OPTIMAL DESIGN OF APP MODIFIED BITUMEN BINDERS FOR ROOFING APPLICATIONS USE OF EXPERIMENTAL DESIGN TECHNIQUES

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Because of constantly increasing economical constraints, the roofing industry is looking more than ever to optimize its products. In practice, this means that the manufacturers of polymer modified roofing sheets aim to use the lowest amount of polymer possible without jeopardizing membrane characteristics.

This is particularly true for polypropylene modified binders for which various types of recycled polymers may be used. The quality of these polymers is variable and their impact on final performance is not always well-understood. To guarantee their specifications, manufacturers are often obliged to use an excess of polymers and to perform frequent and costly analysis.

Optimization is possible if one can make use of reliable models relating polymer and bitumen characteristics to the performance of the final blend. Because of the large number of involved parameters, such models are, however, difficult to establish. This is where experimental design can be a key asset. A correctly designed experimental plan will ensure that the right factors and their possible interactions are taken into account. It also limits the number of experiments to be performed to a strict minimum.

This technique has been applied for a systematic evaluation of different bitumens in combination with a mixture of three types of polypropylene. The study was based on six variables (types of polymer) at two levels (polymer content). This design matrix has been repeated for three bitumens differing by their composition. All blends have been assessed on the major performance criteria (i.e., softening point, penetration, viscosity, and cold-temperature flexibility before and after aging).

The investigation has indeed evidenced how the different polymer and bitumen types affect these criteria. This opens at least two gates for a better control of both performance and costs. On the one hand, it is possible to optimize the final product with an adequate selection of the different blend components, and on the other hand, one can better accommodate and correct the variations of these components.

KEYWORDS

APP modification, experimental design, optimization, roofing, statistics.

INTRODUCTION

The roofing industry, confronted with constantly increasing economical constraints, is looking more than ever to optimize its products. In practice, this means that the manufacturers of polymer modified roofing sheets aim to use lowest amount of polymer possible without jeopardizing membrane characteristics. Unlike the styrene-butadiene-styrene- (SBS-) based market, manufacturers of polypropylene modified binders apply, in general, various types of recycled polymer grades. The quality of such products is variable, so one can clearly understand that this could have a significant impact on the final properties of the membrane. The roofing manufacturer, who tries to guarantee his product quality according to the various specifications, is, therefore, often obliged to use an excess of polymer and to perform frequent and costly analysis. In general, product engineers are looking for a tool that could facilitate this optimization process and are constantly looking for reliable models relating polymer and bitumen characteristics to the final blend properties. Because there is a large number of parameters involved in this process, models are not easy to establish. Hence, most optimization studies performed by roofing manufacturers are less detailed test programs looking at the behavior of a mixture when one or more of the components vary. It is in this process that experimental design can be a key asset: a correctly designed experimental plan ensures that the most important factors and the possible interactions are taken into account while it also limits the number of experiments to be performed. Actually, one can say that a good experimental plan limits the amount of work to a minimum, but yields a maximum amount of information.

EXPERIMENTAL DESIGN

Background

To draw conclusions about cause and effect, a researcher may choose from a whole series of mathematical and statistical

methods and, therefore, risks using a method that is less appropriate for the problem being studied.

The performance of a process or experiment can often be described in terms of one or more responses or dependent variables. In order to control a process, it may be useful to understand the effect on a response of one or more independent variables.

Mathematically, this can be written equation:

$$y=f(x_1,x_2,.....,x_n)$$

y=dependent variable

x_i=independent variables

The effect of an independent variable is the change in response that occurs when the level of the independent variable is changed. In designing an experiment, one has to decide how many trials one wishes to perform and which combinations of variables will be used for each trial. A problem is that there may be no limit to the number of trials one could perform, while on the other hand, there might be a limitation on the conclusions one might draw from the different results after carrying out the experiment.

The following example can help to explain this better:

Suppose one wishes to study a problem having six independent variables, and one decides to use five different values for each independent variable; this means that one has to perform 5^6=15,625 experiments to assess this problem fully. As one can imagine, such a test program is not feasible.

Therefore, it is necessary to reduce such test programs, and for doing that, one has only two possibilities:

- Reducing the number of values for the independent variables—if one takes three values instead of five for each variable, this still leads to a test program with 3^6=729 experiments. In the extreme, one could apply only two values, but in such a case, one would lose all reliability.
- Reducing the number of independent variables—This option also reduces the number of experiments to be performed, but as one can easily understand, it will not give a better understanding of the problem (because certain influencing parameters are not taken into account), and, therefore, this "solution" is inadvisable.

One way to put all variables into a relationship is to establish simple regression equations, linking the various variables (e.g., the weight of a component) with the dependent variables (e.g., a product parameter). If more variables have an impact on the result, it may be useful to apply multiple regression techniques.

It is possible that any of the various resulting equations will offer a useful model, but sometimes it is impossible to reconcile the conflicting information that results when all equations are used at the same time.

Multiple regression analysis is a useful tool as long as one takes great care with the choice of levels of independent variables. Before using multiple regression, one has to examine possible correlations between the input variables because such correlations may cause the regression technique to give misleading results.

This is where experimental design comes into the picture; it allows the effects of the independent variables (at all value levels) to be assessed in an intelligent way by:

- reducing the number of tests;
- taking into account correlations between the input variables;
- optimizing of the test program.

To achieve this, one might use a factorial design at two levels, either in the form of a full experimental plan or in the form of a fractional plan (which allows the number of experiments to be reduced even more). A complete factorial design for a problem with six independent variables (at two levels) needs only 2^6=64 experiments, which is a significant reduction of the number of tests. For small test programs, a full factorial design can be implemented, but for large and complex systems, it is not always feasible to perform the full set of experiments. Fortunately, it is possible to reduce the size of a factorial experiment without sacrificing the most important estimates.

Because the response of a bitumen to a certain polymer formulation depends on the polymer composition and/or percentage and also on the molecular characteristics of the bituminous binder, it is not easy to screen the blend characteristics for other polymer compositions without performing a large series of tests. For such problems, experimental design techniques could prove to be useful as one might expect certain interactions between the different polymer components.[1] To help set up the program and to design the reduced experimental matrix, the advice of a consultant was obtained (D. Caby, France).

Experimental Design Matrix

Bituminous binders

Even based on an optimized design matrix, a lot of blends must be prepared to evaluate the characteristics of a bitumen/polymer combination; in fact, one has to repeat the test program for every other bituminous binder. In order to limit the number and to avoid unnecessary work, a preliminary bitumen screening was performed to select the most optimal set of bituminous binders for further evaluation. Previously, screening of bitumen for modification with polypropylenes was done by making blends, and the results were compared with existing data (see Table 1). A standard polymer mixture

Base Bitumen	Viscosity	Pen 25°C	Pen 60°C	R&B	Flexibility	Disper sion
Bitumen A	5580	41	144	152	-13	average
Bitumen B	3820	43	182	154	-12	fine
Bitumen C	4440	37	152	156	-14	fine
Bitumen D	3000	48	200	152	-9	average
Bitumen E	3320	44	180	153	-10	average

Table 1. Preliminary screening of bitumen (blended with a standard polymer mixture).

	Bitumen A	Bitumen B	Bitumen C
Softening point R&B °C	44.2	43.7	44.0
Penetration at 25°C mm/10	113	115	112
Viscosity at 135°C mm^2/s	325	280	290
IATROSCAN			
asphaltenes %	21.8	17.2	16.3
resins %	22.2	21.1	19.9
aromatics %	47.4	51.2	55.7
saturates %	8.6	10.5	8.1

Table 2. Bitumen analysis data.

was blended into the bitumen and analyzed according to the important product characteristics (all blends are made with a constant percentage of filler).

Only the three most promising bitumens were selected for this study: A, B, and C.

These three binders, having a similar penetration, differ significantly in composition (see Table 2). One of the methods of characterizing the bitumen composition is the Iatroscan method. Several authors have already underlined the relation that exists between the generic composition and the properties of the bitumen/polymer mix.[2,3]

The Iatroscan analysis, a method based on thin layer chromatography combined with a flame ionization detection system (FID), provides a particularly quick and suitable method for determining the generic composition of a bituminous binder.

The system utilizes an adsorption medium in the form of a sintered layer of silica gel bonded to a thin quartz rod (i.e., chromarod). A three-stage solvent development is employed to separate the bitumen into its four main constituents: asphaltenes, resins, aromatics, and saturates. After development, the chromarods are scanned through an FID to burn off the separated components, and the detected signal is used to calculate the generic composition of the bituminous binder.

Selection of polymers

The polymer composition used in the roofing industry is, in general, not based on one single product, but is mostly a blend of different APP, IPP, and copolymers coming from different sources. With a significant supply of recycled products on the market (products having an interesting price advantage), most polymer blends applied by roofing manufacturers are a mixture of high- and low-grade viscosity polymers (see Table 3). By selecting polymers and adapting the individual percentages in the polymer blend, it is possible to achieve a more or less constant polymer quality.

Nevertheless, the often variable quality of the base polymers can have a significant impact on the final properties of the modified binder, and therefore, one can easily understand that keeping the polymer blend's quality as constant as possible requires frequent and costly analysis and checks.

With the advantage of the experimental design approach, one could assess the impacts of a changing polymer formulation without performing a great number of tests.

IPP	Very hard and tough polymer with a MI (Melt Flow Index) ± 8
APP1	$Visc_{180°C}$ ~ 500 cP and $Pen_{25°C}$ ~ 10-20 mm/10
APP2	$Visc_{180°C}$ ~ 3000 cP and $Pen_{25°C}$ 10 mm/10
COPO1	$Visc_{180°C}$ ~ 1 000 000 cP - good flexibility at low temperatures
COPO2	$Visc_{180°C}$ ~ 700 000 cP - harder and less flexible than COPO1
COPO3	$Visc_{180°C}$ ~ 800 000 cP - harder and less flexible that COPO2

Table 3. Polymer characteristics.

Experimental design formulations and design matrix

The base experimental design is based on two levels for six independent variables (IPP, APP1, APP2, COPO1, COPO2, and COPO3). Therefore 2^6 blends must be made, and for each blend, various characteristics are measured:

- softening point ring & ball
- penetration at 25°C (77°F) and 60°C (140°F)
- viscosity at 180°C (356°F)
- cold-temperature flexibility (before and after aging)

The actual percentage values of the independent variables and their variance are, in most cases, derived from experience as one often has an idea of the possible variations in these variables. If not, it is still possible to apply the experimental design approach, but the possibilities to reduce the factorial experiment are much more limited. In this appraisal of the experimental design method, certain percentages are applied for the various polymer grades. As one can imagine, exact figures cannot be mentioned, but to have an idea of the polymer percentages used in this study, the following information can be given:

- maximum polymer level: 26 percent
- center points: 22.5 percent
- minimum polymer level: 19 percent

These figures show that for the center point (average polymer level) blends, a total polymer percentage of 22.5 percent is used (the remaining 77.5 percent being bitumen and filler). This polymer percentage has been chosen in order to obtain a continuous polymer phase in which the bitumen is dispersed (the polymer distribution can be monitored by means of fluorescence microscopy) to give the modified product the required quality.

In production, a blend of bitumen and each type of polymer from Table 3 is manufactured first, followed by an addition of the filler. In this case, the filler content has been held constant. Applying a full experimental plan (this is 2^6=64 experiments) for each bitumen means that because three bituminous binders were selected, a total of 192 experiments have to be prepared (which still represents a considerable amount of work).

Preliminary studies already performed by the authors have shown that interactions between three polymers (and more) can be neglected. As a result of this, the full factorial design can be transformed into a 2^{6-2} fractional plan, which reduces the total number of tests to 16. Having selected three bitumens, this gives a total of 48 blends, which is four times less than the full plan. Finally, the model as (partly) presented in Table 4 has been established (only the first week test program is given; for the following three weeks, a similar test scheme is performed).

Analysis week 1							
	Bitume n	IPP	APP1	APP2	COPO1	COPO2	COPO3
101	A	0	0	0	0	0	0
102	B	1	1	1	1	1	1
103	A	1	1	-1	-1	-1	1
104	C	-1	1	1	1	-1	1
105	C	-1	1	-1	1	1	-1
106	A	-1	1	1	-1	-1	-1
107	B	-1	-1	1	1	1	-1
108	B	0	0	0	0	0	0
109	C	1	-1	1	1	-1	-1
110	A	1	1	1	1	1	1
111	B	-1	-1	1	-1	1	1
112	B	-1	1	1	1	-1	1
113	C	-1	-1	1	-1	1	1
114	A	1	-1	-1	1	1	1
115	C	0	0	0	0	0	0

Table 4. Experimental design matrix.

The figures given in Table 4 must be understood as follows:

- *0* indicates that the center polymer percentage is used (e.g., 3 percent IPP)
- *-1* indicates the minimum polymer percentage (e.g., 2.5 percent IPP)
- *+1* indicates the maximum polymer percentage (e.g., 3.5 percent IPP)

Because all of the different blends were made in a time period of four weeks, it was possible to include reruns of some blends, with a polymer composition identical to the reference level (also called center points). These duplicates provide information on the reproducibility of the test method, but they also indicate whether week-by-week time- dependent effects occur, which could influence the other results of the various blends. If time-dependent effects are noticed, it might be necessary to introduce a statistical compensation of the measured analysis data. One extra additional test with a center point formulation was performed for each bitumen each week; this increased the total number of blends by 12. As such, the whole test program included 60 blends. By performing duplicates on the center points during this four-week testing period, the authors could calculate a deviation factor for a specific test method. A similar factor is also obtained in the analysis of that test procedure when taking into account all performed blends. The results of the various reruns can also be used to verify the assumption of a linear model.

RESULTS: DISCUSSION

Interpretation of the Statistical Data

The first step is to compare the standard deviation at the center points with the residuals. A difference that is too high indicates that the researcher is not fully controlling the test protocols of that specific test. It is sometimes difficult to decide how much data may deviate because the results often depend on the accuracy of the various test methods, and therefore, a good experimental protocol is required in order obtain reliable test results (see Table 5).

In this study, all statistical evaluations and even the design of the experimental plan were performed with the help of a statistical software package, StatGraphics Plus 1.1 (for Windows). Applying the multiple regression analysis procedure, one can calculate the equation parameters for the various input variables (polymer components) as a function of the observed results. Because every initial evaluation includes all the variables, some variables/interactions have no significant effect on the results. Therefore, the next evaluation, and any following, included only these variables with a sufficient significance level; results of this evaluation (for each binder) are presented in Attachment 1.

The objective of this process is to understand the impact of each individual polymer component with respect to the chosen bitumen binder and to evaluate the effect of a percentage change in polymer composition on the final blend characteristics.

Softening Point Ring and Ball

Binder A: R&B=150.4 + 0.68*IPP + 0.19*COPO2 - 0.22*IPP*APP2 - 0.19*IPP*COPO3

Binder B: R&B=152.2 + 0.73*IPP

Binder C: R&B=153.1 + 0.92*IPP + 0.33*APP1 + 0.24*COPO3 + 0.22*IPP*COPO2

The softening point of the modified binders is almost solely determined by the IPP component in the polymer composition. When explaining the influence of a component this way, one ought not forget that the other polymer components also play a role.

But as long as one changes the percentages of these components within the limits of the proposed experimental plan, one will not notice a significant effect on the dependent variable (in this case, the softening point R&B of the blend). Increasing the IPP percentage will have a significant impact on the blend softening point. The constant factor in the model gives an estimation of the dependent variable for a center point polymer formulation. The compatibility of a polymer mixture toward a specific bitumen binder can be evaluated by its effect on the different blend characteristics: a low softening point is often an indication of a poor polymer matrix and, as such, a low modification level.

Binder C, exhibiting a rather high R&B figure, especially compared to Binder A, illustrates this rather well: not only is the constant factor the highest of all tested binders, but the impact on the R&B of an IPP percentage increase is also much higher. In addition, these equations teach that, for hav-

	Center Points	Binder A	Binder B	Binder C
Softening point R&B	0.5525	0.4026	0.8280	0.5022
Penetration 25°C	1.1615	1.2573	1.1685	1.0996
Penetration 60°C	3.6139	8.9956	3.6446	6.5016
Ln (Viscosity 180°C)	0.0209	0.0258	0.0329	0.0234
Cold-temperature flexibility	0.8334	0.9562	1.2012	1.3764
Cold-temperature flexibility (after 4 weeks at 80°C)	1.5184	1.0312	0.8263	1.5232

Table 5. Reproducibility factor for the various test procedures.

ing a certain R&B level, one could apply a lower IPP percentage when using Binder C (or Binder B), than when using Binder A.

One could find that for some binders, other polymer components (or polymer interactions) play a role, but the effect of these is marginal compared to the effect of IPP.

Penetration at 25°C (77°F) and 60°C (140°F)

Binder A: Pen25=41 - 3.21*IPP - 0.78*COPO3
Pen60 =159 - 18.5*IPP

Binder B: Pen25=38 - 2.36*IPP - 0.69*APP2
Pen60 = 187 - 29*IPP - 7*APP1 - 3.1*APP2 - 4*COPO1 - 4.5*COPO2 - 2.4*COPO3 + 3.38*IPP*APP1 + 2.17*IPP*COPO1 + 4.58*IPP*COPO2

Binder C: Pen25=35 - 2.13*IPP - 0.56*APP1
Pen60 = 147 - 19.5*IPP - 5.9*APP1 - 0.14*IPP*APP1

As for the softening point, the IPP percentage is the most decisive factor influencing the penetration of the blend, whether at 25°C (77°F) or at 60°C (140°F). At 25°C (77°F), the differences in penetration are rather small and are in line with the softening point (a higher softening point corresponds, in general, with a lower penetration), but at 60°C (140°F) differences are more discriminating. At 60°C (140°F), Binder B exhibits the highest penetration, which could be a disadvantage in certain situations, but on the other hand, increasing the IPP percentage has a much more important effect. It is also remarkable that for Binder A and Binder C, primarily only the IPP compound has an effect on the penetration, while for Binder B, several other polymer components have an impact.

If a low penetration at 60°C (140°F) is required, the use of Binder B might be less appropriate (unless one increases the IPP content). However, as the penetration for this binder is not influenced only by the percentage of the IPP component, a compromise can be found by changing the percentage of some other polymers.

Brookfield viscosity at 80°C (176°F)

Binder A: Ln (Visc180)=7.82 + 0.047*IPP + 0.032*APP1 + 0.13*COPO1 + 0.091*COPO2 + 0.093*COPO3

Binder B: Ln (Visc180)=7.60 + 0.076*IPP + 0.038*APP1 + 0.025*APP2 + 0.143*COPO1 + 0.094*COPO2 + 0.099*COPO3 - 0.016*IPP*COPO2 - 0.014*IPP*COPO3

Binder C: Ln (Visc180)=7.89 + 0.087*IPP + 0.028*APP1 + 0.13*COPO1 + 0.096*COPO2 + 0.083*COPO3 + 0.011*IPP*APP1 + 0.011*IPP*COPO2

The viscosity, measured by means of a Brookfield rotational viscosimeter at 180°C (356°F) (spindle 28, speed 50 rpm), is an important parameter to evaluate the workability of the modified blend. Products with too high a viscosity are difficult to apply on the carrier layer, but viscosities that are too low cause dripping (and, thus, loss) of the polymer/bitumen mixture. Products having a high viscosity are also less pumpable and can, therefore, provoke problems in certain types of production units. It should be borne in mind that viscosity is also related to various other production factors, which makes it difficult to predict which viscosity figure should be most optimal.

Having a high viscosity, one might increase temperature to obtain an acceptable impregnation of the carrier, but on the other hand, these higher temperatures could provoke unwanted production problems.

The viscosity of a polymer blend is determined by the base bitumen and, of course, by the polymer composition. In general, one observes that the ranking of viscosity runs parallel with that of the penetration at 60°C (140°F). Binder B exhibits the lowest viscosity.

Unlike parameters such as softening point and penetration, for which only a variation of the IPP results in significant changes, the viscosity at 180°C (356°F) is influenced by nearly all polymer constituents (and polymer interactions). The most important, however, are variations in IPP and copolymer percentages. Adapting the APP percentages does not result in a significant viscosity change. All of this does not mean that one can reduce the APP percentage indefinitely; all of the calculated effects are based on the influence within the designed testing frame.

The lower viscosity values found for Binder B can be explained by the different chemical composition of this specific binder: the lower asphaltenes content, meaning a higher maltenes content, suggests that more oils are available to disperse the polymer and the filler.

Eventually, a higher amount of maltenes could be absorbed by the filler (wetting), which could mean that it is not really necessary to increase the IPP content in the polymer mixture. An increase of the filler content might already meet the desired product specifications, without jeopardizing other product characteristics. In this regard, the lower viscosity of Binder B is an asset rather than a disadvantage.

This lower viscosity of Binder B-based blends is not necessarily a negative point: in this study, the filler content was kept constant at a given value (though it is also possible to consider the filler percentage as an independent variable). Up to a certain level, one can increase the filler content without significantly influencing other parameters (such as flexibility, etc.).

Flexibility Cold Bending BDA fresh and aged [four weeks/80°C (176°F)] samples

Binder A: BDA=-16.2 + 0.50*IPP - 0.88*COPO1 - 0.88*COPO2 - 0.75*COPO3 + 0.63*IPP*COPO2 BDA4=-12.7 + 0.94*IPP - 1.06*COPO1 - 0.69*COPO3 - 0.56*IPP*APP1 + 0.81*IPP*COPO1

Binder B: BDA=-14.1 - 0.63*APP1 - 0.75*COPO1 - 0.88*COPO3 + 0.75*IPP*APP1 + 0.63*IPP*COPO1 BDA4=-9.8 + 1.69*IPP - 1.19*APP1 - 0.94*COPO1 - 1.06*COPO2 - 0.44*COPO3 + 0.69*IPP*APP2

Binder C: BDA=-15 + 0.69*IPP - 0.69*APP2 - 0.81*COPO1 BDA4=-10.2 + 1.44*IPP - 0.94*APP2 - 0.94*COPO1 - 1.19*COPO2

The cold-temperature flexibility also showed differences between the tested base bituminous binders, ranking Binder A as being better than both of the other grades.

All polymer constituents play a relatively important role in the cold-temperature behavior of the polymer mixture. The APPs and the copolymers have a positive effect and IPP exhibits a negative effect on cold-temperature flexibility. Actually, this is nothing more than a confirmation of the intrinsic nature of the various polymers the amorphous nature of APP [with glass transition temperatures below -10°C (14°F)] and the semi-crystalline nature of IPP.

This means that if one likes to improve the penetration

and the viscosity of a binder by increasing the IPP percentage, cold-bending temperatures will become negatively affected (but this is directly related to the nature of the IPP polymer). After aging [four weeks at 80°C (176°F)], Binder B and Binder C behave similarly while Binder A seems to be less affected by this aging procedure.

Results: But what can we learn?

In this study, applying the experimental design approach to obtain as much information as possible for a limited number of tests, the effect of the polymer mix composition has been evaluated for different types of bituminous binder. This statistical approach results in several useful models, but it is still the researcher who has to make the final interpretation of the various information given by these equations or models.

By assessing the effects of a change in the polymer mix component percentage (for different bitumen binders), the researcher has a tool to optimize the final polymer mix composition for a specific binder type. It is true that an optimal polymer mixture (composition and percentage) depends of several factors—much more than one can include in a model without making it too complex. Knowing how each individual polymer affects the final product parameters and which polymers do not significantly affect these when lowered in percentage, manufacturers can change the polymer formulation knowing in advance how big the impact on the final membrane will be, as long as one takes into account the fact that data for a production batch are normally different than data for a laboratory mixture.

Applying the equations, presented on the preceding pages, it becomes fairly easy to predict the final characteristics of a bitumen/polymer/filler blend (laboratory-made), knowing the various polymer percentages.

For example, if one should use the polymer amounts corresponding with the center formulation, one might expect following figures (for a blend with Binder A):

- softening point R&B: 150.4°C (303°F)
- penetration at 25C (77°F): 41 mm/10 (1.61 inches/10)
- penetration at 60°C (140°F): 159 mm/10 (6.26 inches/10)
- viscosity at 180°C (356°F): 2490 cP
- flexibility BDA: -16.2°C (3°F)
- flexibility BDA after four weeks of aging: -12.7°C (9°F)

On the other hand, such models provide a handy way to evaluate other alternative types of polymers. If the producer likes to change, for example, COPO1 by an alternative grade, he has only to make a small number of blends and evaluate the data with the standard formulations. This means that evaluating other polymer grades, and even bitumen grades, becomes much easier and requires less time, because the number of experiments to be performed can be limited to a strict minimum. A more detailed study should even allow the establishment of a general formulation that takes into account the effects which occur when one adds or changes a polymer grade.

All these various equations are also useful to correct a polymer composition—for a given binder—if one or more of the characteristics are out of the required specifications.

For example, imagine that a modified binder should fulfill following requirements:

- penetration at 25°C (77°F): 20 to 40 mm/10 (0.78 to 1.57 inches/10)
- penetration at 60°C (140°F): 100 to 150 mm/10 (3.94 to 5.9 inches/10)
- flexibility BDA: <-15°C (5°F)

The values for the standard (center) polymer formulation show that this composition will result in a penetration at 25°C (77°F) and 60°C (140°F) of 41 mm/10 (1.61 inches/10) and 159 mm/106 (26 inches/10), respectively. When adding an additional 0.5 percent IPP to the mixture, one can calculate the effects of this polymer change on the various product parameters by using the different equations. Doing this, one finds the following characteristics for that composition:

- softening point R&B: 151°C (304°F)
- penetration at 25°C (77°F): 37.8 mm/10 (1.49 inches/10)
- penetration at 60°C (140°F): 140.5 mm/10(5.53 inches/10)
- viscosity at 180°C (356°F): 2610 cP
- flexibility BDA: -15.7°C (4°F)
- flexibility BDA after four weeks of aging: -11.8°C (11°F)

Having calculated these estimated figures, the researcher can without any risk approve this change in composition; the "non-conform" characteristics are within specification, while all other characteristics are not drastically changed.

One could even think of one step further: including these, or similar, equations into a computer program will provide a good tool for the optimization of the blend formulation. When entering blend specifications, the program could calculate the most optimal formulation, taking into account parameters such as polymer cost, polymer quality, and even process characteristics (ease of working with a certain polymer, etc.)

CONCLUSIONS

The roofing industry of today is confronted with serious economical constraints; on the one hand, clients like to save money, but on the other hand, they demand the highest quality. Therefore, manufacturers are looking more than ever to optimize their products. Very often, this means that they try to reduce the polymer content as much as possible without jeopardizing the membrane characteristics.

For polypropylene-polymer-based roof membranes, more and more recycled products are applied, and often, these products are of an uncertain quality. To guarantee specifications, frequent checks must be performed, which could increase product cost. Therefore, mathematical models relating polymer and bitumen characteristics to performance could be a great asset. In this study, one of the possible models—experimental design—has been evaluated. The next step should try to relate bitumen compositional data and the physical/chemical nature of the various polymers with the product characteristics, resulting in equations that are independent of the bitumen binder.

Because the response of a bitumen to a certain polymer formulation depends on the polymer composition and/or percentage and also on the molecular characteristics of the bituminous binder, a large series of tests must be performed

to screen the effect of all mentioned parameters on the final properties. One could even integrate effects such as cost, production, and workability in the optimization procedure. Experimental design techniques could help to set up a good and limited test program—performing only the number of tests necessary to draw reliable conclusions.

In this study, the effect of the polymer composition has been evaluated for different types of bituminous binder, and a mathematical model expressing the effect of the polymer composition on the various product parameters has been calculated. Such equations are of significant meaning for the researcher because they can predict the effects when changing the polymer composition. By knowing in advance how each polymer behaves for a certain binder, manufacturers can adapt polymer formulations without the need of performing a whole series of tests. It is even possible to screen other grades of polymer, by performing only a small number of tests and fitting the data into the existing model.

Even if the results of this study are promising and show the potential of the experimental design method as a way to gain better insight into all parameters involved in the production of roof membranes, it is still the researcher who has to interpret the data and apply these in the production process.

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ATTACHMENT 1. STATISTICAL PARAMETERS FOR THE INDEPENDENT VARIABLES

Binder A.	INFLUENCE OF POLYMERS						
	Const.	IPP	APP1	APP2	COPO1	COPO2	COPO3
R&B	150.42	0.6813				0.1938	
Penetration at 25 C	40.968	-3.2163					-0.7788
Penetration at 60 C	158.94	-18.523					
Ln(Visco180 C)	7.823	0.04688	0.03179		0.12856	0.09097	0.093
Cold Bending BDA	-16.15	0.5			-0.875	-0.875	-0.75
Cold Bending BDA (4 weeks)	-12.7	0.9375			-1.0625		-0.6875

Binder A.	POLYMER INTERACTIONS				
	IPP*	IPP*	IPP*	IPP*	IPP*
	APP1	APP2	COPO1	COPO2	COPO3
R&B		-0.2188		-0.1938	
Penetration at 25 C					
Penetration at 60 C					
Ln(Visco180 C)					
Cold Bending BDA			0.625		
Cold Bending BDA (4 weeks)	-0.5625		0.8125		

Binder B.	POLYMER INTERACTIONS				
	IPP*	IPP*	IPP*	IPP*	IPP*
	APP1	APP2	COPO1	COPO2	COPO3
R&B					
Penetration at 25 C					
Penetration at 60 C	3.3756		2.167	4.583	
Ln(Visco180 C)				-0.0158	-0.0142
Cold Bending BDA	0.75		0.625		
Cold Bending BDA (4 weeks)		0.6875			

Binder B.	INFLUENCE OF POLYMERS						
	Const.	IPP	APP1	APP2	COPO1	COPO2	COPO3
R&B	152.15	0.725					
Penetration at 25 C	37.981	-2.355		-0.69125			
Penetration at 60 C	186.716	-29.0001	-6.999	-3.12562	-3.9581	-4.5419	-2.4169
Ln(Visco180 C)	7.604	0.7586	0.0375	0.0245	0.1434	0.094	0.0992
Cold Bending BDA	-14.05		-0.625		-0.750		-0.875
Cold Bending BDA (4 weeks)	-9.75	1.6875	-1.1875		-0.9375	-1.0625	-0.4375

Binder C.	INFLUENCE OF POLYMERS						
Const.	IPP	APP1	APP2	COPO1	COPO2	COPO3	
R&B	153.1	0.9188	0.3313				0.2438
Penetration at 25 C	34.983	-2.125	-0.5588				
Penetration at 60 C	146.795	-19.474	-5.848				
Ln(Visco180 C)	7.895	0.0873	0.0281		0.1288	0.0963	0.0826
Cold Bending BDA	-15.0	0.6875		-0.6875	-0.8125		
Cold Bending BDA (4 weeks)	-10.15	1.438		-0.9375	-0.9375	-1.188	

Binder C.	POLYMER INTERACTIONS				
	IPP*	IPP*	IPP*	IPP*	IPP*
	APP1	APP2	COPO1	COPO2	COPO3
R&B				0.2188	
Penetration at 25 C					
Penetration at 60 C	-0.1406				
Ln(Visco180 C)	0.011			0.0105	
Cold Bending BDA					
Cold Bending BDA (4 weeks)					