

A Design of Experiments Study on the Factors Affecting Variability in the Melt Index Measurement

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ABSTRACT: Melt index (MI) is universally accepted as the primary specification in the thermoplastics industry. The standard according to which the MI measurement is made is ASTM D-1238. However, some variation in the MI measurement is possible, even when all ASTM D-1238 specifications are met. A thorough understanding of the sources and magnitude of variation is essential, since measurement error can have significant economic implications. Error in the MI measurement can cause a resin lot (about 190,000 pounds) to be erroneously classified as off-spec (leading to a loss of over US \$20,000). A Resolution IV, six-factor, $\frac{1}{4}$ fractional factorial design of experiments (DOE) study [with three replicates] was done to quantify the effect of various factors affecting variability in the MI measurement using two high-density polyethylene monitor resins with MI of 1.91 g/10 min ("low" MI) and 36.4 g/10 min ("high" MI). A major challenge was dealing with qualitative (e.g., "cleanliness") and quantitative (e.g., "sample mass" or "temperature") factors in the same DOE. For the high MI monitor, among the factors considered, the most significant (for the ranges considered) were found to be (in the order of importance): die orifice diameter, temperature, die cleanliness, barrel cleanliness, and sample mass. The following factors were also considered but not found (in the ranges considered) to be as significant: piston land (tip) diameter, load, piston cleanliness, preheat time, and piston curvature. For the low MI monitor, among the factors considered, the two most significant (for the ranges considered) were temperature and barrel cleanliness. This article gives practical and useful information for those who make and/or use MI measurements. As others have pointed out, much of such discussion related to melt indexers is also applicable to more sophisticated capillary rheometers. © 1997 John Wiley & Sons, Inc. *J Appl Polym Sci* **65**: 277–288, 1997

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INTRODUCTION

Melt index (MI) is universally accepted as the primary specification in the thermoplastics industry and is the basis on which more than 130 billion pounds of thermoplastic resin (with a dollar value exceeding US \$50 billion) was sold worldwide in 1994.¹ (Worldwide plastics consumption is anticipated to rise by an average annual growth rate of

about 4% for the period 1994–2000). It is extremely important that the MI measurement be made as accurately and precisely as possible, because the economic consequences of MI measurement error can be significant. Measurement error can cause a lot of resin (about 190,000 pounds) to be erroneously classified as "off-spec," and the difference between a lot being in-spec and out-of-spec can be more than US \$20,000. Our best estimate is that between four and eight MI tests are run for each lot of resin produced (including in-lines, repeats/replicates, rechecks by cus-

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tomers, etc.). This corresponds to between 7,500 and 15,000 MI tests being run round the world everyday! Obviously, it is desirable to know as much as possible about the causes and magnitude of variability for a test that is run so frequently.

The standard that specifies how the MI measurement should be made is ASTM D-1238.² However, some variation is possible even when all specifications of this ASTM standard are met. For example, the ASTM standard does not specify an exact sample mass to be used for making the MI measurement; it merely "suggests" a range within which the sample mass should fall. As is shown below, the amount of sample mass used to make the measurement can affect the numerical value of MI obtained. ASTM D-1238 states that it is technically equivalent to ISO-1133³ (although there are minor differences in some of the permissible tolerances; e.g., for MI values in the range between 3.5 and 10 g/10 min, ASTM D-1238 [manual Procedure A] suggests a sample mass of 4–8 g and a measurement time interval of 60 sec, while ISO-1133 specifies 6–8 grams and 30 sec, respectively). ASTM D-1238 is far more detailed, is more widely used, has been in use for a much longer time, and has been validated by several round-robin studies; hence, ASTM D-1238 is the primary standard referenced here for the MI measurement.

The MI is not a fundamental rheological property, since the value encompasses shear and entrance pressure components (the die orifice length to diameter ratio is only 8 mm/2.0955 mm, or 3.82).^{4,5} The MI also does not correlate in any simple manner with processibility, since the measurement is made at a relatively low shear rate (while commercial plastic processing typically occurs at much higher shear rates). Such issues are discussed elsewhere,^{4–6} and hence, that discussion is not repeated here.

It is not the purpose of this article to critique the validity of MI as an appropriate measure for the quality control of plastics or to discuss the interpretation of MI. Instead, this article concentrates only on identifying and quantifying the sources of variability in making the MI measurement. As Reilly and Appel⁴ point out, much of such discussion related to melt indexers is also applicable to more sophisticated capillary rheometers. ASTM D-1238 contains results from round-robin studies, which give information on within-lab and between-lab variability. However, Reference 4 is the only reference in the published literature that the authors have been able to find that discusses

issues related to the subject of making precise MI measurements. While this reference contains useful information and helpful suggestions for making more precise MI measurements, the information is rather anecdotal. A rigorous, thorough study using a design of experiments (DOE) (or similar approach) is not presented. Saucier and Obermiller⁷ have published a thorough DOE study on dynamic oscillatory measurements to determine the linear viscoelastic properties of polymeric melts using a controlled strain rheometer. It is perhaps ironic that such a study should have first appeared for a more complex rheological measurement than for the simple MI!

Typically a lot (roughly 190,000 pounds) of resin is characterized by a single MI value. It is important to recognize, though, that the MI measurement actually tests only a very small fraction of the lot. ASTM D-1238 suggests that a sample mass between 2.5 and 8 g be used (depending on the flow range value). However, the amount of material extruded while the measurement is actually being made is substantially smaller, about 0.35 g for a quarter-inch piston travel (MI < 10 g/10 min) and about 1.39 g for an inch piston travel (MI > 10 g/10 min). This is indeed a unique situation in that such a tiny amount decides whether a 190,000-pound lot is "in-spec" or "out-of-spec" (obviously, at times, multiple measurements and/or multiple samples [e.g., for different rail-car compartments] are used). Essentially, this ratio works out to be equivalent to a measurement on one specimen deciding the fate of 62 million (for MI > 10 g/10 min) or 248 million (for MI < 10 g/10 min) specimens! However, the issue of lot homogeneity (or how representative the sample used in making the MI measurement is of the rest of the lot) is a separate issue and is not dealt with here.

MATERIALS AND EQUIPMENT

Two high-density polyethylene (HDPE) monitor resins 95A ("low" MI) and 92D ("high" MI) were specially prepared and well blended to make them as homogeneous as possible. Monitor resin properties are shown in Table I. These monitor resins are used to maintain statistical control of melt indexers by various Solvay sites worldwide. The averages of a very large number of MI measurements made with these monitors on multiple indexers by multiple operators at multiple sites over a period of several years have been determined to

Table I Monitor Resin Data

Resin Property	“High” MI Monitor HDPE-92D	“Low” MI Monitor HDPE-95A
M_n (Da)	11,900	18,100
M_w (Da)	38,000	91,300
M_z (Da)	84,600	335,900
Polydispersity (M_w/M_n ratio)	3.19	5.04
MI average (g/10 min)	36.4	1.91
MI standard deviation (g/10 min)	0.4	0.03
Power-law index, n (slope of shear-thinning viscosity curve)	0.9	0.7

be 1.91 and 36.4 g/10 min, while the corresponding standard deviations are 0.03 and 0.4 g/10 min, respectively. The application of statistical process control to rheological measurements is discussed elsewhere.^{8,9} The apparent viscosity versus apparent shear-rate curves for both of the monitors measured by capillary rheometry at 190°C with the MI die are shown in Figure 1. For HDPE, the apparent shear rate (in reciprocal seconds) in the melt indexer is given by approximately 2.42 multiplied by the MI value. These rates are marked by an arrow on Figure 1 for both monitors.

The melt indexer used for this study was a Tinius Olsen extrusion plastometer model UE-4-78

controller (style) Y3051 meeting all of the specifications of ASTM D-1238. Prior to use, the temperature was calibrated to NIST-traceable standards and all equipment dimensions were verified to meet ASTM D-1238 specifications. The high MI measurements were made with automated Procedure B with a 1-inch travel (the indexer is equipped with an automated timer capable of measuring up to 0.01 sec). The low MI measurements were made by manual Procedure A with a 3-min cut. The data collected were analyzed with DOE WISDOM software (*W*indows *S*oftware for *D*esign *O*f *E*xperiment *M*ethods; Launsby Consulting, Colorado Springs, CO)^{10,11} and MINITAB

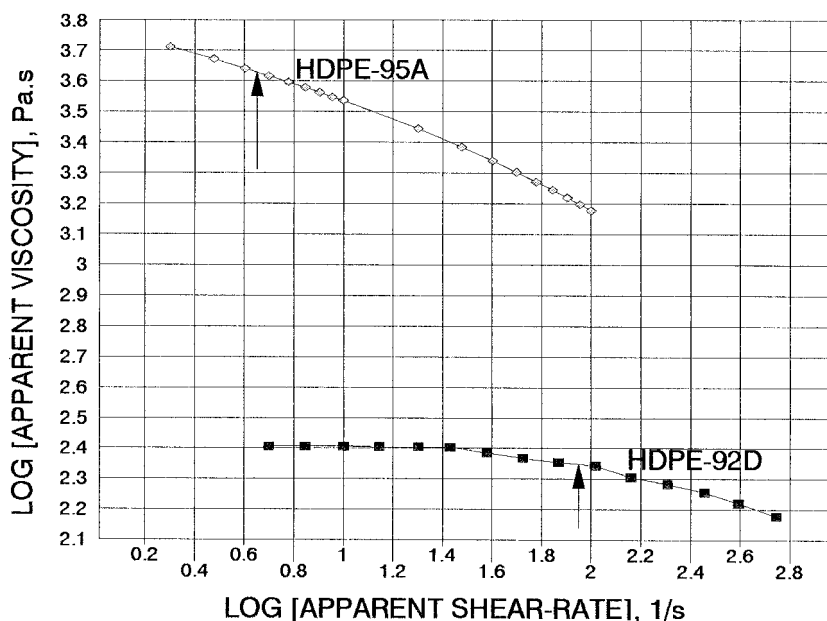


Figure 1 The shear-thinning behavior of HDPE-92D and HDPE-95A measured by capillary rheometry at 190°C with the MI die (the arrows mark the apparent shear rate in the melt indexer).

software. Molecular weights were measured by size exclusion chromatography at 145°C with 1,2,4-trichlorobenzene as a solvent with a Millipore Waters 150-C GPC.

EXPERIMENTAL DESIGN

Experimental design is a statistically based arrangement according to which an experimental program is to be conducted and the selection of the levels of one or more factors or factor combinations to be included in the experiment.¹² The concepts of experimental design are discussed in various references^{10–15}; basically, the two major advantages of using a statistically based design over varying one factor at a time is that interaction effects can be analyzed and precision information can be more easily obtained with fewer experiments (and hence, at lower cost).

A factorial design is one in which several factors are investigated at two or more levels and the interaction between factors may be important.¹⁵ This design consists of running all possible combinations of factors and levels. However, if the number of factors is large, this may increase the number of tests beyond that economically feasible with given resource constraints (especially if the design is replicated to provide more reliable precision information). In such a case, a fractional factorial (a “fraction” of the full factorial design) may be appropriate, where several factors are investigated at several levels but only a subset of the full factorial is run. The fraction is a carefully prescribed subset (on a sound statistical basis) of all possible combinations. A Resolution IV, six-factor, $\frac{1}{4}$ fractional factorial at two levels (high or “+1” level and low or “–1” level) was used in this study.

A major challenge in this study was dealing with qualitative (e.g., “cleanliness”) and quantitative (e.g., “sample mass” or “temperature”) factors in the same DOE. The factors considered in the design for the MI measurement are summarized in Table II and are discussed below.

Die Orifice Diameter

ASTM D-1238 mandates that the orifice diameter must be 2.0955 ± 0.0051 mm. Hence, ideally, it would be desirable to use the top of the spec (a die with orifice diameter = 2.1006 mm) as the “+1” level and the bottom of the spec (a die with

orifice diameter = 2.0904 mm) as the “–1” level. However, dies with such exact diameters were not available for this study; dies with diameters of 2.1448 and 2.0930 mm were used as the “+1” and “–1” levels.

Sample Mass

ASTM D-1238 does not mandatorily require any specific sample mass but merely “suggests” a range of 4.0–8.0 g for samples with flow rates greater than 25 g/10 min and a range of 3.0–5.0 g for samples with flow rates between 1.0 and 3.5 g/10 min. Therefore, for the high MI monitor (HDPE-95A), an 8.0-g sample was used for the “+1” level and a 4.0-g sample was used for the “–1” level. Similarly, for the low MI monitor (HDPE-92D), a 5.0-g sample was used for the “+1” level and a 3.0-g sample was used for the “–1” level.

Temperature

ASTM D-1238 requires that the standard temperature for polyethylene be $190.0 \pm 0.2^\circ\text{C}$, measured in the melt at a location 10 mm above the upper surface of the die. In addition, the temperature of the barrel from 10 to 75 mm above the top of the die must be maintained within $\pm 1\%$ of the set temperature (190.0°C). On the basis of these specifications, 191.9°C (1% higher than the required temperature) and 188.1°C (1% lower than the required temperature) were used as the “+1” and “–1” levels.

Die Cleanliness

Note 14 of ASTM D-1238² states “the degree of cleanliness can significantly influence the flow rate results.” Hence, for this study, it was necessary to quantify “cleanliness” in a reproducible manner. Such a definition of “cleanliness” is necessarily arbitrary. The “dirty” die (or “–1” level) is defined as the die through which the monitor *not* being tested (PE-95A for the high monitor test and PE-92D for the low monitor test) has been purged and then the die is not cleaned before use in the next test. The “clean” die (or “+1” level) is defined as a die that had been cleaned when hot by the removal of any molten plastic sticking to it with a clean metal plug, wiped carefully by a clean cloth rag, and then heated for 4 min in an oven at 500°C . After this cleaning, the die is

Table II Factors Considered and Their Levels

High MI Factor	Low MI Factor	Factor	“-1” Level	+1” Level
A		Die orifice diameter	2.0930 mm	2.1448 mm
B	B	Sample mass (low/high monitor)	3.0/4.0 g	5.0/8.0 g
C	A	Temperature	188.1°C	191.9°C
D	D	Die cleanliness	Dirty or purged with monitor <i>not</i> being tested	Cleaned in oven
E		Piston land (tip) diameter	9.462 mm	9.500 mm
F	F	Barrel cleanliness	Dirty or purged with monitor <i>not</i> being tested	Cleaned by swabs
	C	Purge time	Immediately after charging	5 min after charging
	E	Reference mark	Piston tip 48 mm above top of die	Piston tip 44 mm above top of die
# ^a		Load	2149.2 g	2170.8 g
# ^a		Preheat time	6 min	8 min
# ^a		Piston cleanliness	Dirty or purged with monitor <i>not</i> being tested	Cleaned with polish
# ^a		Piston curvature	3.05 mm	0.32 mm

#^a Factors eliminated by initial screening of high MI monitor.

checked to ensure that it successfully passes the go/no-go gauge test.

Piston Land (Tip) Diameter

ASTM D-1238 specifies that the piston land (tip) diameter be 9.4742 ± 0.0076 mm. Hence, ideally, the “+1” level would have represented a diameter of 9.4818 mm (top of the spec) and a “-1” level would have represented a diameter of 9.4666 mm (bottom of the spec). However, as in the case of die orifice diameter, piston lands with such precise diameters were not available for this study. Instead, the “+1” and “-1” levels were lands with diameters of 9.500 and 9.462 mm, respectively.

Barrel Cleanliness

Note 14 of ASTM D-1238² reports that “three swabs of the barrel” are satisfactory for cleaning and that the barrel is more easily cleaned when hot. The approach used to define barrel cleanliness is similar to that used for defining die cleanliness. The “dirty” barrel (or “-1” level) was a barrel that was purged with the monitor *not* being tested (PE-95A for the high monitor test and PE-92D for the low monitor test) and then used with-

out any further cleaning before the next test began. The “clean” barrel (or “+1” level) was a barrel that was cleaned by swabbing the hot barrel repeatedly with three cotton gun-patches before each test (the gun-patches were discarded after each swab, and new ones were used for the subsequent swab).

Load

ASTM D-1238 specifies that the combined weight of the piston and load must be within a tolerance of $\pm 0.5\%$ of the selected load (2160.0 g); hence, the “+1” and “-1” levels were chosen at 0.5% excess and 0.5% deficit of 2160.0 g.

Preheat Time

ASTM D-1238 specifies a preheat time of 7.0 ± 0.5 min. However, since we had customarily used a 6-min preheat time in the past, we chose the “+1” and “-1” levels at 8 and 6 min, respectively.

Piston Cleanliness

The strategy used for defining piston cleanliness is similar to that used for defining piston and die

cleanliness above. A “dirty” or “-1” level piston is defined as a piston used to purge a barrelful of the monitor *not* being tested and then the piston is used “as is” without any further cleaning. The “+1” level is defined as a piston which is wiped when hot with clean cloth rags until the piston is completely clean without any plastic sticking to it, and then, the piston is polished with a metal polish (we used NOXON®) (Boyle-Midway Household Products, Inc., New York) until it is shiny.

Piston Curvature

To quantify the curvature, the piston was laid flat horizontally and supported on two rollers at a distance of 59 and 142 mm from its tip. The piston was then rotated on its axis, and the difference between the maximum and minimum vertical displacements of the tip (noted in Table II) was measured with a micrometer dial.

Purge Time

ASTM D 1238² requires that the purge be completed at least 2 min prior to the start of the test for materials having MI < 10 g/10 min. The other bound on the purge time is the ASTM D-1238² specification that the total preheat time be restricted to 7.0 ± 0.5 min. Hence, the “-1” and “+1” levels for purge time were set at 7 and 2 min prior to the start of the test (or, in other words, 0 and 5 min after charging the sample).

Reference Mark

The piston has upper and lower reference marks 4 mm apart. ASTM D-1238² requires that, at the start of the test, the piston must be between 48 and 44 mm above the top of the die. The piston is 48 mm above the top of the die when the lower mark on the piston coincides with the top of the cylinder or other suitable reference point (in our case, the reference point was the top of the guiding ring for the piston). This was taken to be the “-1” level. The “+1” level was when the upper mark on the piston coincides with the top of the guiding ring (or the piston is 44 mm above the top of the die).

RESULTS AND DISCUSSION

Initial Screening

Initial screening designs were run on the high MI monitor (HDPE-92D) to reduce the number of

factors to a manageable level. These initial designs (not discussed here in the interests of brevity) suggested that, for the high MI monitor, the following factors were not as significant (in the ranges discussed above) as the others: load, preheat time, piston cleanliness, and piston curvature.

From the power-law equivalent of the Hagen-Poiseuille equation,¹⁶ it is expected that the MI will scale with the load raised to the power $1/n$, where n is the power-law index. Even though the power-law equivalent of the Hagen-Poiseuille equation is not strictly applicable (because the flow in the melt indexer die is neither fully developed nor at steady state), it can still be considered to provide a reasonable first-order estimate. However, the variation in the load considered here ($\pm 0.5\%$) is too small to cause a significant effect. Clearly, a larger variation in load would cause a more substantial effect in the MI. Hence, if one keeps within the ASTM specification of $\pm 0.5\%$ load, then one can be reasonably sure that one potential cause of variability has been removed.

The results of this study indicated that, for the high MI monitor, the piston curvature (as defined above) of 3.05 mm did not cause a significant effect in the MI. This is contrary to the finding of Reilly and Appel,⁴ where a 0.25-mm deflection was found to have a significant effect. This could possibly be due to the fact that Reilly and Appel⁴ used a lower MI resin (7.61 g/10 min) and may have measured piston deflection in a different manner than we did. Reilly and Appel⁴ also found no interaction effect between load and rod curvature.

The fact that a variation of preheat time from 6 to 8 min did not cause a significant effect can be attributed to the fact that the resin is well stabilized; hence, chemical change (degradation/crosslinking) differences between 6 and 8 min of preheat are probably insignificant. The MI (average, 36.4 g/10 min) is also rather high, indicating relatively low molecular weights or short relaxation times. The fact that piston cleanliness was not found to be significant in this study is simply an artifact of the manner in which piston “cleanliness” is defined and must not be interpreted to imply that a thorough cleaning of the piston is not important. “Dirtying” the piston in some other manner than that described above could easily cause a significant effect on the MI.

Since it is inconvenient to vary the temperature (ASTM D-1238² requires that the indexer be stable at the appropriate test temperature for 15 min

Table III High MI (HDPE-92D) Final Design and Data

Run	A	B	C	D	E	F	MI (g/10 min)—First Replicate	MI (g/10 min)—Second Replicate	MI (g/10 min)—Third Replicate
1	-1	-1	+1	+1	+1	-1	36.6	36.8	37.6
2	+1	+1	-1	-1	-1	+1	38.9	39.5	39.6
3	-1	-1	-1	-1	-1	-1	31.9	32.3	31.9
4	+1	+1	+1	+1	+1	+1	42.5	41.9	41.6
5	+1	+1	-1	+1	-1	-1	38.6	38.1	38.8
6	+1	+1	+1	-1	+1	-1	40.0	39.8	40.5
7	-1	-1	+1	-1	+1	+1	34.8	35.5	34.8
8	-1	+1	-1	+1	+1	-1	35.8	34.7	34.6
9	+1	-1	-1	-1	+1	-1	36.6	37.4	37.0
10	-1	-1	-1	+1	-1	+1	35.2	34.6	35.5
11	-1	+1	+1	+1	-1	+1	37.8	37.9	37.2
12	+1	-1	+1	-1	-1	+1	40.7	40.9	39.0
13	+1	-1	+1	+1	-1	-1	41.1	40.9	41.2
14	-1	+1	-1	-1	+1	+1	36.0	36.0	35.1
15	-1	+1	+1	-1	-1	-1	36.6	35.4	35.3
16	+1	-1	-1	+1	+1	+1	38.9	39.4	39.5

before testing is begun), this factor was “blocked” for the low MI test. For a discussion on “blocking” factors, refer to Reference 10. It was verified that “blocking” temperature (as opposed to “randomizing” it) did not affect the results.

CONCLUSIONS

The final design (shown in Tables III and IV with data for the high and low monitors, respectively)

was a Resolution IV, six-factor (A to F in Table II), $\frac{1}{4}$ fractional factorial design. It was replicated three times to obtain a good estimate of precision. The main effects and two-factor interactions are clear of each other, but two-factor interactions are aliased with each other in a Resolution IV design. The two- and three-factor aliasing is shown in Table V (for a discussion on aliasing, refer to Reference 10). It is generally reasonable to assume that main effects dominate and that the three-factor interaction (if any) aliased with the main

Table IV Low MI (HDPE-95A) Final Design and Data

Run	A	B	C	D	E	F	MI (g/10 min)—First Replicate	MI (g/10 min)—Second Replicate	MI (g/10 min)—Third Replicate
1	-1	-1	-1	-1	-1	-1	2.01	2.00	1.95
2	-1	-1	-1	+1	-1	+1	1.92	1.85	1.87
3	-1	-1	+1	-1	+1	+1	1.86	1.88	1.87
4	-1	-1	+1	+1	+1	-1	1.92	1.91	1.92
5	-1	+1	-1	-1	+1	+1	1.86	1.90	1.87
6	-1	+1	-1	+1	+1	-1	1.97	1.96	2.00
7	-1	+1	+1	-1	-1	-1	1.95	1.94	1.99
8	-1	+1	+1	+1	-1	+1	1.84	1.89	1.86
9	+1	-1	-1	-1	+1	-1	2.18	2.19	2.24
10	+1	-1	-1	+1	+1	+1	1.98	1.98	2.00
11	+1	-1	+1	-1	-1	+1	1.94	1.93	1.96
12	+1	-1	+1	+1	-1	-1	2.00	2.00	2.04
13	+1	+1	-1	-1	-1	+1	1.99	1.98	1.98
14	+1	+1	-1	+1	-1	-1	2.04	1.98	2.03
15	+1	+1	+1	-1	+1	-1	2.05	2.10	2.05
16	+1	+1	+1	+1	+1	+1	1.97	1.97	1.98

Table V Two- and Three-Factor Aliasing for Final Design

Interaction in This Column is Aliased With Interactions or Factors on the Same Row in This Column
A*B	C*E
A*C	B*E
A*D	E*F
A*E	B*C and D*F
A*F	D*E
B*D	C*F
B*F	C*D
A*B*C	E
B*C*D	F

effect is negligible in comparison. Hence, from the design used in this study, valid conclusions can be drawn separately about each individual main effect but no valid conclusions can be drawn separately about each individual two-factor interaction (conclusions can be drawn only about the group of two-factor interactions that are aliased with each other).

The results are summarized in Tables VI and VII for the high and low monitors, respectively, with factors and interactions being listed in the order of importance (and are depicted pictorially by Pareto charts in Figs. 2 and 3). The control limits (95% confidence level) for the coefficients determined from the DOE data are 0.21 and 0.009 for the high and low monitors, respectively. The coefficients of the prediction equation give an estimate of the effect varying a certain factor has on MI. For example, varying the die orifice diameter

between the two extremes, as listed in Table II (from 2.0930 to 2.1448 mm), leads to a variation in MI of 4.3 g/10 min (twice the coefficient for die orifice diameter listed in Table VI). The effect of varying the factors between the high and low levels for the high MI monitor (HDPE-92D) is shown pictorially in Figure 4. The corresponding information for the low MI monitor (HDPE-95A) is shown in Figure 5. The results of the various factors are discussed below.

Die Orifice Diameter

Table VI and Figure 6 show that, for the high MI monitor, variation in the die orifice diameter is the most significant source of error (causing, in the high MI test, almost two-thirds of the variation for the range of die orifice diameters considered). This fact is not surprising at all. The power-

Table VI Results of Six-Factor, 1/4 Fractional Factorial DOE Study on High MI Monitor (HDPE-92D)

Factor or Interaction	% Variation Due to Factor	Coefficient	Factor Statistically Significant (S) or Insignificant (I)
A = Die orifice diameter	64.46	2.14	S
C = Temperature	15.77	1.06	S
D = Die cleanliness	6.14	0.66	S
F = Barrel cleanliness	3.28	0.48	S
B = Sample mass	2.99	0.46	S
B*D + C*F	1.90	-0.37	S
A*E + B*C + D*F	0.50	-0.19	I
B*F + C*D	0.41	0.17	I
A*B + C*E	0.36	-0.16	I
A*D + E*F	0.21	-0.12	I
A*C + B*E	0.15	0.10	I
E = Piston land diameter	0.13	0.10	I
A*F + D*E	0.01	0.03	I

Table VII Results of Six-Factor, 1/4 Fractional Factorial DOE Study on Low MI Monitor (HDPE-95A)

Factor or Interaction	% Variation Due to Factor	Coefficient	Factor Statistically Significant (S) or Insignificant (I)
A = Temperature	38.44	0.054	S
F = Barrel cleanliness	30.52	-0.048	S
A*E + B*C + D*F	5.48	0.020	S
C = Purge time	4.82	-0.019	S
D = Die cleanliness	3.63	-0.016	S
E = Reference mark	2.61	0.014	S
B = Sample mass	0.37	-0.005	I

law version of the Hagen-Poiseuille equation¹⁶ shows that the steady-state flow rate will vary as the die orifice diameter is raised to the power $(3 + 1/n)$, where n is the power-law index. As stated earlier, even though the power-law version of the Hagen-Poiseuille equation is not strictly applicable to the flow in the melt indexer, it can still be considered to provide a reasonable first-order estimate. Typically, n for commercial HDPE resins lies roughly between 0.2 and 0.9: $n = 0.9$ for HDPE-92D and $n = 0.7$ for HDPE-95A (refer to Fig. 1); hence, a 1% change in die orifice diameter can be expected to cause about a 4% change in the MI for these monitors. The larger die orifice diameter (2.1448 mm) is about 2.4% bigger than the smaller die orifice diameter (2.0930 mm). From this study, we find that the corresponding MI is 11% higher (39.7 versus 35.4 g/10 min), very close to the 10% increase predicted by the

power-law version of the Hagen-Poiseuille equation. Note that since this factor (die orifice diameter) has such an overwhelming effect, it was not considered in the low MI test (since it would obviously have been predominant in that case also).

Temperature

Temperature was found to be the second most significant factor. Hence, the ASTM recommendation of restricting temperature variation to between $\pm 0.2^\circ\text{C}$ is well justified. Assuming that HDPE follows an Arrhenius temperature dependence with an activation energy of 27 kJ/mol,⁸ a 1% change in temperature at 190°C is expected to cause a shift of 2.9% in MI. In this study, for the high MI test, we measure a shift of 2.9% (corresponding to the temperature coefficient of 1.06 given in Table V). For the low MI test, we mea-

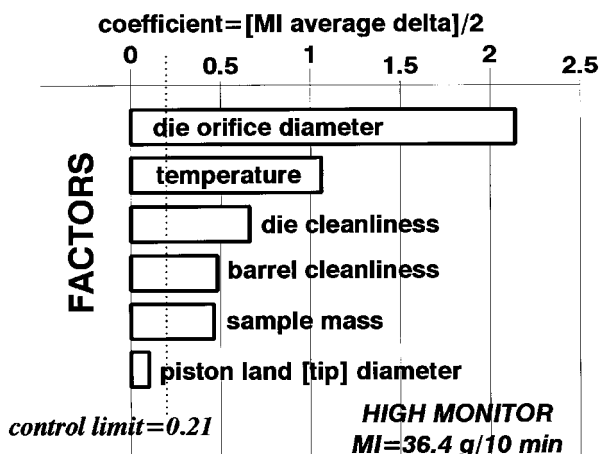


Figure 2 A Pareto chart of the coefficients for the high MI monitor (HDPE-92D) DOE test (control limit = 0.21 is shown by the dotted line).

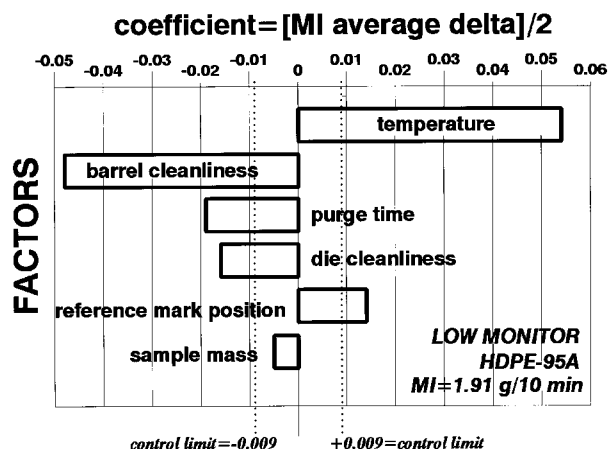


Figure 3 A Pareto chart of the coefficients for the low MI monitor (HDPE-95A) DOE test (control limits of ± 0.009 are shown by the dotted lines).

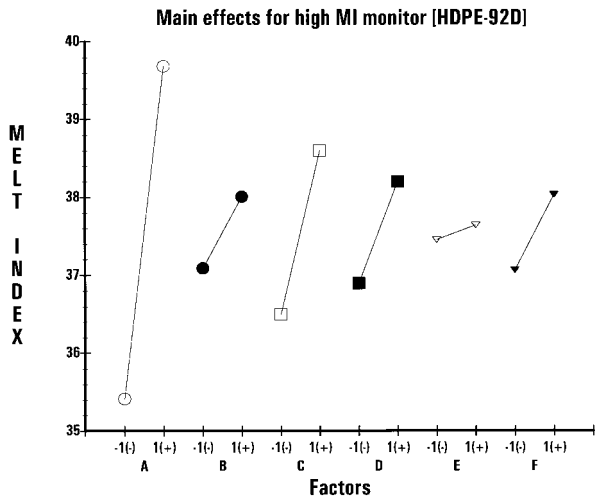


Figure 4 The effect of varying factors on the measured MI for the high MI monitor (HDPE-92D).

sure a shift of 2.8% (corresponding to the temperature coefficient of 0.054 given in Table VI). Note that Reilly and Appel⁴ report that having either one or two heater bands did not cause any significant change in MI as long as the ASTM specification of $\pm 0.2^\circ\text{C}$ within the die region is met. They also found that different temperature gradients in the barrel did not cause a significant effect on the material they studied ($\text{MI} = 21 \text{ g}/10 \text{ min}$), although they emphasize that this should not be interpreted to mean that precise temperature control is not critical. In this study, we did not study the effect of number of heater bands (the Tinius-Olsen indexer used has only single zone heater

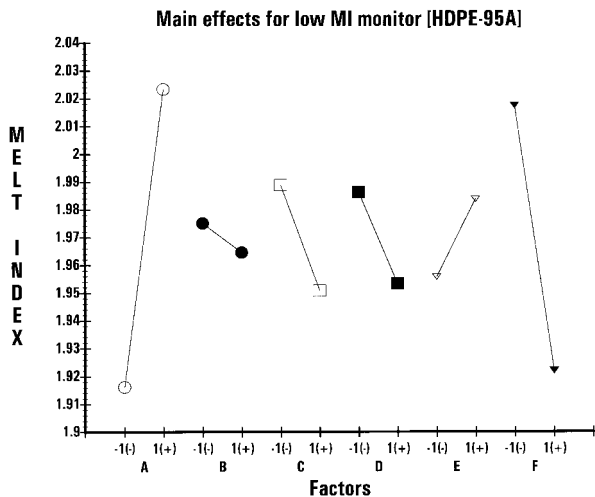


Figure 5 The effect of varying factors on the measured MI for the low MI monitor (HDPE-95A).

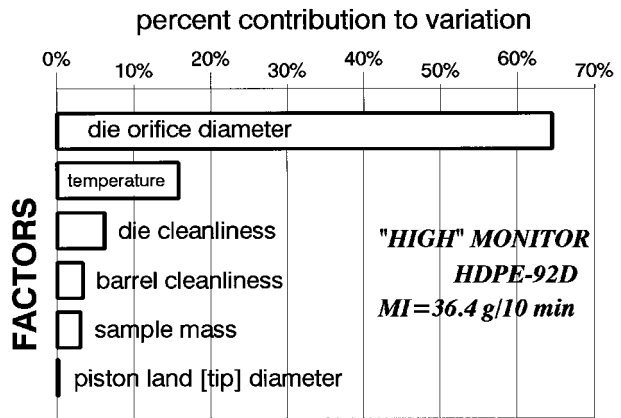


Figure 6 The relative contribution (in percentage terms) of the various factors to the variability in the MI measurement for the high MI monitor (HDPE-92D).

control) or the effect of different temperature gradients. To give an idea of temperature uniformity within the barrel, the temperatures measured in the melt 10.0, 12.7, 35.0, and 75.0 mm above the top of the die were 190.10, 190.08, 190.07, and 189.52°C, respectively.

Die and Barrel Cleanliness

The need for through cleaning is emphasized in References 2 and 4. For the high MI monitor, die cleanliness is found to be more important than barrel cleanliness. The converse is observed for the low MI monitor. Also, the coefficients for barrel and die cleanliness are positive for the high MI monitor but negative for the low MI monitor (Figs. 2 and 3, respectively). One may speculate on the reasons for this (in terms of slip, which may be induced in the low MI monitor by residual high MI monitor or the difficulty that a high MI [low viscosity] monitor may have in pushing out low MI [high viscosity] residues, etc.). It would be desirable to study the surfaces of "clean" and "dirty" barrel and die orifice surfaces by analytical surface chemistry techniques but that was not possible in this study.

Sample Mass

Sample mass was significant for the high MI monitor but not for the low MI monitor. This is consistent with the fact that the range for the sample mass permitted by ASTM D-1238² (and thus used in this study) for the high MI monitor (4–8 g) is twice that for the low MI monitor (3–5 g). The

results in Table VI suggest that it may be desirable to tighten the rather generous range (4–8 g) permitted by ASTM D-1238 for MI > 25 g/10 min. It is desirable to select a specific sample weight for each MI range and stick as close to that as possible for all tests in that MI range. Reilly and Appel⁴ recommend a maximum permissible tolerance of ± 0.1 g. They also mention that if weighing the sample to this tolerance is an impossible burden, then the use of a scoop, beaker, etc., to at least obtain a constant volume of sample for every test is desirable.

Piston Land [Tip] Diameter

Piston land (tip) diameter was not found to be significant, even though the ranges we used were outside the specifications permitted by ASTM D-1238. It seems safe to conclude that as long as one is within the ASTM specification, piston land diameter will not be an issue.

Purge Time

Purge time was found to be, after temperature and barrel cleanliness, the third most significant factor for the low MI monitor. Although we did not study purge time as a factor in the high MI test reported in this article, our historical experience has been that for high MI samples (especially polypropylene), a higher purge time gives more consistent results (probably because entrapped air or volatiles have a chance to escape before the full load is imposed).¹⁷ Hence, we recommend purging for 5 min after loading the sample (a longer purge time is not permitted by ASTM D-1238²).

Reference Mark Position

Of all of the factors found statistically significant for the low MI monitor, this was the last one. ASTM permits a range of 4 mm in the reference mark position; however, for the sake of consistency, it is recommended that the test always be started at the same position.

SUGGESTIONS FOR FURTHER WORK

While it is hoped that this work provides a useful starting point, it is by no means the final word on this subject. We hope that this article will spur others to investigate the effect of factors not con-

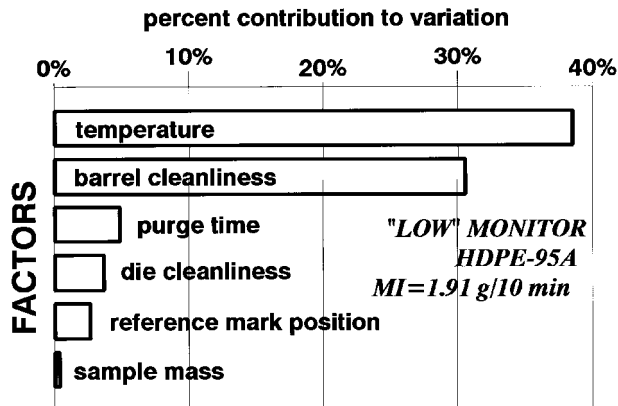


Figure 7 The relative contribution (in percentage terms) of the various factors to the variability in the MI measurement for the low MI monitor (HDPE-95A).

sidered in this study (e.g., barrel diameter, piston land length, and die length). Different definitions of "cleanliness" should be explored (random or un-reproducible processes of "dirtiness" are unfortunately inevitable in real life, but it is important to bear in mind that the DOE format cannot handle these). The use of more complex designs (e.g., central composite design) to examine separately the effect of each individual two-factor interaction and curvature in the prediction equation is recommended. In addition, the effect of lower and higher MI resins than those used in this study as well as different types of resins (polypropylene, nylon, etc.) should be studied to determine how generic our findings are. Only a 2.16-kg weight was used in this study; it would be worth considering how the use of other weights (5, 10, or 21.6 kg) would affect the results. As Reilly and Appel⁴ point out, much of the discussion related to such issues for melt indexers is also applicable to more sophisticated capillary rheometers.

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