The influence of bubble mechanics on some properties of blown film

Christopher J. S. Petrie and Ann Petrie
Department of Engineering Mathematics,
University of Newcastle, Newcastle upon Tyne, NE1 7RU, U.K.

25th February, 1999

Abstract

A series of experimental runs were carried out on a small blown film line to investigate the effect of processing parameters on the mechanical properties of the film that was produced. Three different dies were used and for each die the throughput (mass flow rate), freeze line height and blow-up ratio were varied, with take-up velocity adjusted to give a fixed nominal gauge (film thickness). One polymer (a low density polyethylene) was used throughout and the extrusion temperature was also kept constant.

The measured properties included machine direction tensile strength and transverse direction tensile strength. The best correlation was found to be with characteristic strain rates in the machine and transverse directions. These quantities have the advantage that they can be estimated from easily measurable process parameters and offer the possibility of estimating effects due to scale-up.

1 Introduction

About 25 years ago, the mechanical properties (and some others) were measured for LDPE film produced under systematically varied conditions. This was reported only in an internal report to Visqueen Ltd., then part of ICI Plastics (Petrie, 1975b), and is now published for the first time. In this paper we concentrate on a few aspects of the results, as a basis for predicting film properties from processing conditions. First of all we shall mention recent related work, to set these results in context.

It is perfectly possible to undertake a research project and write a paper on the film blowing process without discussing the extensional flow that takes place in the film bubble at all (Debbaut *et al.*, 1998). We start with this remark not in any spirit of controversy – indeed the emphasis in this paper is not, in fact, on the **analysis** of the extensional flow – but because we want to emphasize, right from the start, the limitations of this study. The work of Debbaut *et al.* addresses primarily questions of extrusion line design and optimization, looking both at extruder screw design and spiral mandrel die design. The polymer which they study is LLDPE rather than LDPE and limitations in production rate are identified as arising primarily from the occurrence of surface defects (sharkskin) and from bubble instability. Here we shall ignore the important aspects of the process on which Debbaut *et al.* concentrate and continue to urge the importance of extensional flow (Petrie, 1979, 1997).

In another piece of relatively recent work, Liu, Bogue and Spruiell (1995a) report on experimental studies of the interrelation between the various bubble parameters (blow-up ratio, draw ratio or take-up ratio, inflation pressure, melt temperature and cooling air flow) for three polymers. One motivation they give for this is to seek an understanding of what they term the "counterintuitive" behaviour of models of the blown film bubble – namely that an increase in bubble radius is associated with a decrease in inflation pressure. We shall not embark on a detailed discussion of this work here, but note that Liu *et al.* observe "intuitive" behaviour at low values of the blow-up ratio (BUR) (between 0.5 and 1.5) and "counterintuitive" behaviour at high BUR (between 2 and 3) under some circumstances. These experimental observations are complemented (Liu, Bogue and Spruiell, 1995b) by a simplified theory which they compare with the established theory (Pearson and Petrie, 1970a, 1970b, 1970c; Petrie 1975b, 1983). There are differences, but Liu *et al.* conclude their second paper by remarking that "The reason for this difference is not understood".

Kurtz (1992), in a list of key challenges which he identifies for polymer processing, gave blown film bubble stability as one of his five key challenges. Sharkskin and scale-up are two more which are highly relevant to the film blowing process. He later emphasizes (Kurtz, 1995) the point that prediction of stresses, rather than bubble shape, is the real test of success in modelling the blown film process. Petrie (1997) takes this up and develops the point that stresses in the film (at the freeze-line in particular) are the key factor that one can relate on the one hand to processing parameters and on the other hand to the microstructure and properties of the film produced. In a sense the work of Liu *et al.* is pursuing the same goal, since the inflation pressure, whose behaviour they discuss, is directly related to the stresses in the film.

We seek, here, to make a contribution to the task of relating the properties of blown film to processing parameters We consider those parameters which are most obviously related to the biaxial stretching of the film which takes place after extrusion, in the blown film bubble as the polymer cools and solidifies. At this stage we consider how far the task can be accomplished without detailed analysis of the flow, the heat transfer or the microstructure of the polymer. This might be termed a pragmatic view, or possibly a gross over-simplification. Authors, such as those cited above, have concentrated on other aspects of the task and it is clearly desirable to bring all these approaches together. One difficulty is that, in the cited work (Debbaut *et al.*, 1998; Liu *et al.*, 1995) film properties are not reported.

The work which seems to be closest in spirit to ours is that of Tas (1994). He is the only author of whom we are aware who reports directly on relations between processing parameters and film properties. He clearly identifies the stresses in the film as it solidifies as the key quantities which link the mechanics of the film formation to its final properties. There are obvious similarities, as well as clear differences, between Tas' study and ours.

Tas studies three different polymers using a single die, while we use a single polymer and three different dies. Both here and in Tas' work, LDPE is used and there is no attempt to investigate the effect of extrusion temperature. Extruder operation is not altered at all by Tas (except for the screw speed and hence the output rate in one run out of the 30 that he reports), while we have systematically varied output rate. Tas has also kept the freeze-line height (FLH) fixed (for each polymer) while we have adjusted the cooling air flow to give two or three different freeze-line heights with other parameters being kept the same. We shall allude to other aspects of Tas' work in the discussion below. The two sets of results are largely complementary and more detailed study of both sets might prove informative.

One final remark is worth making here, with possible comparisons between this and other work in mind. It is very easy **to say** "we study the effect of one parameter, holding the others constant". However, with the film blowing process in particular, it is not easy **to achieve** this and not at all easy to do so **unambiguously**. The point is this: if we say we are studying the effect of blow-up ratio, then clearly we supply more air to the bubble interior and inflate the bubble. If we do not adjust the nip roll speed, then the film thickness will be reduced as the transverse direction stretching is increased, with machine direction stretching unaltered. If, on the other hand, we wish to make the comparison for the same thickness of film, we must reduce the nip roll speed as we increase the blow-up ratio. In either case, if the cooling airflow is not adjusted, the freeze-line height will change. Almost certainly whatever else we do, the bubble shape will change and this, like the temperature history along the bubble, is outside our direct control, and is not totally controllable by any means.

Hence it is important to specify in detail what changes are made and what quantities are adjusted is any series of experiments. Some of the contradictory information, for example that which is summarized in Table 4 below, can be explained in this way. Of course such a table oversimplifies, which in itself may explain the fact that there are apparent contradictions.

2 Experimental procedure

The polymer used was a film grade low density polyethylene (LDPE) produced by ICI (XJF 46/51, MFI = 2). It had a zero shear viscosity of around 20,000 Nsm^{-2} at the extrusion temperature, which was kept more or less constant at around 160C. A 2.5” (6.3cm) extruder capable of delivering up to 120Kg/hr was used with three different dies, of diameters 4” and 8” (10cm and 20cm) and die gaps 0.033” and 0.066” (0.084cm and 0.167cm). The three dies have dimensions given in Table 1. These dies are appreciably larger than those in the experiments of Tas (4cm die diameter) and Liu *et al.* (1.5cm die diameter).

Table 1: Experimental die dimensions.

Die	Diameter (cm)	Gap (cm)
A	10	0.084
B	20	0.084
C	10	0.167

Table 2: Missing runs.

Run no:	Nominal BUR	Die	Nominal output lb/hr/in	FLH cm	Comment
11	2	A	8	40-60	Instability (lack of cooling capacity)
12	2	A	6	40-60	Achieved (no properties measured)
24	2	B	8	80	Instability (lack of cooling capacity)
30	2	C	4	80	Instability
36	2	C	8	40	Instability

A set of 40 experimental runs were planned, of which 4 were not carried out because of failure to achieve steady operation at the planned conditions. A fifth run was carried out, but film properties were not measured, so that we have 35 sets of results. The missing runs are given in Table 2. The runs were planned for two values of the “nominal BUR”, three values of the extruder output and two freeze-line heights, giving 12 runs with each die. For two of the sets of conditions with die A, a third freeze-line height was tried, and for two sets of conditions with die C a film of gauge 76μ was

Table 3: Experimental runs.

Run no:	Nominal BUR	Die	Flow rate g/s	FLH cm	DR V_1/V_0	BUR R_1/R_0
1	2	A	6.3	50	16.72	1.29
4	2	A	6.3	60	16.02	1.31
5	2	A	9.4	80	16.02	1.25
10	2	A	12.5	75	16.65	1.33
15	2	B	12.5	40	16.25	1.29
16	2	B	12.5	75	16.67	1.3
20	2	B	18.7	45	17.8	1.28
19	2	B	18.7	65	19.08	1.27
23	2	B	25	65	18.43	1.28
27	2	C	6.3	42	32.79	1.32
31	2	C	9.4	40	36.62	1.28
32	2	C	9.4	60	36.24	1.26
35	2	C	12.5	70	33.32	1.3
3	4	A	6.3	45	8.59	2.52
2	4	A	6.3	80	8.69	2.71
13	4	A	9.4	43	8.14	2.59
7	4	A	9.4	65	8.11	2.53
6	4	A	9.4	80	8.39	2.58
14	4	A	12.5	40	7.58	2.57
8	4	A	12.5	50	8.6	2.51
9	4	A	12.5	78	8.62	2.51
18	4	B	12.5	65	8.53	2.47
17	4	B	12.5	75	7.7	2.47
22	4	B	18.7	65	9.12	2.5
21	4	B	18.7	80	9.06	2.52
26	4	B	25	70	9.23	2.55
25	4	B	25	80	9.39	2.5
29	4	C	6.3	40	16.13	2.54
28	4	C	6.3	80	17.15	2.52
34	4	C	9.4	63	16.26	2.59
33	4	C	9.4	75	16.31	2.58
38	4	C	12.5	57	16.25	2.52
37	4	C	12.5	80	16.59	2.54
39	4	C	6.3	85	9.16	2.57
40	4	C	12.5	80	8.69	2.55

produced, instead of the 38μ gauge produced for all the other runs. The "nominal BUR" is the ratio of layflat width to die diameter, $\pi R_1/2R_0$, so that "nominal BUR" values of 2 and 4 correspond to actual values, $\mathrm{BUR} = R_1/R_0 = 1.27$ and 2.55. The extruder output was set at 4, 6 and 8lb/hr/in (mass flow rate per unit of die circumference) so that actual mass flow rates for die B were double those for dies A and C.

The process parameters set for the experimental runs are given in Table 3, the run number being that from the planned order of the experiments. Runs 39 and 40 are the two which gave the film of double thickness. Achievable values of FLH for this extruder/die/cooling system combination generally lay between 40cm and 85cm. The high FLH instability involved a varicose oscillation in bubble shape, often observed at start-up and in a few cases never damped out. The practical cure for this is normally to increase the cooling airflow, hence the attribution of failure to lack of cooling capacity. With low FLH, an instability involving switching of the bubble between running clear of the air ring and sitting on the air ring deflector was a limitation. In runs 8, 13, 14 and 38 the bubble was sitting on the deflector, and this does appear to affect some film properties. Run 20 was thought to be close to this condition.

3 Results

The relation between machine direction tensile strength (MDTS) and a characteristic machine direction strain rate (MDSR) is shown in Figure 1. Considering the large variety of processing conditions and the large number of factors that are varied, this is a remarkably simple relation for predicting one mechanical property of the film. The MDSR is defined as the logarithmic strain in the machine direction, $\ln(V_1/V_0)$ divided by a characteristic time, taken to be the freeze-line height (FLH) divided

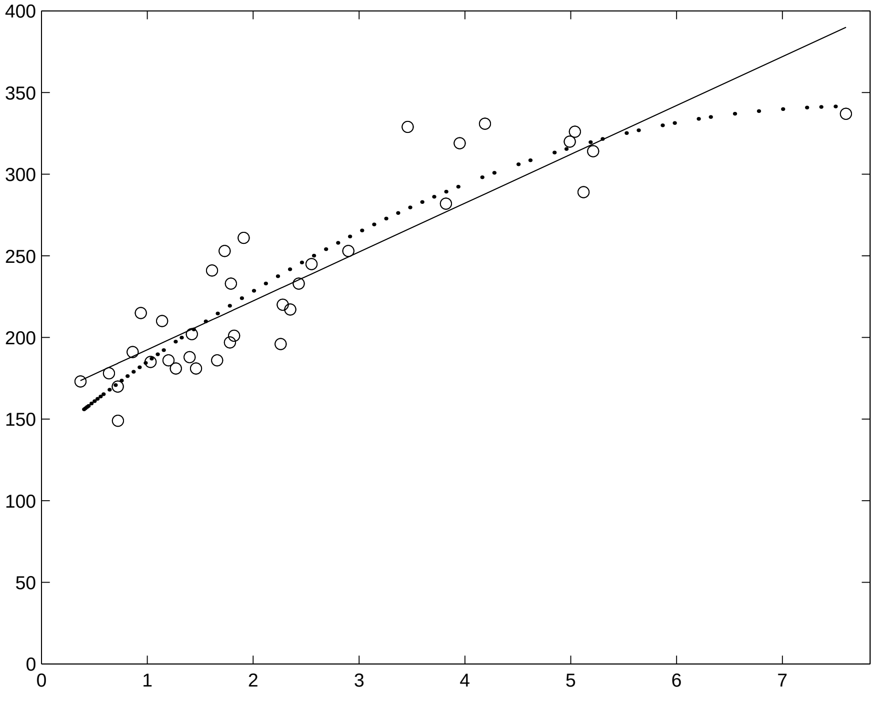

Figure 1: MDTS plotted against MDSR, with fitted linear and quadratic regression lines.

by the velocity of the film at the freeze-line, V_1:

$$\mathrm{MDSR} = (V_1/\mathrm{FLH}) \ln(V_1/V_0) \qquad (1)$$

The arbitrariness of the choice of characteristic time in the MDSR may be investigated by using a second characteristic strain rate, $\mathrm{MDSR2} = (V_0/\mathrm{FLH}) \ln(V_1/V_0)$ and it can be seen in Figure 2(a) that this is not as helpful; the scatter of data about the fitted line is much greater than in Figure 1. Since Tas presents his results as a function of draw ratio, DR ($= V_1/V_0$), we examine this possibility in Figure 2(b) and find that, for our results it is much less informative.

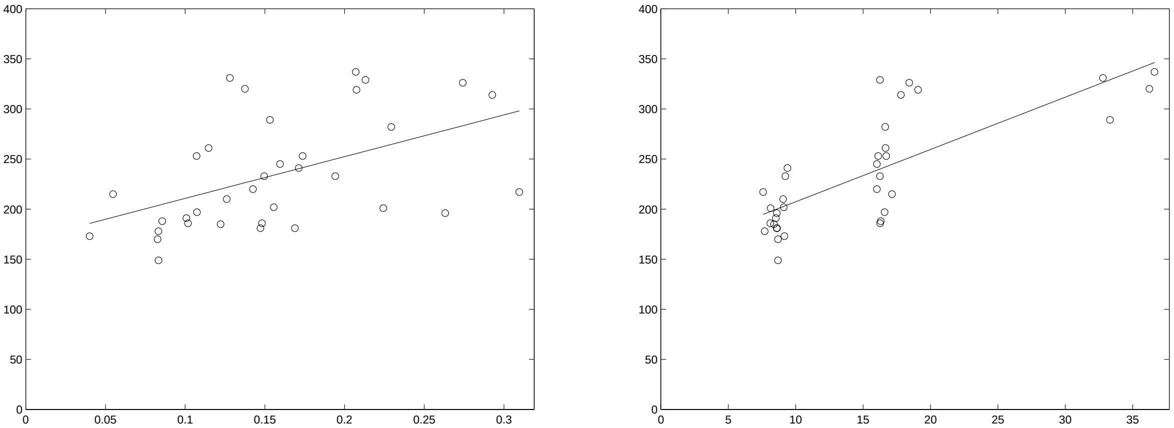

Figure 2: (a) MDTS plotted MDSR2; (b) MDTS plotted against against draw ratio, V_1/V_0.

Statistical investigation confirms what the Figures tell us. The linear and quadratic equations for MDTS are as follows:

$$\mathrm{MDTS} = 163 + 29.9\mathrm{MDSR} \qquad (2)$$

and

$$\mathrm{MDTS} = 135 + 53.7\mathrm{MDSR} - 3.49\mathrm{MDSR}^2 \qquad (3)$$

An analysis of variance for the regression line in Equation (2) gives $F_{1,33} = 126.0$ and there is a probability $p < 10^{-12}$ of a value exceeding this occurring by chance. Similarly an analysis of variance for Equation (3) gives $F_{2,32} = 82.1$, with the improvement obtained by including the quadratic term being significant; t=-2.95 corresponds to a probability of 0.6%. The dependence of MDTS on MDSR2 is not significant (as is probably obvious from Figure 2(a) while the dependence on draw ratio

$$\mathrm{MDTS} = 155 + 5.22(V_1/V_0) \qquad (4)$$

is highly significant. This may serve as a warning against a careless use of statistical tests. The “significance” referred to is saying that the mean square distance of the measured values of MDTS are from the regression line is significantly smaller than the variance of MDTS. However, equation (4) is not a useful predictor for our data, since the draw ratio has two main values, $V_1/V_0 \approx 8$ and $V_1/V_0 \approx 16$, each of which was used for around 15 runs. The line shown in Figure 2(b) does not (and cannot) help in predicting the variation among runs with the same draw ratio, while equations (2) and (3) do make a useful contribution to that prediction. We conclude that the relations proposed in Equations (2) and (3) are as good as we are likely to get if our aim is to predict MDTS from a single variable (or combination of variables).

The transverse direction tensile strength, TDTS, is less successfully predicted. In fact an attempt to use the corresponding variables, TDSR and TDSR2, as predictors gives nothing statistically significant. (The TD strain rates are defined as $\mathrm{TDSR} = (V_1/\mathrm{FLH})\ln(R_1/R_0)$ and $\mathrm{TDSR2} = (V_0/\mathrm{FLH})\ln(R_1/R_0)$.) If we exclude the runs where the bubble was sitting on the deflector, four of which are those with the highest TDSR, but a TDTS which is not appreciably improved , we do a little better. Figure 3 shows the two regression equations, (5) and (6),

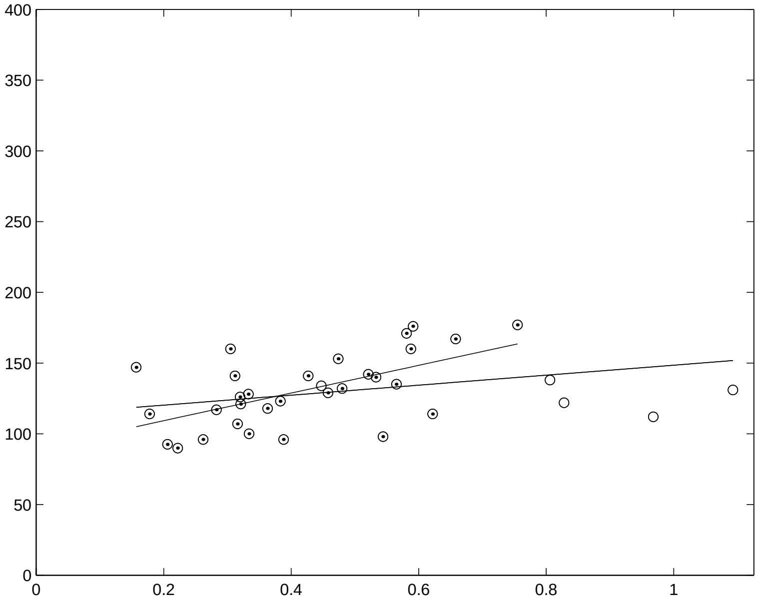

Figure 3: TDTS plotted against TDSR with fitted linear regression lines using 30 ($\odot$) and 35 (o) values.

$$\mathrm{TDTS} = 113 + 35.3\mathrm{TDSR} \tag{5}$$

which uses all 35 runs and

$$\mathrm{TDTS} = 89.7 + 97.8\mathrm{TDSR} \tag{6}$$

which uses the 30 runs which are not affected by the deflector (i.e. excluding runs 8, 13, 14, 20 and 38).

The regression equation (6) is statistically significant in a formal sense ($F_{1,28} = 14.67$; a value exceeding this has a probability of about 0.1% of happening by chance). In contrast, equation (5) is not statistically significant ($F_{1,33} = 3.86$; a value exceeding this has a probability of 6% of happening by chance). In fact the regression of TDTS on BUR,

$$\mathrm{TDTS} = 94.6 + 17.0\mathrm{BUR} \tag{7}$$

is also statistically significant but it is even more true here than for Equation (4) that we do not have a useful predictor for our data because we have only two values of BUR; the graph is similar to Figure 2(b).

4 Discussion

We might ask why the MDTS predictor, Equation (2), is better than the TDTS predictor, Equation (5). One important feature of the results is that the TDSR has a considerably smaller range of values than the MDSR. Because we are not claiming that either characteristic strain rate will be the sole parameter determining tensile strength, the smaller the range of values, the more likely that other factors, unaccounted for in our predictor, will contribute appreciably to the variation in the results. This makes it much less likely that TDSR will be a good predictor on the basis of the results we have obtained. In our results, the TDTS is generally about half the MDTS and the TDSR is often less than one tenth of the MDSR. If we sought a balanced film (in the simple sense of having equal MDTS and TDTS) we should, according to Equations (2) and (6), need

$$\mathrm{TDSR} = 0.75 + 0.3\mathrm{MDSR} \tag{8}$$

This would require TDSR values in the range 0.9 to 2.3 for MDSR in the range, typical of our observations, of 0.6 to 5. Use of Equations (3) and (6) does not seriously affect this conclusion. The observed TDSR values in fact lie in the range 0.2 to 0.8 which corresponds to the unbalanced film properties we find.

The use of a characteristic strain rate rather than a characteristic strain as a predictor of tensile strength may seem surprising. We have the idea that the MDTS will be determined by the polymer microstructure in the film and that will be influenced by the orientation and stress in the polymer as it solidifies. We might tend to think that near solidification, the polymer will behave in an elastic manner rather than a viscous manner.

It is perhaps misleading to label the regression equations, Equations (4) and (2) as elastic and viscous respectively. It is clear that, if we compare two runs with the same draw ratio (total MD strain) but different freeze-line heights, the orientation and stress at the freeze-line will be different because of relaxation effects, not to mention different thermal histories. It is not certain, *a priori*, which way the effect of a greater freeze-line height might tend, but it seems most plausible that it will allow greater relaxation and reduce the stress. One might say that the "frozen-in strain" would be only the strain accumulated in a, presumably small, time interval before the freeze-line and that, for a given draw ratio, the greater the freeze-line height and the smaller the freeze-line velocity, V_1, the smaller that "frozen-in strain". Suffice it to say that time scales are important and that the viscoelastic nature of the material is, in some sense, important in assessing the stress and microstructure in the film as it solidifies. The comparison of the various regression equations for the MDTS, discussed above, clearly bear this out.

An historic, and still relevant, study of the effect of processing on film properties was carried out by Clegg and Huck (1961) and their results are reported in detail by Holmes-Walker (1969), pp 84-86. MDTS, TDTS, impact strength and tear strength (MD and TD) are reported as functions of BUR and output (mass flow rate) in response surfaces which show the trends in each property. There is also a table showing the effect on these five mechanical properties of die gap, melt temperature, BUR, FLH and haul-off speed. We record some trends reported by Clegg and Huck in Table 4.

Glanvill (1971) records accepted wisdom, for the benefit of "all technical staff engaged in the plastics converting industry". His section on the effect of processing parameters on mechanical and optical properties consists of a number of "trend graphs" which seek to show "the effect of altering one variable only, other variables being held constant." As we have seen, the interrelation of variables does not make this easy to do (or indeed to specify unambiguously). The only trend graph for tensile strength which Glanvill includes shows TDTS decreasing, and MDTS decreasing even more rapidly,

Table 4: Trends in properties.

Property	Parameter varied	Reported trend			
		Glanvill	Clegg & Huck	this work	Tas
MDTS	DR	-	unaffected	increases	increases
MDTS	BUR	decreases	decreases	decreases	-
TDTS	DR	-	unaffected	unaffected	decreases
TDTS	BUR	decreases	increases	increases	-
MD tear	BUR	decreases	decreases	decreases	-
TD tear	BUR	decreases	increases	little effect	-
Impact	BUR	increases	increases	increases	-
Impact	FLH (high BUR)	increases	increases	unaffected	-
Impact	FLH (low BUR)	decreases	increases	unaffected	-
Impact	DR/BUR	-	-	decreases	decreases

with increasing BUR. We have seen, Equation (7), that TDTS increases with BUR which contradicts the reported trend. A regression of MDTS against BUR gives

$$\mathrm{MDTS} = 394 - 77.0\mathrm{BUR} \tag{9}$$

with $F_{1,33} = 83.84$, highly significant but again not a very useful predictor for our data (with only two values of BUR). These and other trends in mechanical properties are summarized in Table 4. We have not included many of the results of Tas (1994) here since Tas plots mechanical properties against draw ratio (or MD stress at the freeze-line) and not against BUR (which he does vary) or FLH (which he does not vary). Note that the films produced by Tas have different thicknesses, and it is claimed by Horsley (1969), p.56, talking about impact strength, that “it is difficult, however, to reduce results obtained at different thicknesses to a common basis”.

Further investigation of the MD to TD balance in mechanical properties is proposed, since the production of a “balanced” film is often a desirable aim in choosing operating conditions. In doing this we are also motivated by Tas’ presentation of impact strength as a function of the ratio DR/BUR. Our results for impact strength show the same trend, but it is clear that this ratio alone does not explain much of the variation we have in impact strength. A consideration of the ratio of measured MDTS/TDTS does not give any more useful results, largely because for our results the 35 runs involve only four different values of DR/BUR. The trends are as expected but it seems that other factors influence the balance of mechanical properties. These investigations are continuing.

Acknowledgements

The experimental work reported here was undertaken in collaboration with ICI and Visqueen Ltd and both the financial support (direct and indirect) and the permission to publish are gratefully acknowledged. The experiments were carried out by M.J. Vidler under the supervision of G.G. Turner at the Visqueen factory in Stevenage. The planning also involved contributions from G. Jay and B.W. Overton of Visqueen (then part of ICI) and P.L. Clegg, F.N. Cogswell and P. Lamb of ICI Plastics (as it then was). The contributions of all these people are gratefully acknowledged, as is a period of study leave from Newcastle University which allowed the work to be undertaken.

References

Clegg, P.L. and Huck, N.D., *The effect of extrusion variables on the fundamental properties of tubular polythene film*, Plastics, London, **26**, 114 (1961)

Debbaut, B., Goublomme, A., Homerin, O., Koopmans, R., Liebman, D., Meissner, J., Schroeter, B., Reckmann, B., Daponte, T., Verschaeren, P., Agassant, J.-F., Vergnes, B. and Venet, C., *Development of high quality LLDPE and optimised processing for film blowing*, Int. Polym. Proc., **13**, 262-270 (1998).

Glanvill, A.B., *Plastics Engineer's Data Book*, Machinery Publishing Co., Brighton (1971).

Holmes-Walker, W.A., *Extrusion and Calendering*, Chapter 3 in *Thermoplastics: Effects of Processing*, Ed. R.M. Ogorkiewicz, Iliffe Books (for the Plastics Institute), London, 76-109 (1969).

Horsley, R.A., *Assessment of performance*, Chapter 2 in *Thermoplastics: Effects of Processing*, Ed. R.M. Ogorkiewicz, Iliffe Books (for the Plastics Institute), London, 31-75 (1969).

Kurtz, S.J., *Some key challenges in polymer processing technology*, in *Recent Advances in Non-Newtonian Flows*, ASME, **AMD-Vol 153/PED-Vol 141**, 1-13 (1992).

Kurtz, S.J., *Relationship of stresses in blown-film processes*, Int. Polym. Proc., **10**, 148-154 (1995).

Liu, C.-C., Bogue, D.C. and Spruiell, J.E., *Tubular film blowing. Part 1. On-line experimental studies*, Int. Polym. Proc., **10**, 226-229 (1995a).

Liu, C.-C., Bogue, D.C. and Spruiell, J.E., *Tubular film blowing. Part 2. Theoretical modelling*, Int. Polym. Proc., **10**, 230-236 (1995b).

Pearson, J.R.A. and Petrie, C.J.S., *The flow of a tubular film. Part 1. Formal mathematical representation*, J. Fluid Mech., **40**, 1-9 (1970a).

Pearson, J.R.A. and Petrie, C.J.S., *The flow of a tubular film. Part 2. Interpretation of the model and discussion of solutions*, J. Fluid Mech., **42**, 609-625 (1970b).

Pearson, J.R.A. and Petrie, C.J.S., *A fluid-mechanical analysis of the film-blowing process*, Plastics & Polymers, **38**, 85-94 (1970c).

Petrie, C.J.S., *An investigation into the effect of output rate and scale-up on blown film properties*, Internal report, ICI/Visqueen Ltd (1975a).

Petrie, C.J.S., *A comparison of theoretical predictions with published experimental measurements on the blown film process*, AIChE J., **21**, 275-282 (1975b).

Petrie, C.J.S., *Elongational flows*, Pitman, London (1979).

Petrie, C.J.S., *Film blowing, blow moulding and thermoforming*, Chapter 7 in *Computational Analysis of Polymer Processing*, Eds. J.R.A. Pearson and S.M. Richardson, Applied Science Publishers, London, 217-241 (1983).

Petrie, C.J.S., *Recent ideas in extensional rheology*, in *Polymer Processing Society Europe/Africa Regional Meeting - Extended Abstracts*, Ed. J. Becker, Chalmers University of Technology, Gothenburg, Sweden, KN4:2 (1997)

Tas, P.P., *Film blowing: from polymer to product*, Doctoral thesis, Technical University of Eindhoven (1994).