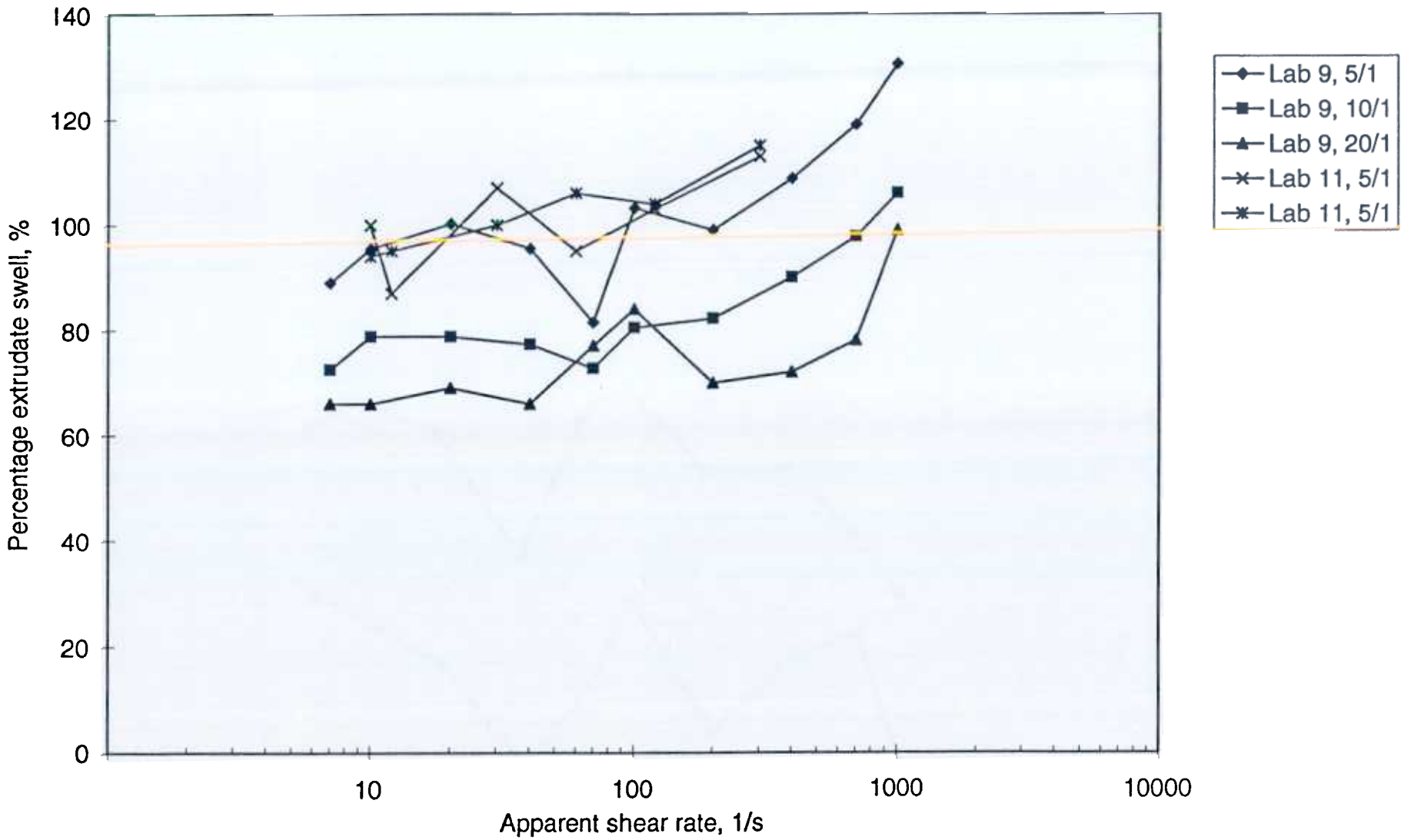

Figure 1 Comparison of extrudate swell results using method 1 for HFU000 at 190 °C.
Legend specifies the laboratory number and the capillary die length (mm) / diameter (mm) ratio.

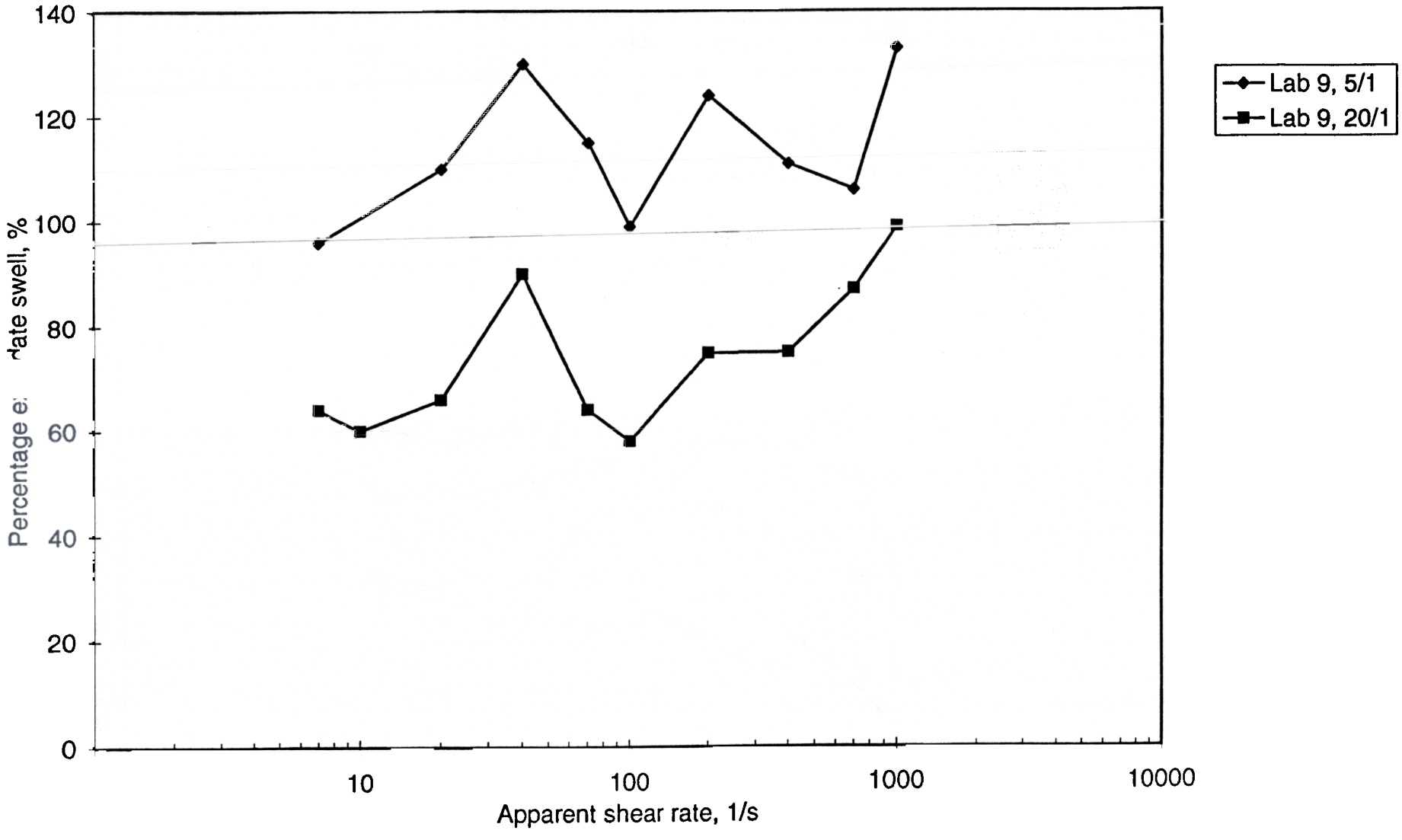

Figure 2 Comparison of extrudate swell results using method 1 for HFU000 at 210 °C.
Legend specifies the laboratory number and the capillary die length (mm) / diameter (mm) ratio.

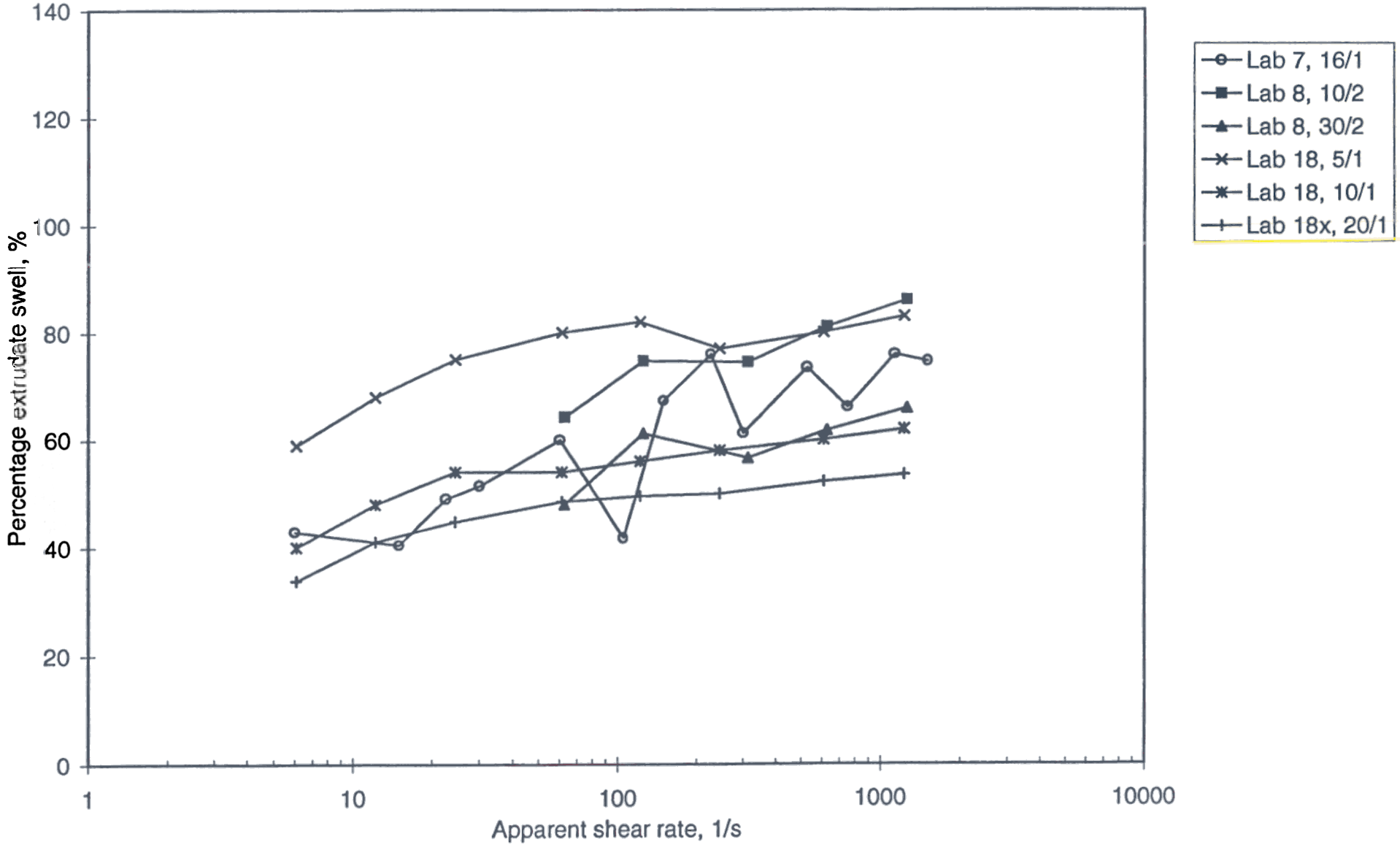

Figure 3 Comparison of extrudate swell results using method 2 for HFU000 at 190 °C.
Legend specifies the laboratory number and the capillary die length (mm) / diameter (mm) ratio.

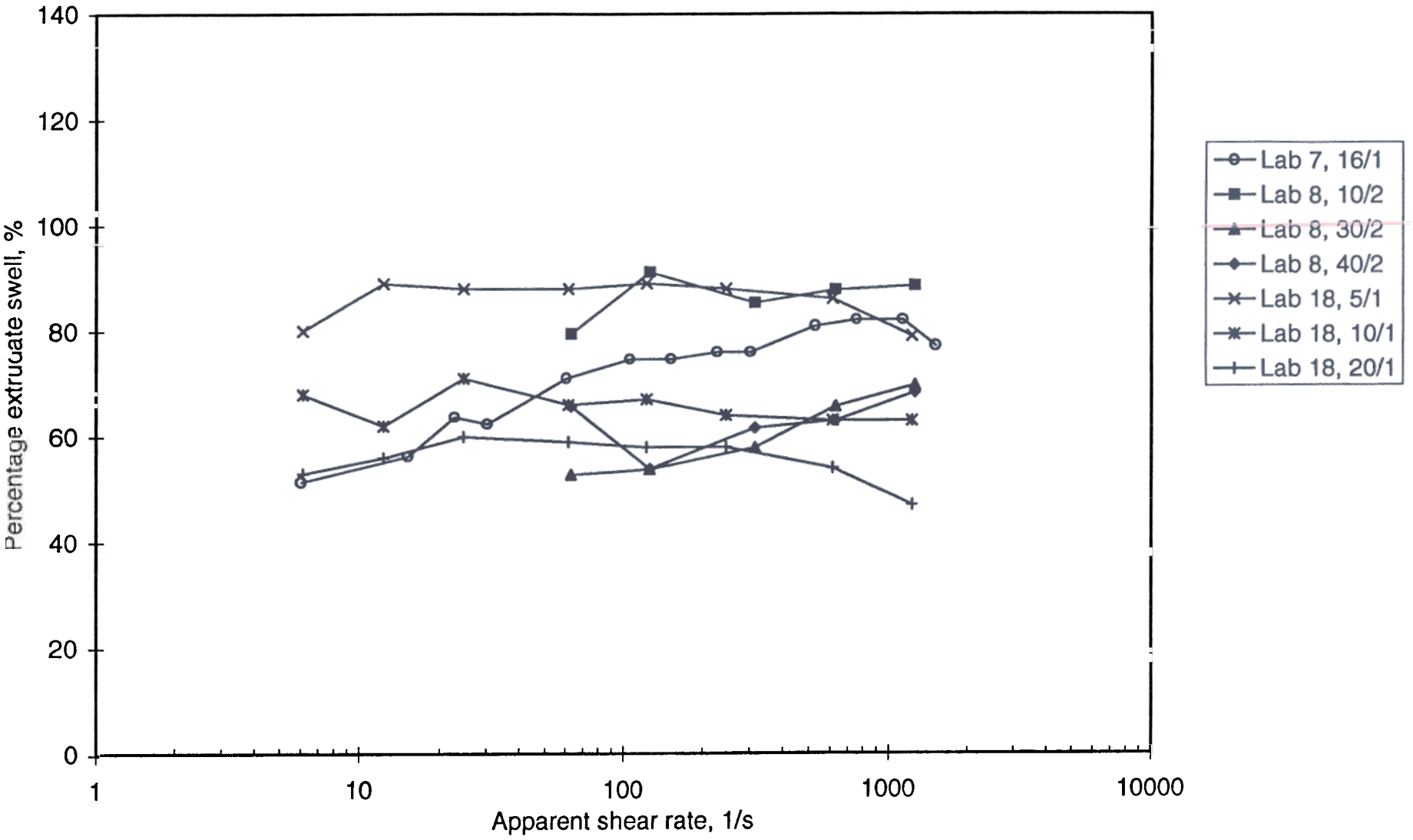

Figure 4 Comparison of extrudate swell results using method 2 for HFU000 at 210 °C.
Legend specifies the laboratory number and the capillary die length (mm) / diameter (mm) ratio.

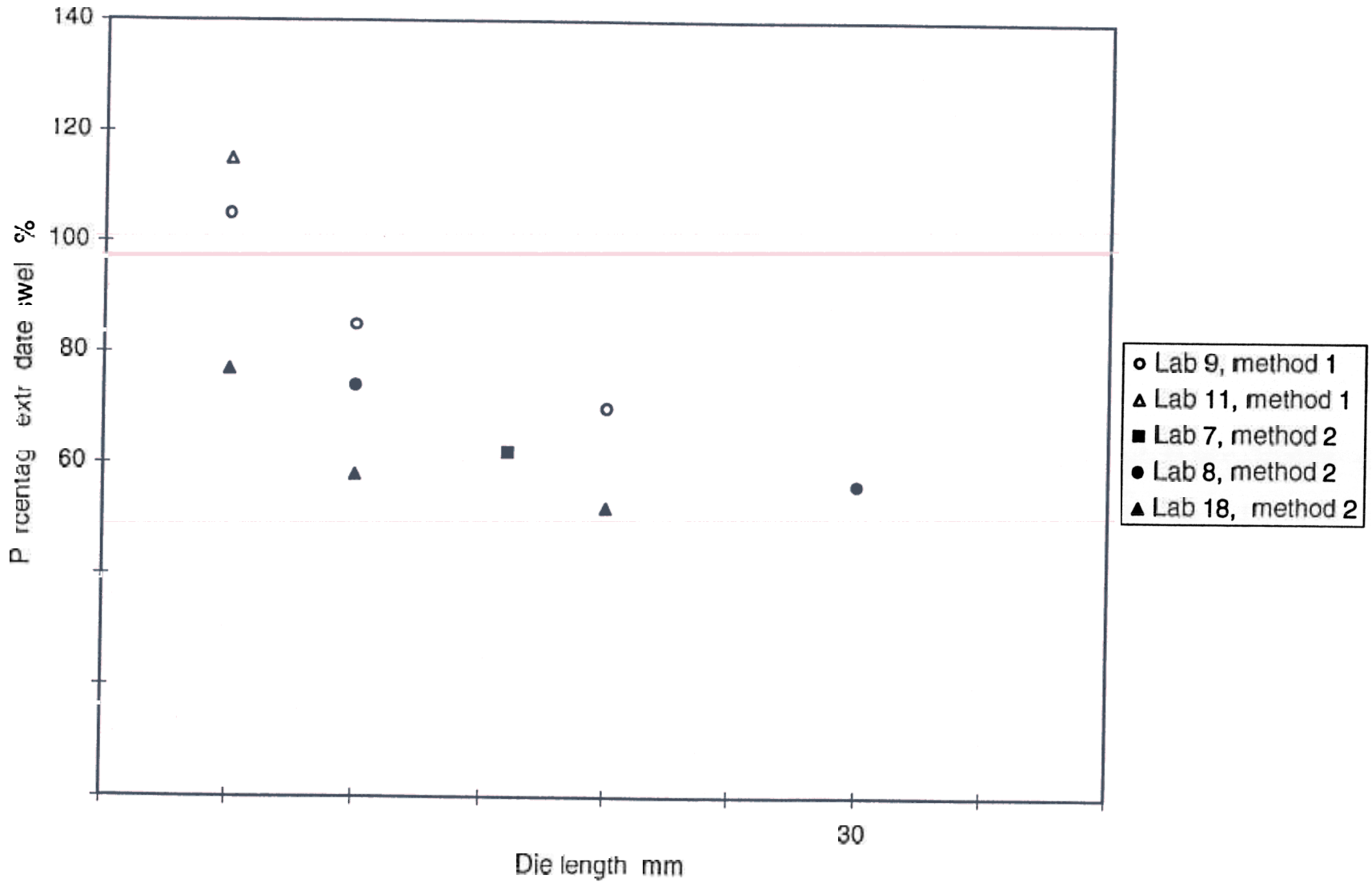

Figure Comparison methods used and the effect die length extrudate swell values for HFU000 190 °C and 300

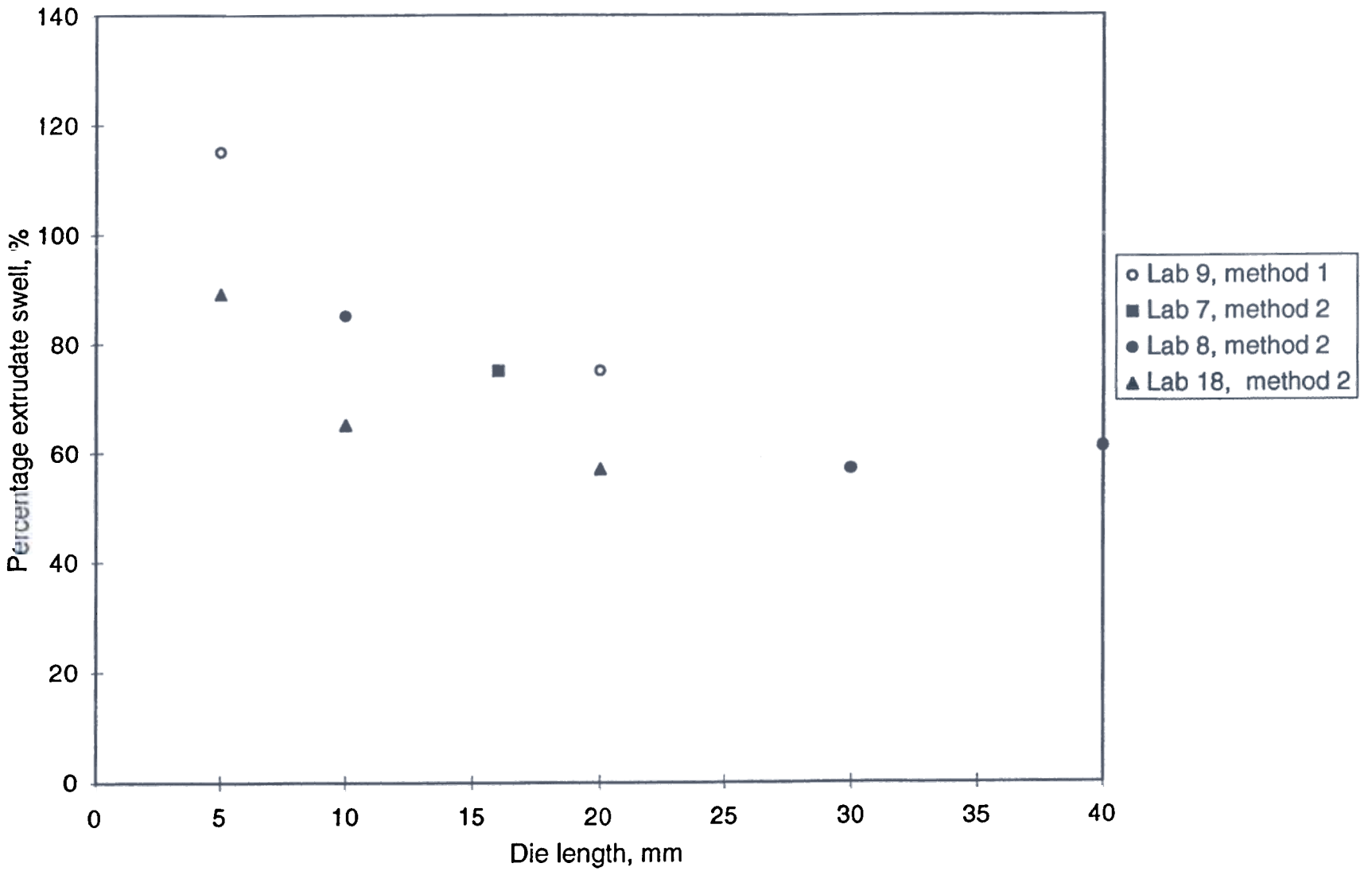

Figure 6 Comparison of methods used and the effect of die length on extrudate swell values for HFU000 at 210 °C and 300 s^{-1}.

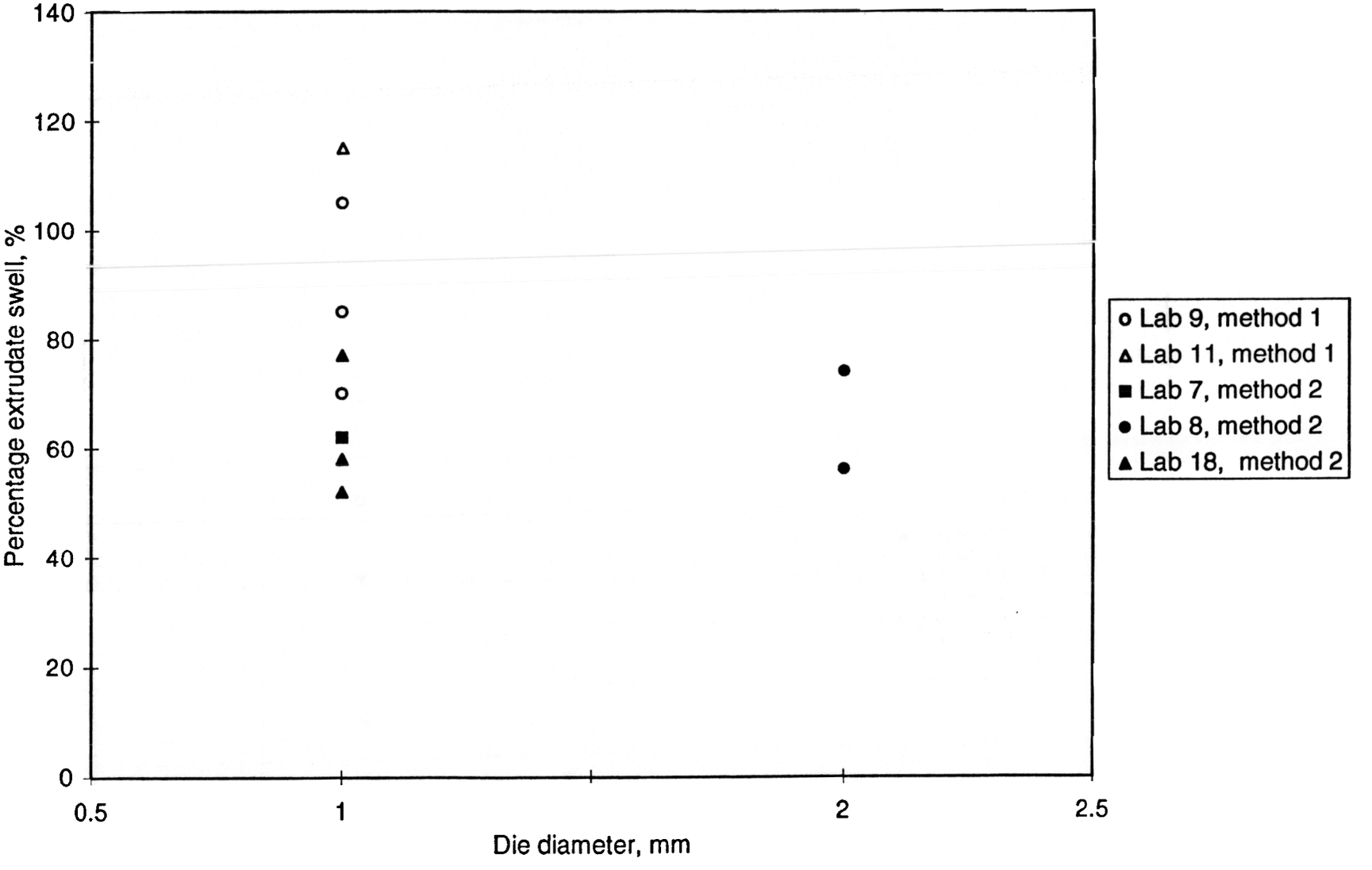

Figure 7 of methods used and the effect of die on extrudate swell values for HFU000 at 190 °C and 300 s^{-1}.

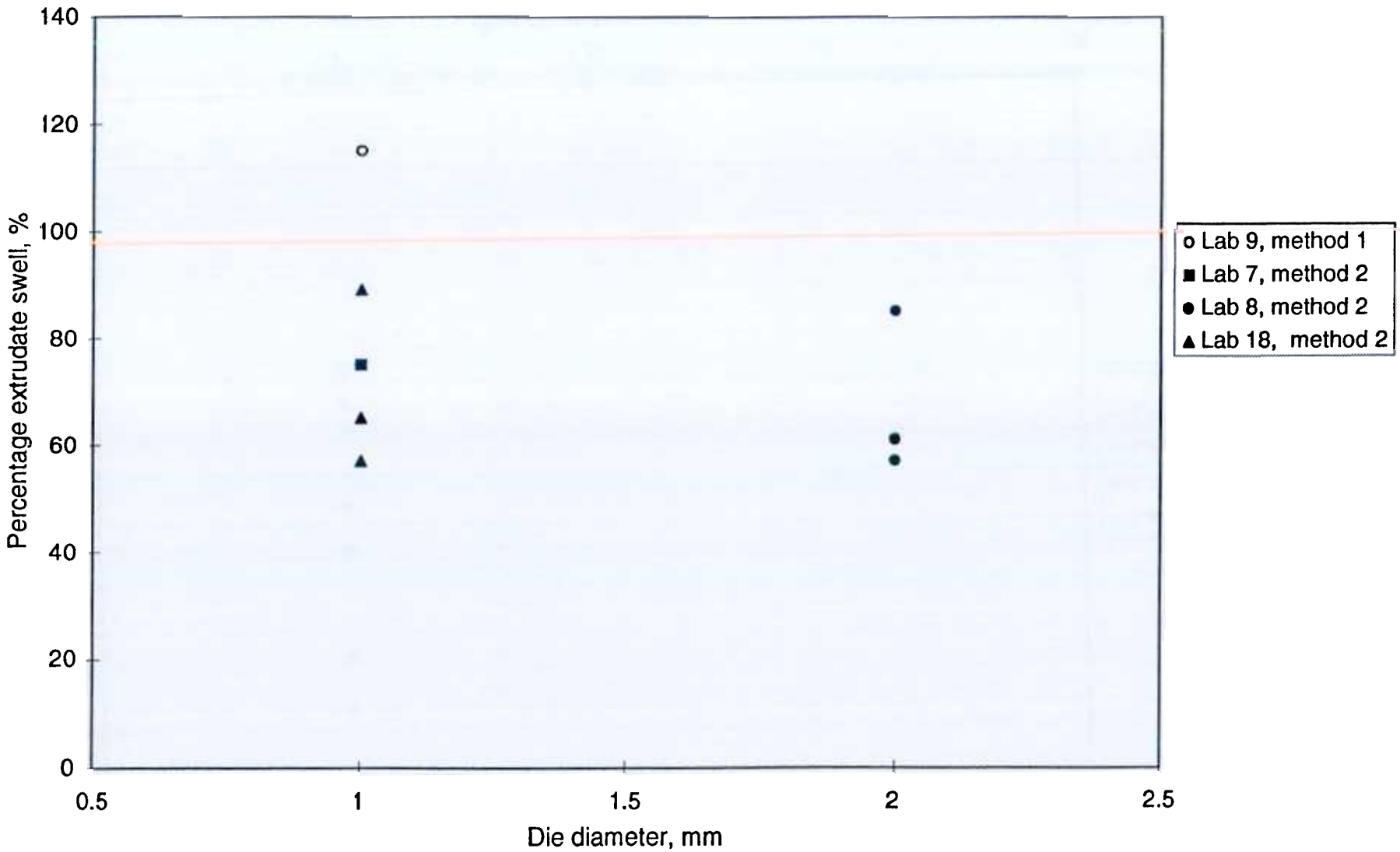

Figure 8 Comparison of methods used and the effect of die diameter on extrudate swell values for HFU000 at 210 °C and 300 s^{-1}.