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## An Evaluation of the Effects of Varying Composition and Processing on Several Encapsulating Resins

K. B. Wischmann, E. V. Thomas

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## **AN EVALUATION OF THE EFFECTS OF VARYING COMPOSITION AND PROCESSING ON SEVERAL ENCAPSULATING RESINS**

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### **ABSTRACT**

This investigation describes how a statistically designed experiment can be useful to characterize the relationship between a fundamental material property such as the glass transition temperature,  $T_g$ , and various processing parameters, e.g. composition, cure time, and temperature. To illustrate, formulation weighing errors can have a dramatic affect on material properties such as thermal, mechanical, and electrical properties. The glass transition temperature was selected for monitoring because it represents the materials state of cure and it is relatively easy to determine. Specifically, EPON 828 systems cured with diethanolamine and Shell Z, respectively, were investigated plus a mixture of the latter that employed aluminum oxide as a filler. This investigation showed that  $T_g$  changed very little with cure temperature in the DEA system compared to Shell Z, whereas the latter system appeared to display synergistic effects contrary to the DEA system. In the filled formulation, loading level had very little effect on  $T_g$ . The significance of this study is that the relationship between  $T_g$ , the composition and processing factors can be used to help diagnose the cause of misprocessed material.

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INTRODUCTION

Sandia National Laboratories, SNL, has been involved in encapsulating electronic devices for over 30 years. During this time our laboratory has developed a number of encapsulants tailored to withstand a variety of environments; a description of these materials can be found elsewhere (1). After development, these encapsulants are employed in numerous manufacturing scenarios. During encapsulating operations, processing specifications are used to define materials ratios, cure times, and temperatures. Variability is built into the specification to allow the manufacturer some latitude in controlling the process. For example, curing ovens at Allied-Signal are controlled to  $\pm 6^{\circ}\text{C}$ ; as a result there can be a  $12^{\circ}\text{C}$  range in cure temperature from high to low. Because of the allowance of a  $12^{\circ}\text{C}$  range in cure temperature, it is of interest to know how the physical properties of the encapsulant vary with processing over this cure temperature range. Similar situations exist with cure time and material composition. For example, if an error is made during weighing or if an abnormal cure time is employed, again what is the effect on physical properties?

To insure quality material, it would be advantageous to have a measurement that would be sensitive to perturbations of encapsulant compositions and cure schedule. If a definitive measurement was available, then discrepant material could be identified on the basis of this measurement. As a candidate, we decided to measure the glass transition temperature,  $T_g$ . Certainly, there are other physical properties that could be measured e.g. mechanical strength, fracture toughness, etc.; however,  $T_g$  is a good measure of the state of cure in an encapsulant, and is relatively easy to determine. In fact, when trouble-shooting misprocessed material, the  $T_g$  is often determined and compared to a known value. Unfortunately, when abnormal values are obtained, one does not know exactly what accounts for the discrepancy.

Therefore, this study was conducted to help understand the relationship between  $T_g$  and 1) the cure schedule and 2) encapsulant composition. Specifically, three formulations were studied, two unfilled, EPON 828/DEA and EPON 828/SHELL Z, and one filled, EPON/SHELL Z/ $\text{Al}_2\text{O}_3$ . This paper describes the experimental strategies and associated analyses used in this investigation as well as drawing some practical conclusions based on this work.

## EXPERIMENTAL APPROACH AND PROCESS

Specimens of the three encapsulants, EPON 828/Z, EPON 828/DEA, and EPON 828/Z/Al<sub>2</sub>O<sub>3</sub>, were produced under various conditions; SNL processing specifications, 9927109, 9927019, and 9927061, respectively were closely followed. The glass transition temperatures of these specimens were determined using a DuPont 940 thermomechanical analyzer, TMA. TMA is a technique in which the deformation of a substance is measured under static load as a function of temperature. The T<sub>g</sub> is measured at the onset of softening. The conditions of operation were: 1) a 50g weight was used in the penetration mode, 2) samples were run at 5°C/min; duplicate samples were taken at two different locations in a given coupon. Preliminary standardization experiments indicated this determination was subject to a measurement uncertainty of about  $\pm 2^\circ\text{C}$ .

For the EPON 828/Z formulations, both filled and unfilled, the nominal cure schedule consists of 4 hours at ambient temperature followed by 4 hours at 54°C and 16 hours at 93°C. Thus, the specific processing factors that were considered are the time at ambient temperature and the cure times and temperatures of the other two cure steps. The cure schedule for EPON 828/DEA involves simply a dwell at 71°C for 24 hours, so that only two processing factors were considered, the cure time and temperature of the single cure step.

The compositional factors that were studied depend on the particular formulations. The unfilled formulations are binary systems, while the filled formulation is a ternary system. The compositions (parts by weight, pbw) and weight fractions (in parenthesis) of the 3 formulations are shown in Table I.

Table 1. Resin Formulations

1#	EPON 828	100pbw (.833)	2#	EPON 828	100pbw (.893)
	Shell Z	20pbw (.167)		DEA	12pbw (.107)
3#	EPON 828	100pbw (.238)			
	SHELL Z	20pbw (.048)			
	Al <sub>2</sub> O <sub>3</sub>	300pbw (.714)			

In the case of the binary unfilled systems, the percentage or weight fraction of the curing agent is sufficient to describe the composition. The compositional factor considered in these cases is percentage by weight of the curing agent. The specific compositional factors in the ternary filled system can be described in a number of ways. The particular set of compositional factors considered in this case will be described later.

An efficient way to study the effects of processing and composition on  $T_g$  is through a statistically designed factorial experiment. In such an experiment, the levels of experimental factors are systematically varied, in this case to obtain formulations with a variety of compositional and processing attributes. By using a factorial experimental strategy, it is possible to assess the effect of experimental factors, individually and to some degree jointly, on  $T_g$ , with greater efficiency than haphazard, or one-factor-at-a-time strategies achieve.

Initially, the experimentation was limited to the unfilled EPON 828/Z system. The total number of experimental factors considered with respect to this system was six. Because of the relatively large number of experimental factors considered and because of the relative shortage of testing resources available, the experimental design chosen was a quarter-fraction of a  $2^6$  factorial design (2), supplemented with several centerpoints that replicate the nominal processing and compositional conditions. A full  $2^6$  factorial experiment would have involved producing specimens at each of the 64 possible combinations of the six factors when each factor could assume one of two levels. This was not economically feasible. The design used required a total of only 16 samples. The processing conditions of the 16 samples were chosen in a way so that the individual effects of the factors on  $T_g$  could be assessed. Results from this experimentation with the EPON 828/Z system indicate that variations in the factors that determine the first two cure steps had no significant effects on  $T_g$ . Therefore, subsequent experimentation with the filled and unfilled EPON 828/Z systems involved only the composition and the time and temperature of the third cure step as experimental factors. In the case of the EPON 828/DEA system, only three experimental factors (percent DEA, cure time, and cure temperature) were considered. The design chosen, a full  $2^3$  factorial supplemented with several centerpoints, was suitable for estimating interactions among the three factors. After analyzing the results from this initial experiment, it was concluded that additional experimentation was needed in order to characterize  $T_g$  over the ranges of the three factors. This additional experimentation was needed to model quadratic effects.

#### MODELING OF $T_g$

The experimental results provide the basis for developing descriptive and predictive models. The degree of modeling of the experimental results depended on the particular system. With respect to the unfilled systems, attempts were made to develop predictive models of  $T_g$  as a function of composition and the processing variables. In the case of the filled system, the primary intent was to model  $T_g$  only at the descriptive level due to the complexity introduced by a third mixture component ( $Al_2O_3$ ).

The predictive models that relate Tg to the experimental factors are simple functions of the experimental factors. For example, the case of the EPON 828/Z unfilled system (with 15 pbw curing agent) the model is

$$\hat{T}_g(t, T) = \hat{\beta}_0 + \hat{\beta}_1 \cdot t + \hat{\beta}_2 \cdot T + \hat{\beta}_3 \cdot t \cdot T,$$

where t is the cure time, and T is the cure temperature.  $\hat{T}_g$  is the fitted value of Tg, and  $\hat{\beta}_0$ ,  $\hat{\beta}_1$ ,  $\hat{\beta}_2$ , and  $\hat{\beta}_3$  are estimated model parameters. Various other models were considered in this and the other cases. The models that were selected were the simplest (among the various alternatives that were considered) that provided an adequate fit to the experimental data.

The models constructed in this study provide estimates of the average value of Tg that is expected at specified values of the experimental factors. Individually, the observed responses (Tg) differ from the fitted responses ( $\hat{T}_g$ ) because of the error in measuring Tg and the difference between the model and reality (model error). Based on earlier measurement error studies, we believe that the model error is sufficiently small in these cases, i.e., the models approximate reality rather well. Therefore, we believe that, in general, most of the differences between Tg and  $\hat{T}_g$  are due to measurement error.

## RESULTS AND DISCUSSION

EPON 828/DEA The unfilled systems will be discussed first. EPON 828 cured with diethanol amine (DEA) has been used as an encapsulant at SNL for about 30 years. The primary use of this system is to encapsulate or impregnate transformer windings and capacitor coils. The experimental strategy for this two component system was relatively simple. The first step in setting-up such an experiment is to bound the problem, that is, dictate the most abnormal set of processing conditions envisioned, while being able to produce a cured material for analysis. If the encapsulant does not cure, obviously, something is wrong. In the nominal formulation, 100pbw EPON 828 is reacted with 12pbw DEA per specification 9927019. Since composition is a variable, we introduced a deliberate weighing error. The lower and upper limits for this study were set at 9pbw and 18pbw; anything outside these limits would result in incomplete cure. For cure time and temperature, 16 hours at 71°C is specified. The experimental range considered was from 8 to 72 hours and 60° to 80°C. A long cure time was employed to evaluate post cure effects. The reported Tg for encapsulant processed under nominal conditions is about 70°C. In this study, we found that the Tg of encapsulant processed under nominal conditions was slightly lower, ~68°C.

As previously mentioned, experimentation was performed in stages as it became apparent that the results originating from the initial design were not sufficient to model the relatively complex way in which Tg varied over the range of the experimental factors. The empirical model developed from the experimental data is

$$T_g = \beta_0 + \beta_1 \cdot D + \beta_2 \cdot D^2 + \beta_3 \cdot \log(-3 + .5t) + \beta_4 \cdot T + \beta_5 \cdot D \cdot \log(-3 + .5t) + \epsilon, \text{ where}$$

$T_g$  = observed glass transition temperature (degrees C),

$D$  = % by weight DEA,

$t$  = cure time (hours),

$T$  = cure temperature (degrees C), and

$\epsilon$  = is a random error with zero mean and standard deviation,  $\sigma$ .

The relatively complicated form of the model dependence on cure time ( $t$ ) is admittedly ad hoc. However, it does match the experimental data well.

Least-squares regression yielded the set of parameter estimates (in this case,  $\hat{\beta}_0, \hat{\beta}_1, \hat{\beta}_2, \hat{\beta}_3, \hat{\beta}_4, \hat{\beta}_5$ ) that minimized the sum (over all observations) of the squared differences between the fitted responses ( $\hat{T}_g$ ) and the observed responses ( $T_g$ ). The parameter estimates (and respective standard errors) obtained by least-squares regression are,

$$\begin{aligned} \hat{\beta}_0 &= -102. & (13.), \\ \hat{\beta}_1 &= 22.0 & (1.8), \\ \hat{\beta}_2 &= -.816 & (.069), \\ \hat{\beta}_3 &= 30.3 & (4.3), \\ \hat{\beta}_4 &= .169 & (.077), \text{ and} \\ \hat{\beta}_5 &= -1.35 & (.34). \end{aligned}$$

The standard errors of the parameter estimates can be used to judge the range of uncertainty in the parameter estimates. For instance, if the experiment was repeated with a corresponding new analysis, it would not be unlikely that a given parameter estimate would differ from the original estimate by about a standard error. However, it would be unusual for the new estimate to differ by more than two standard errors from the original estimate.

The fitted glass transition temperature ( $\hat{T}_g$ ), obtained by substituting the parameter estimates into the model equation, can be displayed graphically as a function of the experimental factors in a number of ways. Figures 1-3 are contour plots that show  $\hat{T}_g$  versus cure time and pbw DEA at three cure temperatures, 60°, 71° and 80°C. The magnitude of typical deviations of the observed value of  $T_g$  from  $\hat{T}_g$  is about 4°C ( $\hat{\sigma} \approx 4$ ). Appendix A displays the experimental conditions (% DEA, cure time, cure temp) along with  $T_g$  and  $\hat{T}_g$  for all experiments.

Figures 1-3 show that the estimated optimum DEA level (yielding the highest Tg) is between 12 and 13 pbw. Processing specifications call out 12 pbw DEA. The contour plots show that more or less DEA results in a lower Tg. Less DEA probably results in undercure whereas larger amounts may introduce plasticization/dilution effects. At a constant curing agent level and cure time, e.g., 12 pbw and 24 hrs, cure temperature appears to have little effect; that is, going from a 60° to 80°C only increases the Tg from 68° to 71°C. Increasing the cure time above the nominal 24 hours, at a constant DEA level and cure temperature, e.g. 12 pbw DEA and 60°C appears to increase the Tg somewhat. This may indicate some post curing effects, but this phenomenon is not well understood. The kinetics of this system are currently being investigated. Nevertheless, the most dramatic effects on Tg, relative to the nominal conditions, are observed at low cure time and either high or low DEA levels.

EPON 828/Z The second system to be discussed is EPON 828 cured with SHELL Z according to specification 9927109; the nominal ratio is 100/20pbw, respectively. SHELL Z is a eutectic mixture of methylene dianiline (MDA) and meta-phenylene diamine (MPDA). SHELL Z is employed in SNL formulations to achieve high use-temperature materials; average Tg's are 117°C. A screening experiment (one-quarter fraction of a 2<sup>6</sup> factorial) was performed to examine the effects of variations of the percent curing agent and the cure schedule on Tg. The determinants of the cure schedule are illustrated in Figure 4 and are as follows: the time at room temperature, the time and temperature at the second cure step, and the time and temperature at the third cure step. The ranges of experimental factors are indicated in Table 2. To reiterate, the purpose of this experiment was to identify which of the six factors considered has a significant influence on Tg.

Table 2. Range of Experimental Factors  
for EPON 828/Z Systems

<u>Factor</u>	<u>Low Level</u>	<u>Nominal</u>	<u>High Level</u>
1. Shell Z Content	15pbw	20pbw	25pbw
2. Cure Time (1 <sup>st</sup> step)	2 hours	4 hours	24 hours
3. Cure Temp (2 <sup>nd</sup> step)	43°C	54°C	67°C
4. Cure Time (2 <sup>nd</sup> step)	2 hours	4 hours	16 hours
5. Cure Temp (3 <sup>rd</sup> step)	81°C	93°C	140°C
6. Cure Time (3 <sup>rd</sup> step)	4 hours	16 hours	48 hours

Statistical analysis of the experimental results from the screening experiment indicated that the percent curing agent and the third curing step affected Tg significantly. Variations in the times and temperature (at least in the ranges that are considered) of the first and second cure steps have little or no effect on Tg.

Additional experiments were performed so that the relationship between Tg and the three important experimental factors could be modeled. The results from the original screening experiment and the additional experiments are presented in Appendix B along with the associated experimental conditions (pbw Shell Z, third cure time, third cure temperature).

The manner in which the third cure time and temperature affect Tg depends somewhat on the curing agent level. For a fixed cure schedule, Tg increases dramatically when the percent cure agent varies from a low to intermediate level. When the percent cure agent varies from an intermediate to high level there is no significant change in Tg. Therefore, two empirical models, were constructed for the Shell Z system. One is associated with the low level of cure agent, the other is associated with intermediate to high levels of cure agent.

When the curing agent content is 15pbw, the following model is a good approximation to the observed value of Tg.

$$Tg = \beta_0 + \beta_1 \cdot t + \beta_2 \cdot T + \beta_3 \cdot t \cdot T + \epsilon, \text{ where}$$

- Tg = observed glass transition temperature (degrees C),
- t = cure time (hours) of the third cure step,
- T = cure temp (degrees C) of the third cure step, and
- $\epsilon$  = a random error with standard deviation,  $\sigma$ .

The estimates of the model parameters ( $\beta_0, \beta_1, \beta_2, \beta_3$ ) obtained by least-squares regression along with their respective standard errors are,

$$\begin{aligned} \hat{\beta}_0 &= 81.5 && (.13), \\ \hat{\beta}_1 &= -1.21 && (.38), \\ \hat{\beta}_2 &= .0496 && (.14), \text{ and} \\ \hat{\beta}_3 &= .0163 && (.004) \end{aligned}$$

Although the parameter estimate in the model corresponding to T ( $\hat{\beta}_2$ ) is smaller than its standard error, it is included so that the interaction model is complete. Figure 5 illustrates the contours of this estimated response surface. Notice the strong synergistic effect on Tg when both time and temperature are increased. The observed Tg's typically deviate from this estimated response surface by about 3°C (i.e.,  $\hat{\sigma} \approx 3^\circ\text{C}$ ).

In the case where the level of curing agent is moderate to high (20-25%), the synergy between time and temperature disappears. Therefore, a simpler model can be used to represent Tg. That is,

$$Tg = \beta_0 + \beta_1 \cdot t + \beta_2 \cdot T + \epsilon.$$

The parameter estimates (and respective standard errors) in this case are,

$$\begin{aligned}\hat{\beta}_0 &= 24.2 & (12), \\ \hat{\beta}_1 &= .183 & (.061), \text{ and} \\ \hat{\beta}_2 &= .918 & (.13).\end{aligned}$$

This model was constructed by pooling experimental results associated with moderate to high levels of curing agent (20-25%) as it appears changes in curing agent content within this range affected Tg very little, if at all. In this case  $\hat{\sigma} \approx 4^\circ\text{C}$ . Figure 6 represents the expected values of Tg when the curing agent is in the range from 20-25pbw.

Without data corresponding to levels of curing agent between 15 and 20pbw, it is not possible to model Tg in that range. It is clear that Tg decreases when the curing agent level decreases from 20 to 15pbw. However the precise way in which Tg is decreasing is not estimable.

Upon comparing curing agent ratios, the observed difference in Tg between the 20 and 25pbw SHELL Z under nominal processing conditions was at most 2-3°C, whereas the difference between 15 and 20pbw SHELL Z under similar conditions was about 20°C. Again, similar to DEA, low curing agent ratios make dramatic differences. Low cure times and temperatures also show marked effects. At the nominal composition, Tg drops to around 100°C with a 4 hour cure at 80°C compared to about 115°C with the nominal cure. Therefore, abnormally low Tg's would be probably due to low curing agent levels. To illustrate, when the composition contains 15pbw Z, only extreme increases in time and temperature will raise the Tg above 100°C. When the composition has at least 20pbw Z, or higher, increased cure time has little effect on Tg; only 3-5° increases are realized. This indicates that the post-cure effects are small which is desirable. In contrast to the DEA cured system, cure temperature has a dramatic effect on Tg. For example, at 20pbw SHELL Z and 24 hours cure, the Tg at 81°C cure is about 102°C whereas when cured the same length of time at 105°C, the Tg is about 124°C. The data shows that dramatic changes in Tg/state of cure can occur as a result of changes in processing. These changes in state of cure are often reflected in differing mechanical and electrical properties which could jeopardize the function of the encapsulated component.

EPON 828/Z/Al<sub>2</sub>O<sub>3</sub> The next system that will be discussed is a filled system. The addition of a third compositional variable (filler) changes the nature of the experimental strategy used. The compositional space can be represented by a ternary diagram. The formulation discussed here consists of 300pbw Al<sub>2</sub>O<sub>3</sub>, 100pbw EPON 828 and 20pbw SHELL Z, or in terms of weight fraction, .714, .238 and .048 respectively. As previously mentioned, moderate-to-large variations in the first and second curing steps had at most minimal effects on Tg in the SHELL Z formulations. Therefore, the only experimental factors considered were the third processing step (time and temperature) and composition. Tables 3 and 4 outline the matrix of experiments run.

Table 3. Compositions by Weight Fraction  
(Al<sub>2</sub>O<sub>3</sub>, EPON-828, SHELL Z)

1. (.7712, .1904, .0384)
2. (.676, .2856, .0384)
3. (.752, .1904, .0576)
4. (.6568, .2856, .0576)
5. (.714, .238, .048)

Table 4. Curing Schedules\*

	Cure Temp. #3	Cure Time #3
A	81°C	4 hours
B	81°C	48 hours
C	93°C	16 hours
D	104°C	4 hours
E	104°C	48 hours

\* Cure steps 1 and 2 were at nominal conditions  
(4 hours at room temperature followed by 4 hours  
at 130°F)

A total of 25 experiments were run, 5 compositions, each cured at 5 different conditions. The experiments are nonlinear with time in order to determine any post cure effects. The compositions studied are shown in Figure 7, which is a portion of a ternary diagram. The central point (composition 5) is the nominal composition. The other compositions (1, 2, 3, 4) define the compositional region of interest. This region was selected by allowing the mole ratios of EPON-828 and Shell-Z to vary individually by as much as 20%. For instance, composition 1 contains 20% less EPON-828 and 20% less Shell-Z (in terms of weight fractions) than the nominal composition. Note that compositions 1 and 4, however, have the same EPON-828/Shell Z stoichiometry as the nominal composition. It is believed that the region defined by compositions 1, 2, 3, and 4 span compositions that could realistically arise due to weighing errors. The processing conditions studied at each composition are shown in Figure 8, again the center point represents the nominal conditions. The experimental results are shown in Figure 9. For the range of cure schedules considered, T<sub>g</sub> was observed to be very similar for compositions 3, 4 and 5. Except for the low time, low temperature cure, composition 1 is similar to these three compositions with respect to T<sub>g</sub>. Composition 2 yielded markedly different T<sub>g</sub>'s than the others. This composition is low in curing agent while high in epoxy. Therefore, similar to the unfilled system, T<sub>g</sub> is not overly sensitive to composition except when the curing agent/epoxy ratio is very low.

The effects of cure schedule on Tg can also be seen in Figure 9. The general qualitative effects of cure temperature and cure time are similar across composition. In general, Tg increases with increasing time and temperature. Quantitatively, the effects of cure temperature and cure time depend on composition. The strongest effects are seen in compositions 3,4 and 5. The smallest effects are seen in composition 2. The dependence of Tg on cure schedule can be easily modeled in compositions 3, 4 and 5. Figure 10 graphically illustrates the estimated relationship between Tg and the cure time (t) and temperature (T). This estimated relationship is:

$$\hat{T}_g = -118 + 16T^{1/2} + 1.94t^{1/2}$$

Superimposed on the contour plot are the average Tg's (with respect to compositions 3, 4 and 5) for each of the processing conditions.

In an effort to further understand the sensitivity of Tg to a low curing agent/epoxy ratio, several additional experiments were performed. The new compositions, see Table 5, were cured at the nominal cure conditions.

Table 5. Weight Fractions and Tg's of New Compositions  
EPON 828/SHELL Z/Al<sub>2</sub>O<sub>3</sub>

	<u>Weight Fractions</u>	<u>Tg</u>
6.	(.2476, .0384, .714)	112
7.	(.2618, .0432, .695)	110
8.	(.2856, .048, .6664)	113

Figure 11 displays the Tg observed at each of these three new compositions in addition to the Tg observed at the other five compositions (nominal cure schedule). As these observed Tg's are relatively close to those observed at compositions 3, 4 and 5, it is clear that Tg changes very rapidly as the composition moves from one of these compositions to composition 2. Therefore, Tg is a good indicator of discrepant composition only when the curing agent/epoxy ratio is much less than the nominal ratio.

From these results, it was concluded that Tg increases approximately linearly with the square roots of cure time and temperature over the bounded experimental region. At the nominal composition, the difference between the low-temperature, low-time Tg and the high-temperature, high-time Tg is about 17°C. Tg is relatively constant over the range of composition considered except when the composition has a very low curing agent/epoxy ratio. Therefore, Tg could be used to identify material that has been processed incorrectly or that is markedly different with respect to the curing agent/epoxy ratio. This indirect measure of the curing agent/epoxy ratio would be much easier to measure than the composition, directly. A very low value of Tg (say below 100°C) would suggest a low curing agent/epoxy ratio.

## SUMMARY

This investigation demonstrates how a statistically designed experiment can be useful to characterize the relationship between a fundamental material property such as Tg and various process and compositional factors. For example, at a given curing agent level, the DEA cured system showed considerably less change in Tg than the SHELL Z system as temperature was increased. In the filled Shell Z system, the loading level of Al<sub>2</sub>O<sub>3</sub> did not have a very large effect on Tg as did curing agent level or temperature. However, when the curing agent/epoxy ratio is much less than the nominal value, Tg is reduced considerably.

The significance of this study is that the observed relationship between Tg and the composition and processing factors can be used to help diagnose the cause of discrepant material. For example, when there is a significant deviation in Tg compared to nominal, the process engineer would have data to substantiate his or her supposition that the material does not have enough curing agent or that the encapsulant may have experienced too high a curing temperature. Hopefully, this work will stimulate the use of similar experimental strategies to systematically and economically investigate the effects of compositional and processing factors on materials and their properties.

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Appendix A - Experimental Results and Model Predictions (EPON 828/DEA)

PBW DEA	Time (Hrs)	Temp	T <sub>g</sub>	$\hat{T}_g$	T <sub>g</sub> - $\hat{T}_g$
9	8	60.0	39	40.2	-1.2
9	8	71.1	44	42.0	2.0
9	8	82.2	42	43.9	-1.9
9	8	82.2	46	43.9	2.1
9	24	71.1	55	59.4	-4.4
9	48	60.0	66	64.2	1.8
9	48	71.1	70	66.1	3.9
9	48	82.2	69	67.9	1.1
9	72	71.1	68	69.6	-1.6
9	72	71.1	74	69.6	4.4
10	8	71.1	49	48.6	0.4
10	24	71.1	60	64.6	-4.6
12	8	71.1	59	56.8	2.2
12	24	60.0	65	68.4	-3.4
12	24	71.1	72	70.2	1.8
12	24	71.1	75	70.2	4.8
12	24	71.1	71	70.2	0.8
12	24	82.2	64	72.1	-8.1
12	24	82.2	72	72.1	-0.1
12	48	71.1	78	75.4	2.6
12	72	60.0	72	76.3	-4.3
12	72	71.1	79	78.2	0.8
12	72	82.2	73	80.1	-7.1
13.5	24	71.1	72	70.2	1.8
15	8	60.0	50	54.9	-4.9
15	8	71.1	61	56.8	4.2
15	8	82.2	56	58.7	-2.7
15	24	71.1	72	66.4	5.6
15	48	60.0	70	68.2	1.8
15	48	71.1	71	70.1	0.9
15	48	82.2	77	72.0	5.0
18	12	71.1	43	45.0	-2.0
18	12	82.2	50	46.9	3.1
18	24	60.0	48	46.0	2.0
18	24	71.1	44	47.9	-3.9
18	24	82.2	49	49.8	-0.8
18	72	60.0	50	49.4	0.6
18	72	71.1	47	51.3	-4.3
18	72	82.2	55	53.2	1.8

Appendix B - Experimental Results and Model Predictions (EPON 828/Z)

PBW (Z)	Time (Hrs)	Temp	Tg	$\hat{T}_g$	Diff
15	4	82.2	86.0	86.1	-0.1
15	4	82.2	84.7	86.1	-1.4
15	4	104.4	86.8	88.6	-1.8
15	4	104.4	89.0	88.6	0.4
15	16	93.3	95.0	91.0	4.0
15	48	82.2	91.4	91.6	-0.1
15	48	82.2	91.2	91.6	-0.4
15	48	104.4	112.7	110.1	2.6
15	48	104.4	106.9	110.1	-3.2
20	4	93.3	111.0	110.6	0.4
20	16	82.2	107.0	102.6	4.4
20	16	93.3	116.5	112.8	3.7
20	16	93.3	115.0	112.8	2.2
20	16	104.4	125.0	123.0	2.0
20	48	93.3	120.0	118.7	1.3
25	4	82.2	87.5	100.4	-12.9
25	4	82.2	95.3	100.4	-5.1
25	4	104.4	119.5	120.8	-1.3
25	4	104.4	113.5	120.8	-7.3
25	16	93.3	119.0	112.8	6.2
25	16	93.3	112.5	112.8	-0.3
25	48	82.2	109.0	108.5	0.5
25	48	82.2	101.8	108.5	-6.7
25	48	104.4	126.5	128.9	-2.4
25	48	104.4	118.8	128.9	-10.1
25	48	104.4	131.0	128.9	2.1

Estimated T<sub>g</sub> versus Cure Time and PBW(DEA)  
Cure Temp = 60 deg C.

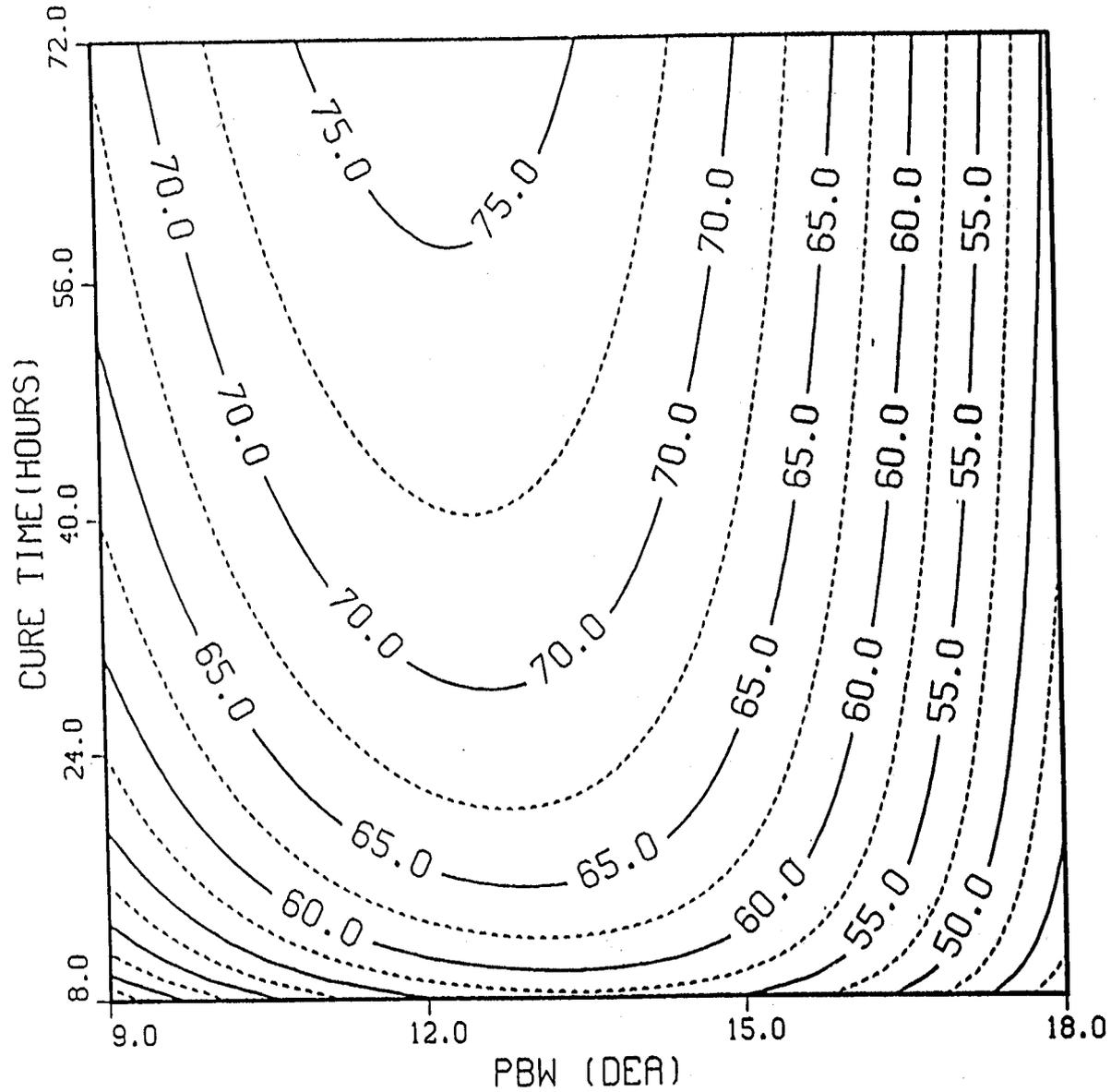


Figure 1.

Estimated  $T_g$  versus Cure Time and PBW(DEA)  
Cure Temp = 70 deg C.

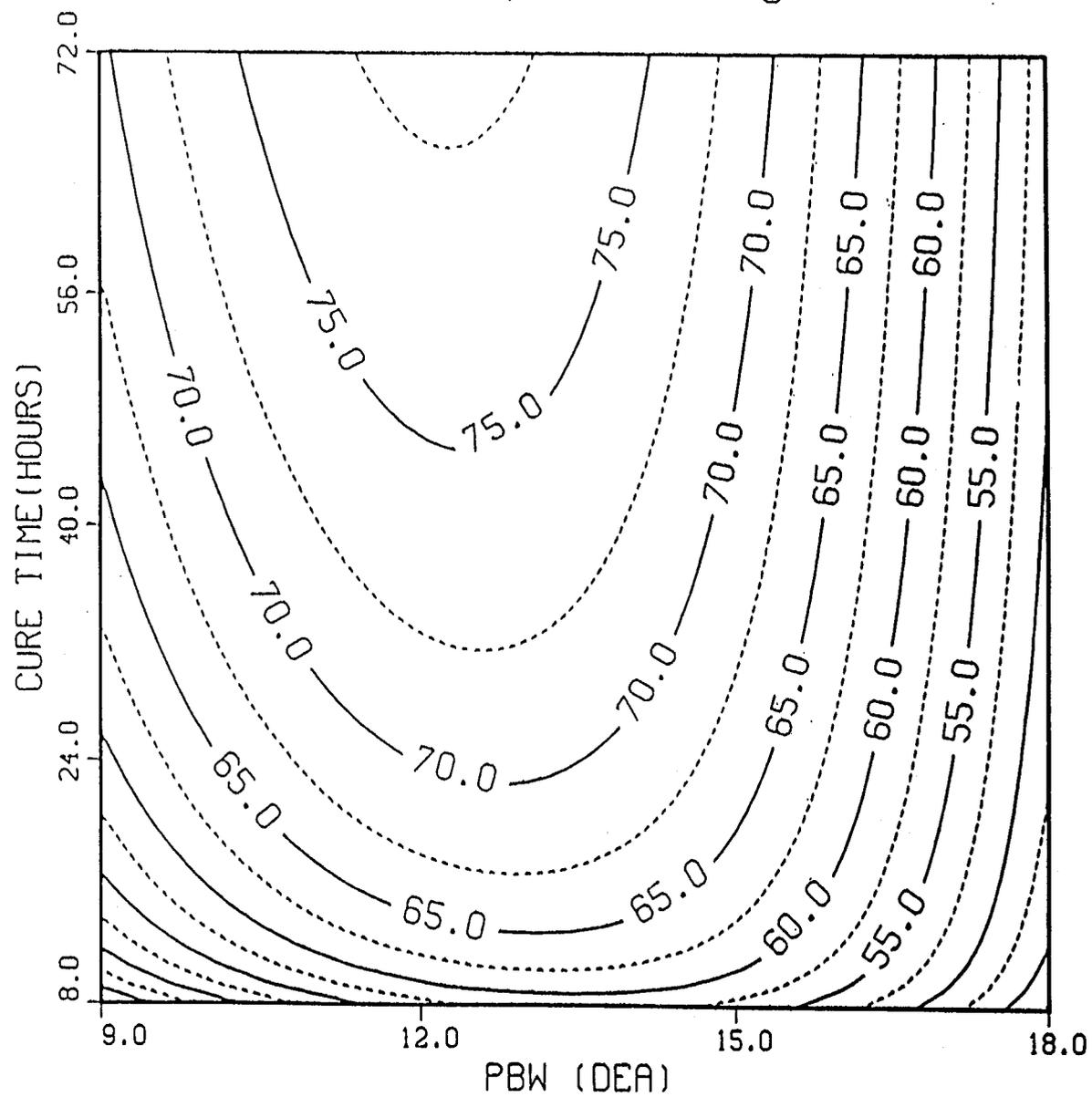


Figure 2.

Estimated T<sub>g</sub> versus Cure Time and PBW(DEA)  
Cure Temp = 80 deg C.

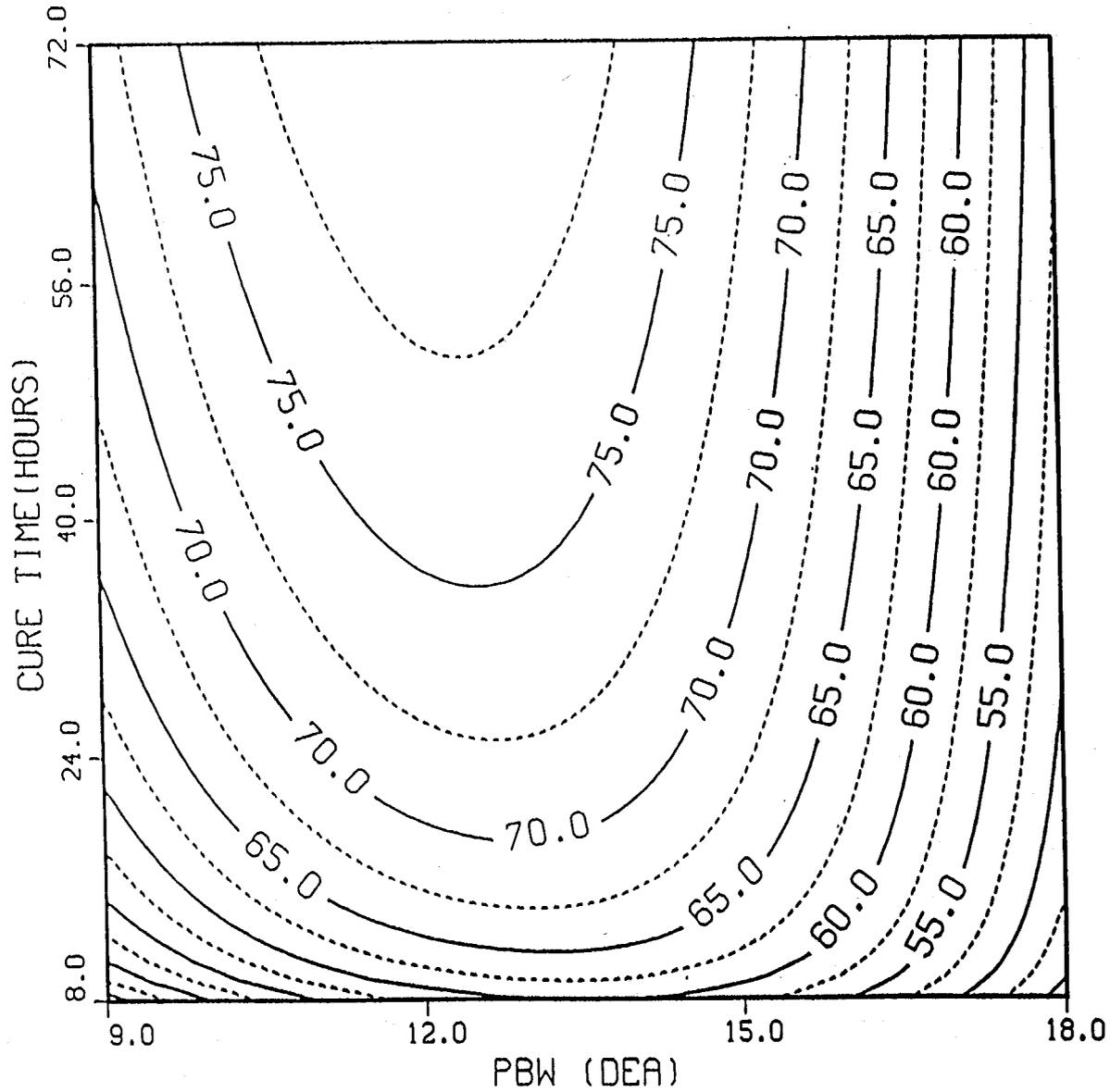


Figure 3.

# CURE SCHEDULE

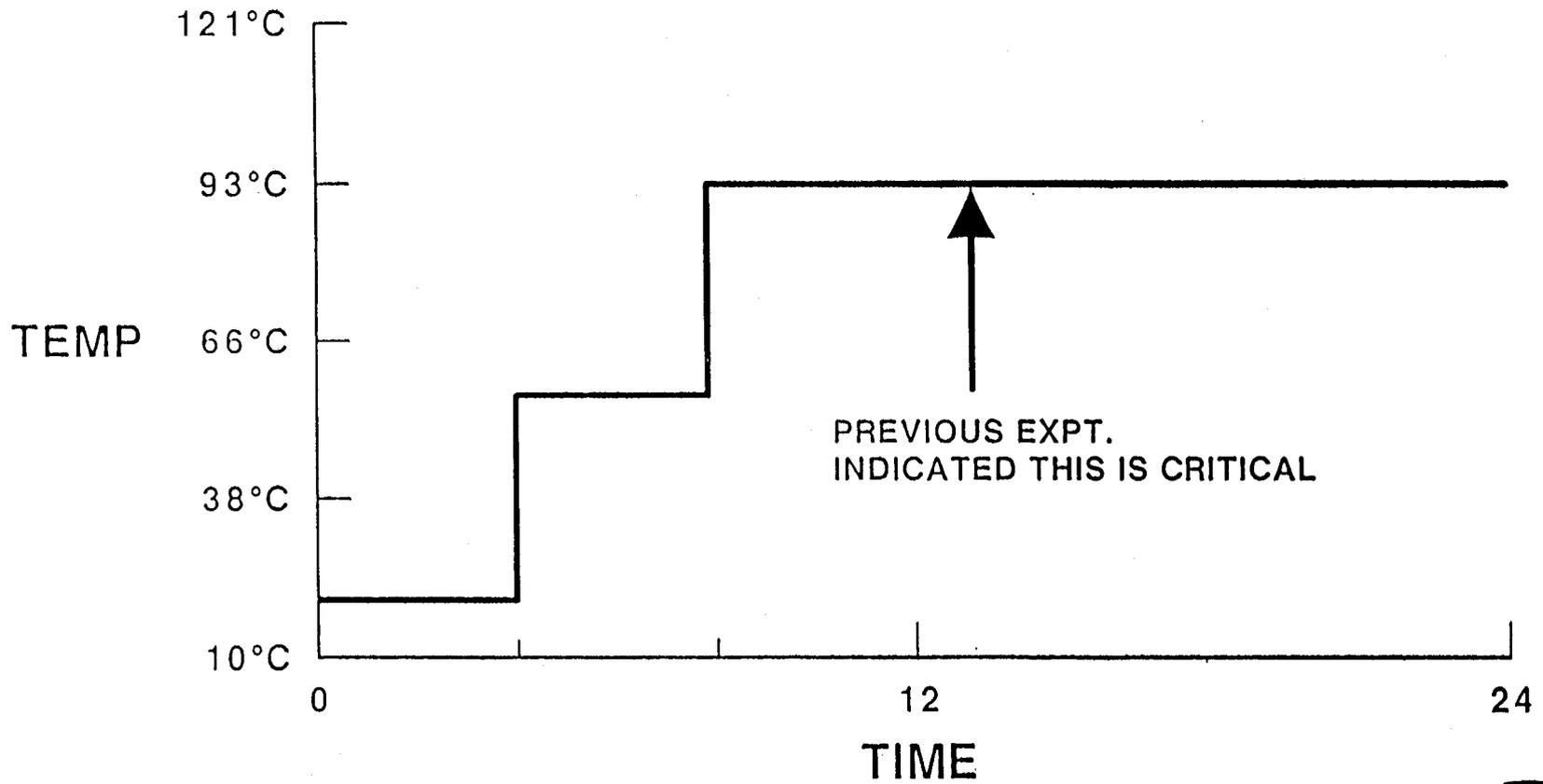


Figure 4.

88D7000.03



# Estimated T<sub>g</sub> versus Cure Time and Temperature PBW(Z) = 15

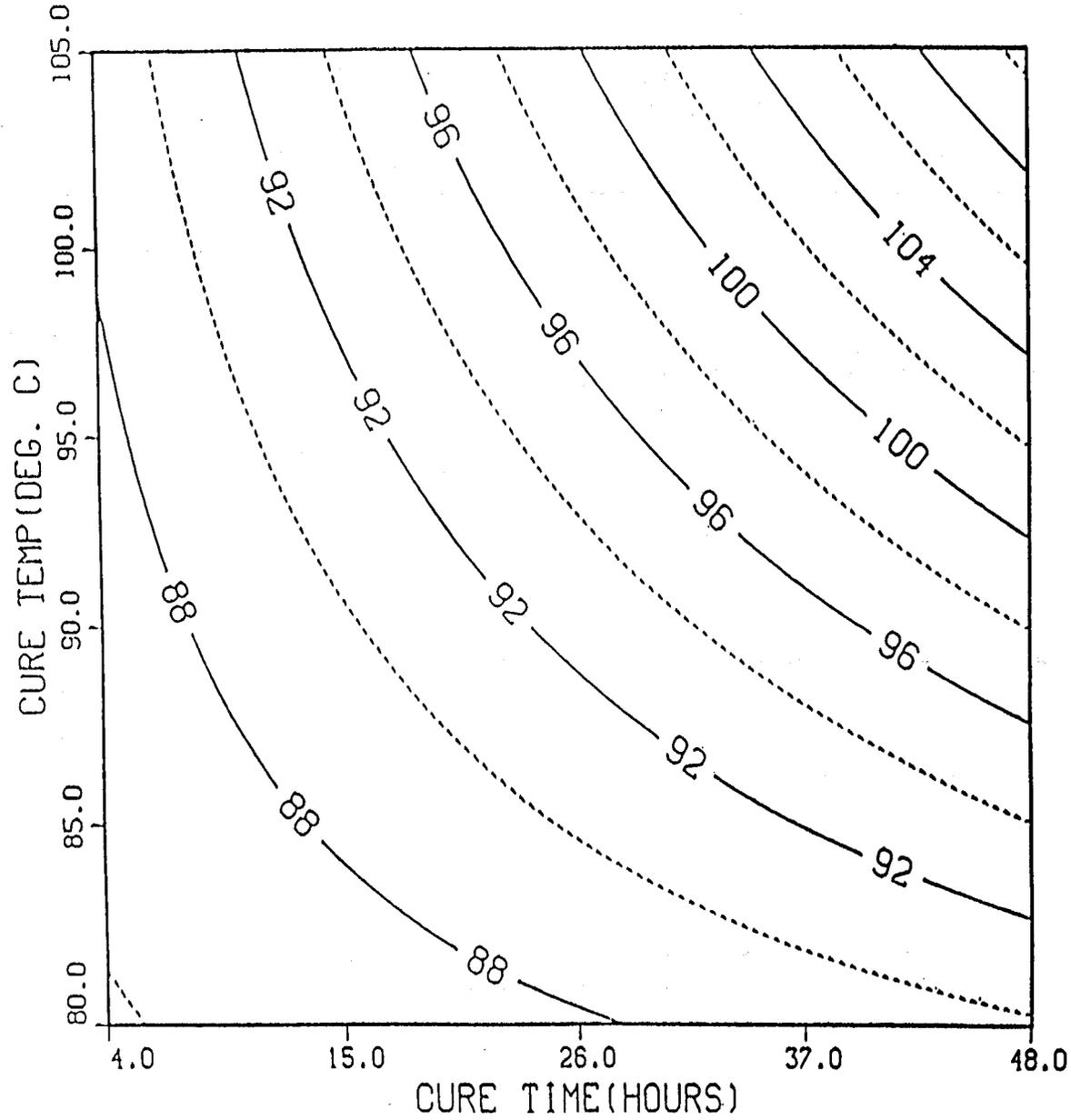


Figure 5.

# Estimated Tg versus Cure Time and Temperature 20 < PBW(Z) < 25

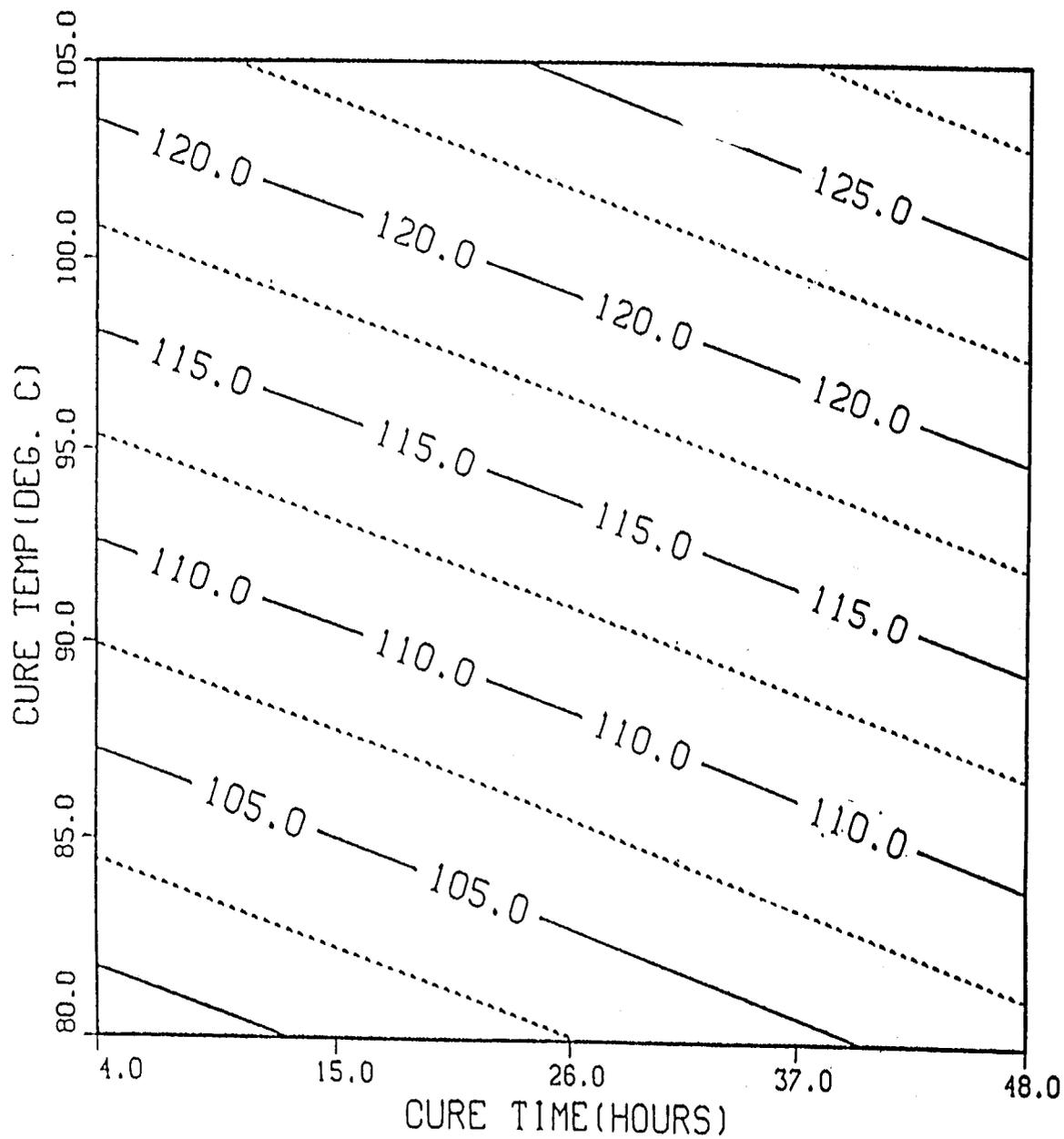


Figure 6.

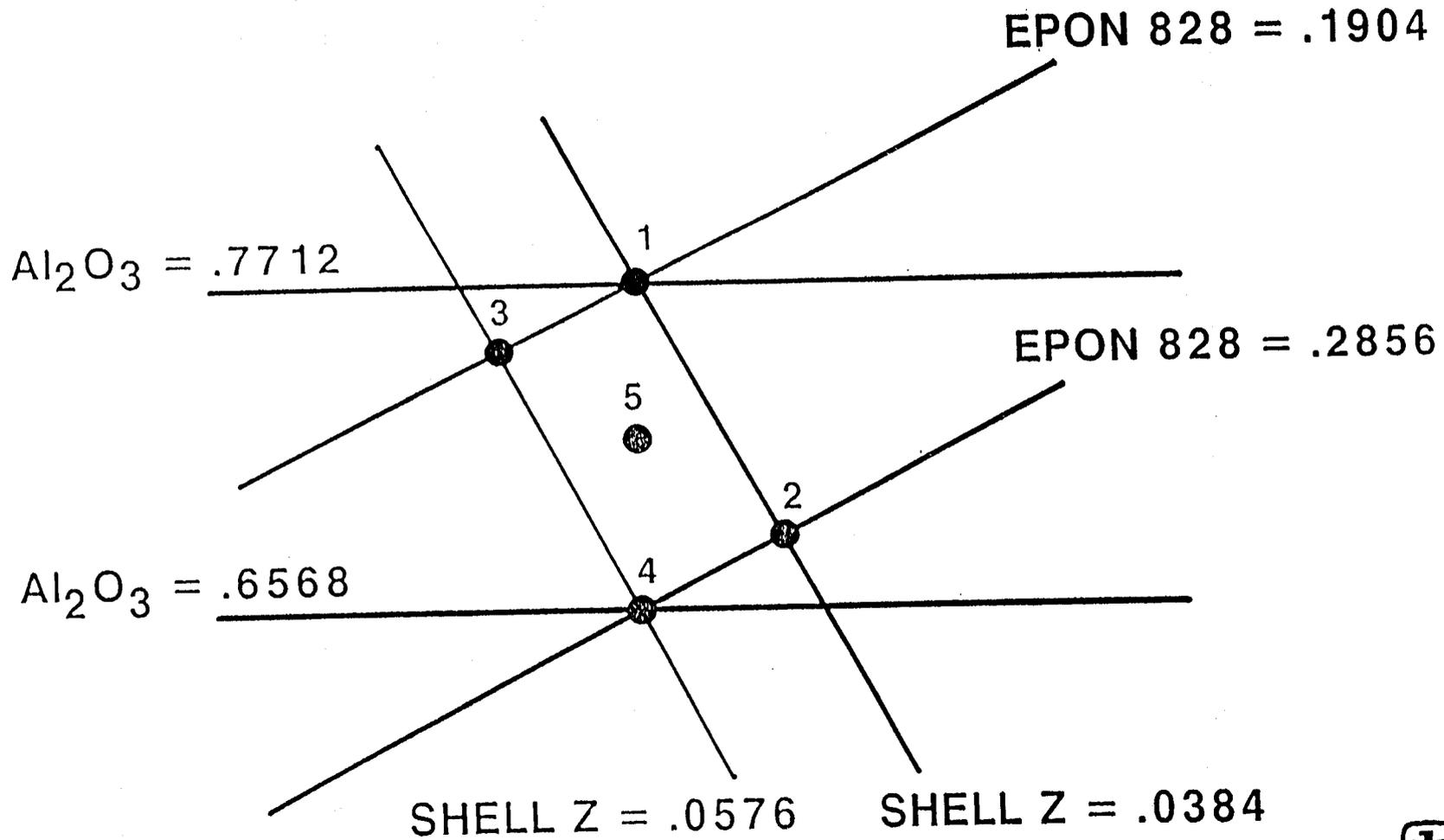


Figure 7.



# PROCESSING CONDITIONS

4 HOURS @ RT

4 HOURS @ 54°C

FOLLOWED BY

CURE TIME - 3

CURE TEMP - 3

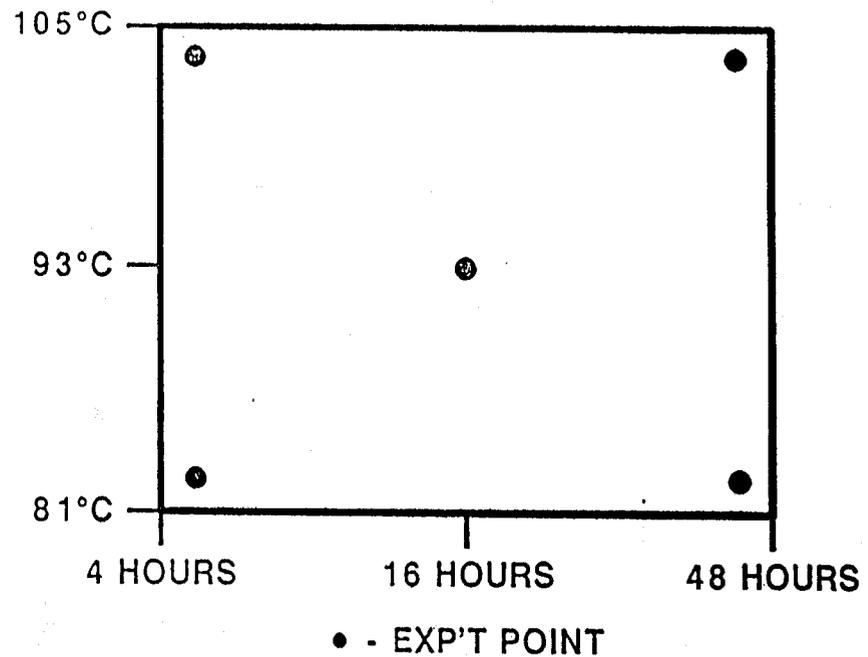
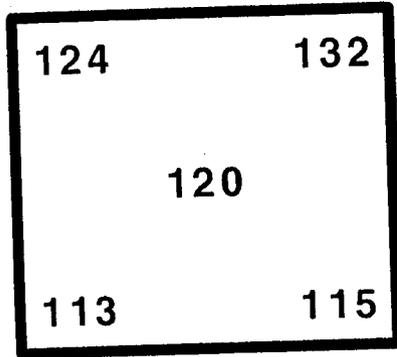


Figure 8.

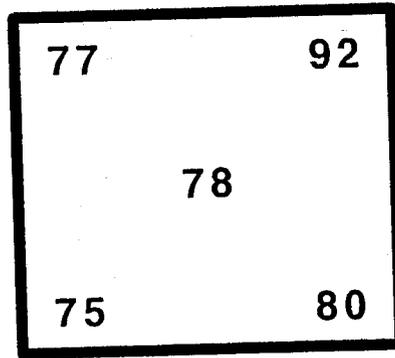


88D7000.02

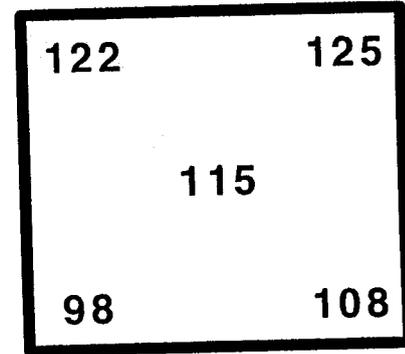
# EXPERIMENTAL RESULTS



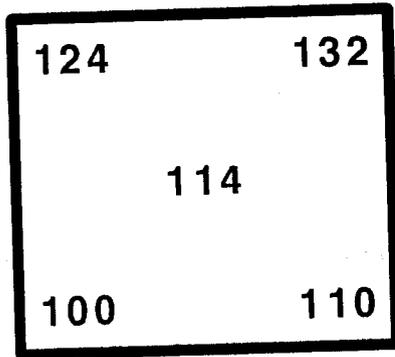
#1



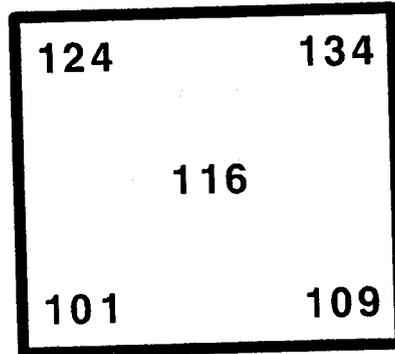
#2



#3



#4



#5

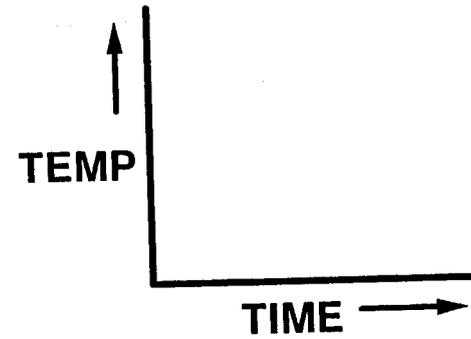


Figure 9.



Figure 10 - Estimated Dependence of Tg on Curing  
Given Mixtures 3., 4., 5.

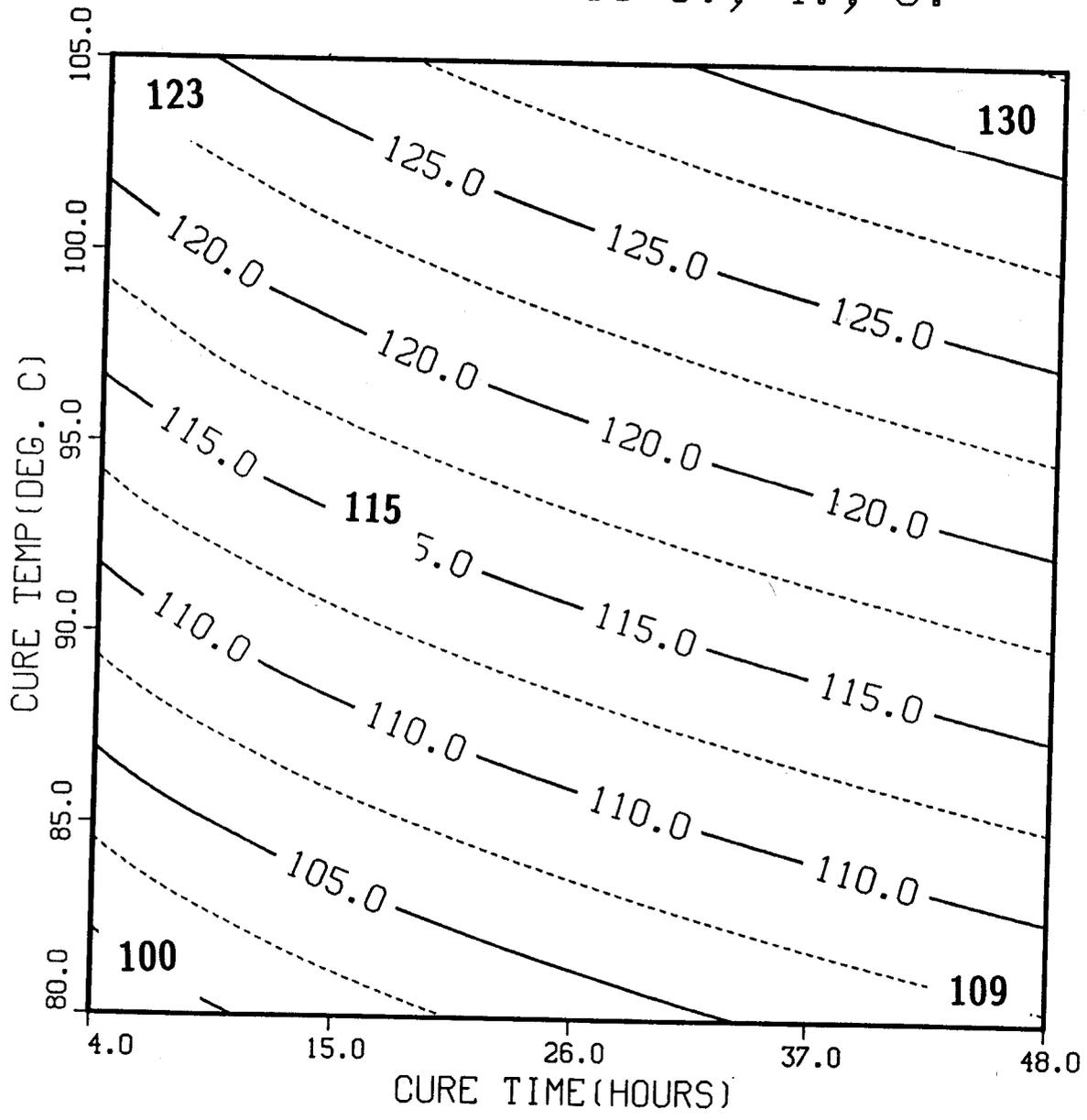


Figure 10

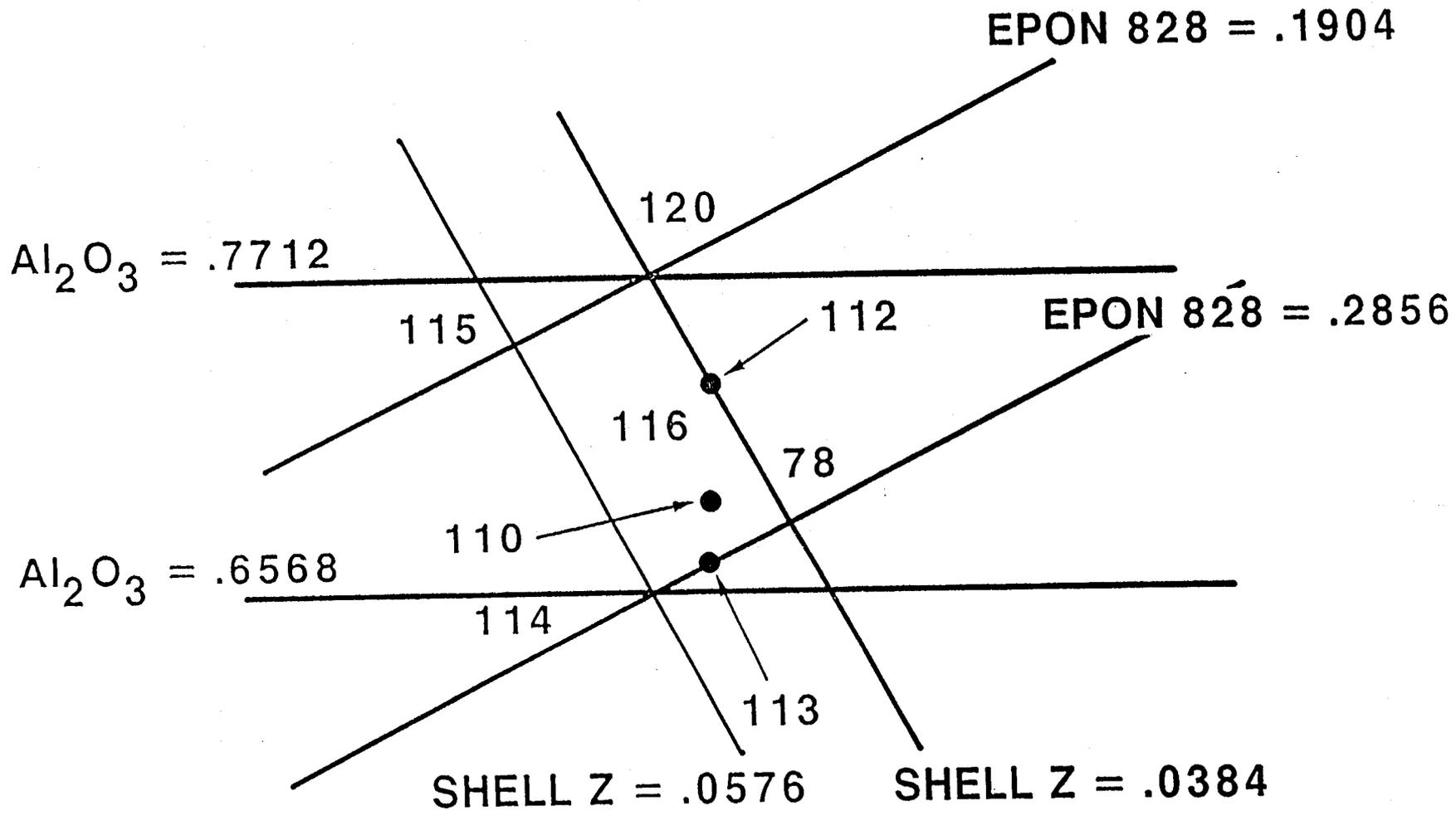


Figure 11.



Distribution:

SNL

1 1521 S. N. Burchett  
1 1811 C. Arnold  
1 1812 C. L. Renschler  
1 1813 D. Adolf  
1 1813 P. B. Rand  
1 1824 M. R. Keenan  
1 2361 T. J. Williams  
1 2561 F. M. Bacon  
1 2561 D. K. Morgan  
1 2561 C. E. Spenser  
1 2561 W. E. Newman  
5 3141 S. A. Landenberger  
8 <sup>3145</sup> ~~3141~~ <sup>Document Processing</sup> C. L. Ward for DOE/OSTI  
3 3151 ~~W. I. Klein~~ <sup>G. L. Esch (acting)</sup>  
1 6612 A. Smith  
1 7220 R. R. Prairie  
1 7223 R. G. Easterling  
1 7470 J. A. Sayre (acting)  
1 7472 J. A. Sayre  
1 7472 H. W. Arris  
1 7472 S. N. Hoier  
1 7476 F. P. Gerstle, Jr.  
1 8311 D. L. Lindner  
1 8311 R. E. Gott  
1 8524 J. A. Wackerly  
12 7472 K. B. Wischmann  
12 7223 E. V. Thomas

Allied Signal

R. S. Sanders, D/837  
F. N. Larsen, D/837  
H. M. McIlroy, D/837  
T. E. Neet, D/837  
E. W. Grotheer, D/837

Mound Labs

J. DiCharo

GEND

B. J. Wells  
R. Antepencko  
R. Solingio

Oak Ridge

G. Dorsey



8232-2/071268



00000001 -



8232-2/071268



00000001 -



8232-2/071268



00000001 -

