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Application of chemometrics to the production of friction materials: Analysis of previous data and search of new formulations

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Abstract

A friction material is a composite containing up to 18 different components which can be chosen among a large number of possible raw materials having different characteristics, like graphite, sulphides, metals, fibers, rubbers, resins and fillers. The requested form is obtained by moulding at controlled pressure and temperature. In order to prepare new formulations having good performances, the problem is to choose the best raw materials and to mix them in the optimal proportions. Since the quality of a formulation is not expressed by a single value, but several responses have to be taken into account at the same time (friction coefficient, comfort, wear, etc.), the analysis of the data obtained from different formulations is quite difficult. In this study an approach to the analysis of this kind of data is presented, in order to evaluate different products on the basis of a small number of 'quality indicators'. The techniques of experimental design are successfully applied in order to investigate the effect of process variables on the performances of the product and to perform a screening of the raw materials for new optimal formulations.

Keywords: Experimental design; Mixture design; Friction material; Multicriteria optimization

1. Introduction

A friction material is a very complex product, since it is a mixture of more than 15 components, which can be chosen among more than 800 possible raw materials. The performances of the product depend on the process variables and on the percentage of the raw materials selected. Moreover, it must be considered that there are constraints, i.e. not all blends among the components are possible.

Another important problem is that the quality of a formulation is not expressed by a single value, but several responses have to be taken into account at the same time (friction coefficient, comfort, wear, etc.). Furthermore, different customers give them different importance.

For the complexity of the problem, a 'one variable at a time' experimentation (OVAT design), where the compounder changes the components, their proportions and the setting of the process variables,

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requires many experiments and cannot assure to find new good formulations. In general, a strategy consisting in performing small variations around an acceptable end product is applied.

In this study an approach to the analysis of data from different friction materials in order to obtain the maximum information for characterizing the products is presented and a strategy for searching new formulations is successfully employed.

2. Analysis of previous data

The most important response for friction material is the friction level, which should comply with the customer request. It is usually evaluated by dynamometer-bench, where several stop conditions are simulated, at different speed, temperature and pressure. The classical way to compare the results of different dyno-tests is to draw several graphs showing the behaviour of the friction material at the different conditions. Fig. 1 reports an example of these curves for a certain product. The comparison between these graphs is quite difficult and a good analysis of the results is not possible.

Data of this kind can be handled by chemometric techniques. After a certain number of formulations have undergone the dyno-test, a data matrix is obtained, where each row (object) is one friction material and each column (variable) is the friction level measured at the corresponding conditions. Chemometric methods are very useful tools for visualizing the information contained in the data, for identifying groups of similar products, and for finding relationships between the variables.

The computations required for the work reported in this paper were performed by means of a package for multivariate analysis, PARVUS [1], and a package for experimental design, NEMROD [2].

Since each test is characterized by several tens of variables and these variables are highly correlated, principal component analysis (PCA) can be applied in order to reduce the dimensionality of data and to describe the different formulations by means of a few new uncorrelated variables, linear combinations of the original ones. PCA applied to several data sets showed that usually 2 or 3 principal components are significant and easily interpretable. A very simple comparison of different formulations is allowed.

The following example refers to 26 formulas de-

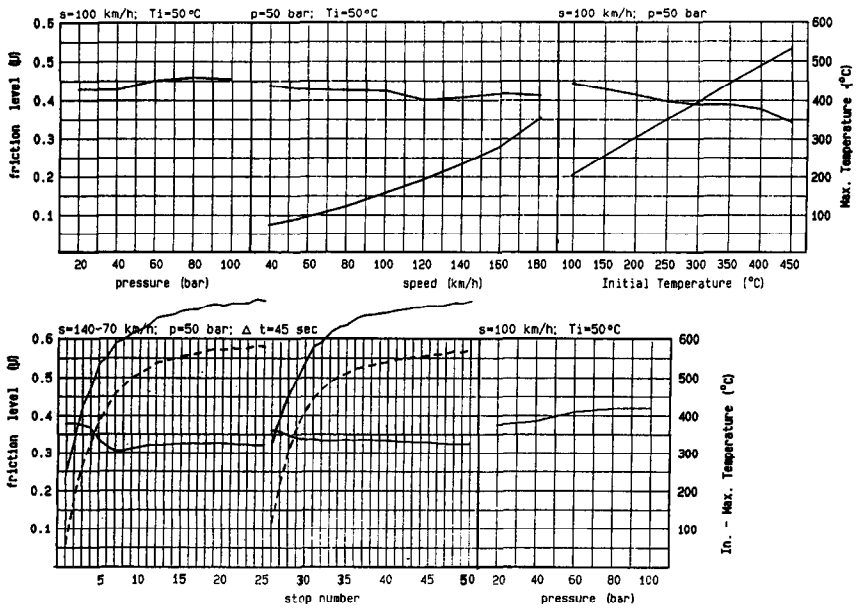

Fig. 1. An example of plots showing the results of a dyno-test performed on a certain product.

scribed by the friction coefficient measured at 55 different brakes in the dyno-test. The first two principal components retain 70.6% of the total variance.

After raw varimax rotation of loadings [3], it can be noted that varivector 1 describes the performance at high temperatures, and varivector 2 describes the performance at low temperatures (Fig. 2). In fact, the group of brakes at high temperature (variables 20–55) has important loadings on varivector 1, while the group of brakes at low temperature (variables 1–19) has important loadings on varivector 2.

It is important to note that scores on varivectors 1 and 2, after range scaling within the range 0 to 100, can be used as 'quality indicators' of the products instead of the original variables: score on varivector 1 is the most important indicator.

However, performing a dyno-test is rather expensive and time-consuming; in addition to the variables from the dyno-test, a set of physical properties, cheaper and faster to be obtained, can be measured on the products.

Especially during the first phase of the study, it would be useful to perform a general screening of new formulations simply on the basis of physical properties, so that only the promising products have to undergo the dyno-test.

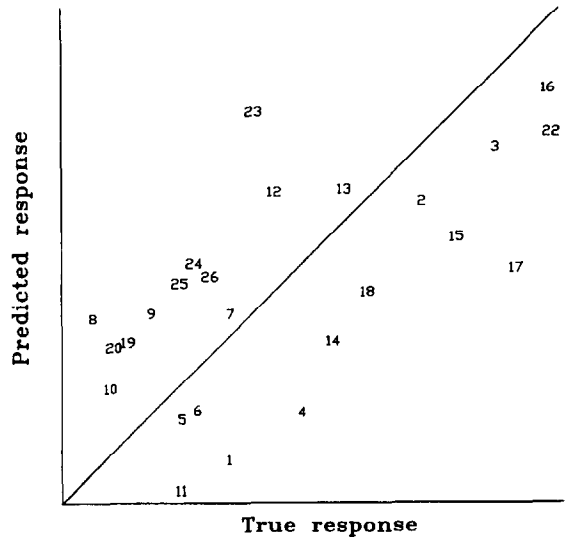

Fig. 3. PLS plot: true response versus predicted response.

Partial least squares (PLS) regression [4] was applied in order to find a relationship between a block of 7 physical variables and the score on varivector 1 for the same 26 products of the example shown before. The PLS model with 2 latent variables explains 41.8% of the variance in prediction: cross-validation

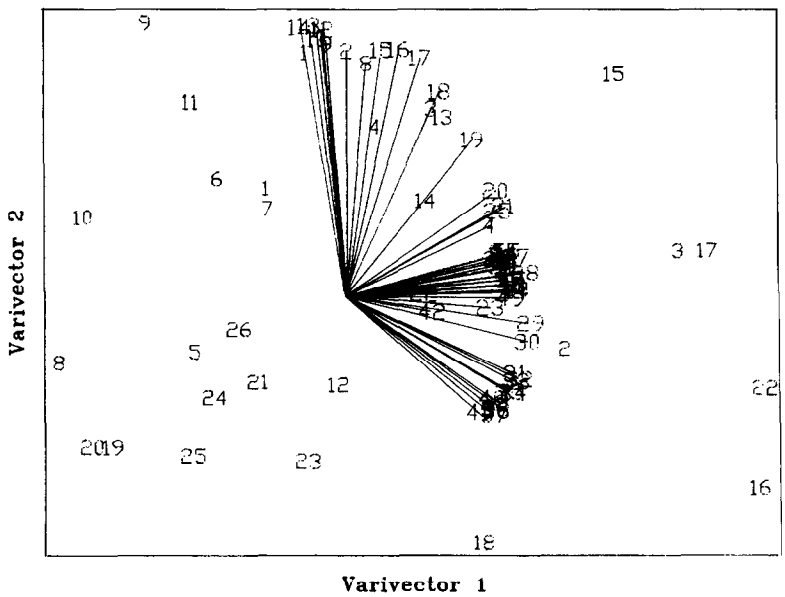

Fig. 2. Varivector biplot: scores of the objects and loadings of the variables.

Table 1
Study of process variables: experimental domain

Factors	Level 1	Level 2	Level 3	Level 4
A	small	medium	large	mixture of small and large
B	type a	type b	–	–
C	0.5:1	1:1	2:1	3:1
D	2:1	3:1	4:1	5:1
E	type a	type b	–	–
F	type a	type b	–	–
G	small	large	–	–

with the leave-one-out procedure was used. Fig. 3 shows the relationship between the true response and the predicted response. A satisfying correlation was found, as indicated by the predictive ability of the model: even if the value of variance explained seems to be quite low, however it is sufficient to refuse the worst products.

In reality, the validation performed is not completely correct. In fact, objects are left out only in the computation of the PLS model, but not in the computation of the PC model. For a full validation [5] the objects must be left out from the very first phase of the analysis. The results obtained after having deleted some objects in the data set and having repeated PCA and PLS are in accordance with the results above reported: this is due to the fact that there were no outliers in the data set under study, as shown in Fig. 2, where the objects are uniformly spread.

3. Experimental design for qualitative variables

The performance of a friction material depends not only on the components and on their proportions, but also process variables can have an important effect on the product. An experimental design was applied in order to investigate the effect of 7 process variables. The results of the dyno-test (55 different brakes) were transformed according to PCA and raw varimax rotation of loadings: the score on varivector 1 after range

Table 2
Study of process variables: experimental matrix and responses

Experiment	A	B	C	D	E	F	G	Score on varivector 1
1	1	1	1	1	1	1	1	54
2	1	1	2	2	1	2	2	39
3	1	2	3	3	2	1	1	35
4	1	2	4	4	2	2	2	44
5	2	2	1	2	2	2	1	14
6	2	2	2	1	2	1	2	21
7	2	1	3	4	1	2	1	49
8	2	1	4	3	1	1	2	55
9	3	2	1	3	1	2	2	80
10	3	2	2	4	1	1	1	79
11	3	1	3	1	2	2	2	16
12	3	1	4	2	2	1	1	6
13	4	1	1	4	2	1	2	10
14	4	1	2	3	2	2	1	0
15	4	2	3	2	1	2	2	100
16	4	2	4	1	1	1	1	82

scaling (range 0–100), which is, as already said, an indicator of the performance at high temperature, was the response variable.

Table 1 reports the experimental domain (for reasons of industrial secrecy, the true name of the raw materials cannot be reported). Factors A, B, E, F, G are qualitative; factors C and D, though intrinsically quantitative, are considered qualitative, in order to point out the behaviour at each proposed level. For sake of simplicity, no significant interactions between the factors were supposed to be present; this assumption was supported by previous experiments. As a consequence, a Free-Wilson model [6] was postulated. In it the first column of the model matrix is a column of 1's (to calculate the constant term); for each of the variables, $(l_v - 1)$ columns are required, l_v being the number of levels of the variable v. In our case the model contains 14 terms (1 constant term + 1 term for each factor at 2 levels + 3 terms for each factor at 4 levels). The complete matrix requires 1024 experiments: D-optimal matrices having a number of experiments from 14 to 18 were computed and their characteristics were analyzed. The solution at 16 experiments was chosen, since it allowed the best compromise between the number of experiments and the quality of the information. Table 2 reports the experimental matrix and the values of the response.

After multilinear regression, the following equation relating the factors with the response was obtained:

$$Y = 45 - 6A_1 - 14A_2 - 3A_3 - \mathbf{28B_1} - 8C_1 - 12C_2 + 3C_3 - 2D_1 - 5D_2 - 3D_3 + \mathbf{49E_1} + 5F_1 - 6G_1$$

The interpretation of a model for qualitative variables is slightly more complex than that for quantitative variables. For each factor, the reference level is the highest one. For instance, for factor A, level 1 has a coefficient of -6: this means that working at level 1 the response obtained is 6 units lower than working at level 4.

The analysis of the regression coefficients shows that factors A, C, D, F and G are not important. Factors B and E are significant ($p < 0.05$ and $p < 0.01$, respectively). When B is at level 2 (type b) and when E is at level 1 (type a), a better performance at high temperature is obtained. The 4 formulations having factor B at level 2 and factor E at level 1 (9, 10, 15, 16) are by far the best ones.

Replicates of some experiments performed in a following time showed that the differences between the 4 best formulations have the same magnitude of experimental error.

4. Experimental design for mixtures

Friction materials are a composite and their evaluation should be done with experimental design for mixtures [7]. A detailed study requires a number of experiments (preparation of a number of mixtures) not acceptable for problems of time and costs involved in the experimentation. In addition, the complete mathematical model including linear terms, quadratic terms and interactions is quite difficult to be interpreted.

To simplify the study, the materials can be divided in classes of components, each of them including several members.

In a first phase, three categories (abrasives, lubricants and metals) were studied, with the goal of identifying the effect of their proportions on the performances of the product; they are called 'major components'. Each major component is actually a ternary mixture of raw materials called 'minor components'.

Since it is an explorative study, we are interested not in computing a mathematical model, but simply in obtaining an idea about the behaviour of the products, in order to plan a further phase studying in detail the most interesting zone of the experimental domain.

The mixture abrasives + lubricants + metals constitutes 45% of the total formulation; the remaining part was not considered in this study and therefore it was kept constant. The major components can be represented as a simplex in 2 dimensions (equilateral triangle): the vertices correspond to the formulations in which the whole variable part (45%) is formed only by one of the major components and the other two are absent. For simplicity, the vertices are indicated as 100% of one of the major components, considering that this 100% is in reality the 45% of the formulation. However, as the three major components must be present in mixture, at first the constraints summarized in Table 3 have been proposed. These constraints are not coherent; in fact, if a mixture is

Table 3
Original constraints of the three major components

Major components	Experimental domain
Abrasives	5–33%
Lubricants	5–50%
Metals	5–50%

formed by the maximum amount of abrasives and lubricants (33% + 50%), the minimum amount of metals will be 17%, and the same for lubricants when abrasives and metals are at the maximum percentage. The real constraints are given in Table 4.

Fig. 4 shows the experimental domain under study; it has 4 vertices, two of which are very close. A further reduction of the experiments is possible if we replace these two points with the average of this short edge.

Table 4
Real constraints of the three major components

Major components	Experimental domain
Abrasives	5–33%
Lubricants	17–50%
Metals	17–50%

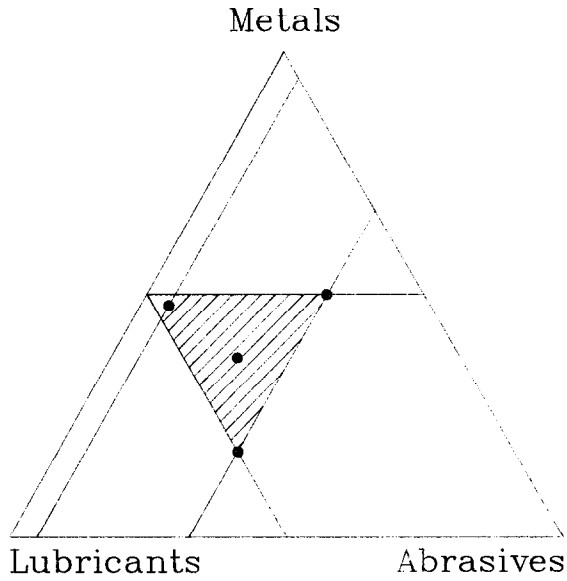

Fig. 4. Simplex of the major components: experimental domain and experimental points.

The experimental matrix includes only 4 points

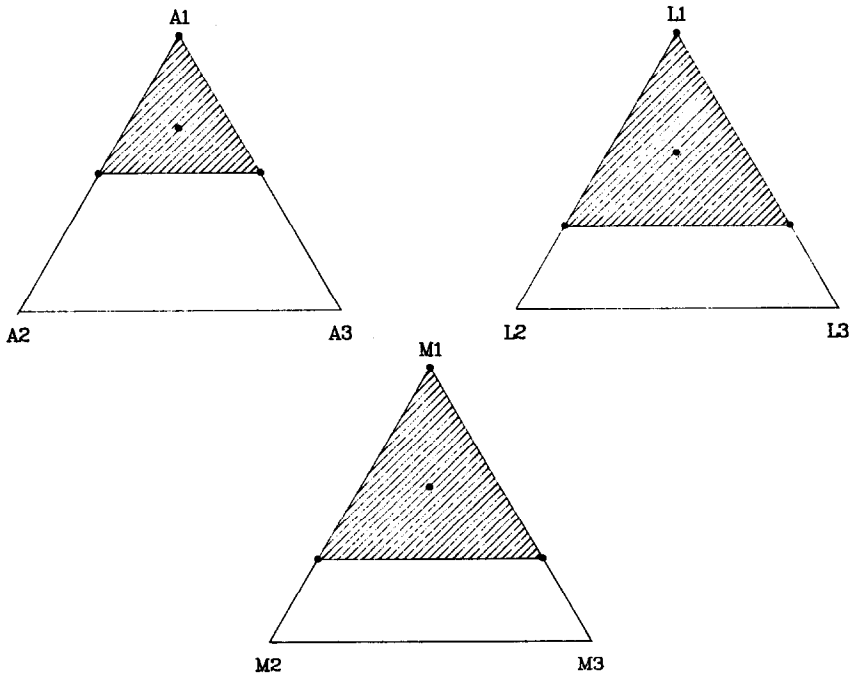

Fig. 5. Simplex of the minor components: experimental domains and experimental points of abrasives, lubricants and metals.

Table 5
Experimental matrix for the major components

Experiment	Abrasives (%)	Lubricants (%)	Metals (%)
1	33	50	17
2	33	17	50
3	5	47.5	47.5
4	22	39	39

(the 3 vertices plus one point located at the centre of the experimental domain), corresponding to the formulations given in Table 5. For each major component, 3 minor components were investigated; inside each major component one of the minor components has a lower bound (see Table 6). Fig. 5 shows the experimental domains for the mixtures of the three minor components of the categories abrasives, lubricants and metals, respectively. In each case, since only a lower constraint is present, a new simplex was obtained: 4 possible mixtures correspond to its vertices plus the central point. Under this approach a complete experimental plan includes 48 experiments: for the 4 mixtures of major components (different proportions of abrasives, lubricants and metals), 12 mixtures are studied (4 mixtures of abrasives A1, A2 and A3, 4 mixtures of lubricants L1, L2 and L3 and 4 of metals M1, M2 and M3). In the first phase we decided to investigate the effect of different proportions of the 3 major components and of the 3 lubricants. Then 16 experiments were performed: at each point of the experimental space of the major components 4 mixtures having different proportions of lubricants L1, L2 and L3 were prepared. The other major components (abrasives and metals) had composition corresponding to the central point of their simplex (i.e. the mixtures of components 1, 2 and 3 at

Table 6
Constraints of the minor components

	min. %	max. %
Abrasives		
A1	50	100
A2	0	50
A3	0	50
Lubricants		
L1	30	100
L2	0	70
L3	0	70
Metals		
M1	30	70
M2	0	30
M3	0	30

Table 7
Study of mixtures: experimental plan and responses

Experiment	Abrasives (%)	Lubricants (%)	Metals (%)	L1 (%)	L2 (%)	L3 (%)	Score on varivector 1	Wear (g)
1	33	50	17	30	70	–	84	15.2
2	33	50	17	30	–	70	63	15.2
3	33	50	17	100	–	–	62	27.5
4	33	50	17	54	23	23	60	11.3
5	33	17	50	30	70	–	70	14.2
6	33	17	50	30	–	70	74	12.4
7	33	17	50	100	–	–	81	25.3
8	33	17	50	54	23	23	100	14.3
9	5	47.5	47.5	30	70	–	35	21.7
10	5	47.5	47.5	30	–	70	24	9.7
11	5	47.5	47.5	100	–	–	0	16.3
12	5	47.5	47.5	54	23	23	48	10.9
13	22	39	39	30	70	–	62	12.3
14	22	39	39	30	–	70	45	8.3
15	22	39	39	100	–	–	84	20.1
16	22	39	39	54	23	23	58	8.6

the centre of the experimental space of abrasives and of metals, respectively). The experimental plan and the responses are reported in Table 7: for each mixture the proportions of the 3 major components are reported together with the proportions of the 3 lubricants. The results of the dyno-test (75 different brakes) were transformed according to the previously described procedure (PCA and raw varimax rotation of loadings). Two responses were taken into account: score on varivector 1 (scaled within the range 0–100), which can be considered a global indicator of the quality of the product (to be maximized) and wear, computed as difference between the weights before and after the dyno-test (to be minimized). Of course, since there is a certain direct correlation between the 2 responses, a compromise will be looked for. The results of the 16 experiments are also displayed in Fig. 6.

Mixtures poor in abrasives (experiments 9, 10, 11, 12) have the worst performances (the smallest values of score on varivector 1 – see Fig. 6a). Mixtures in which L1 was the only lubricant (experiments 3, 7, 11, 15) are generally characterized by higher values of wear, sometimes together with bad performances (Figs. 6c–d).

After deletion of these 7 experiments (Fig. 7), it can be seen more clearly that higher amounts of abrasives result in an improvement of the performances: there are no important differences between L2 and L3, except that higher amounts of L2 seem to give slightly better performances but also worse values of wear.

This trend is confirmed also by Fig. 8, in which the experiments are plotted on the plane of the two responses. To determine the 'best' formulation, a multicriteria approach is needed; due to the very high experimental error, all the methods producing linear combinations of the responses [8] cannot be used. The

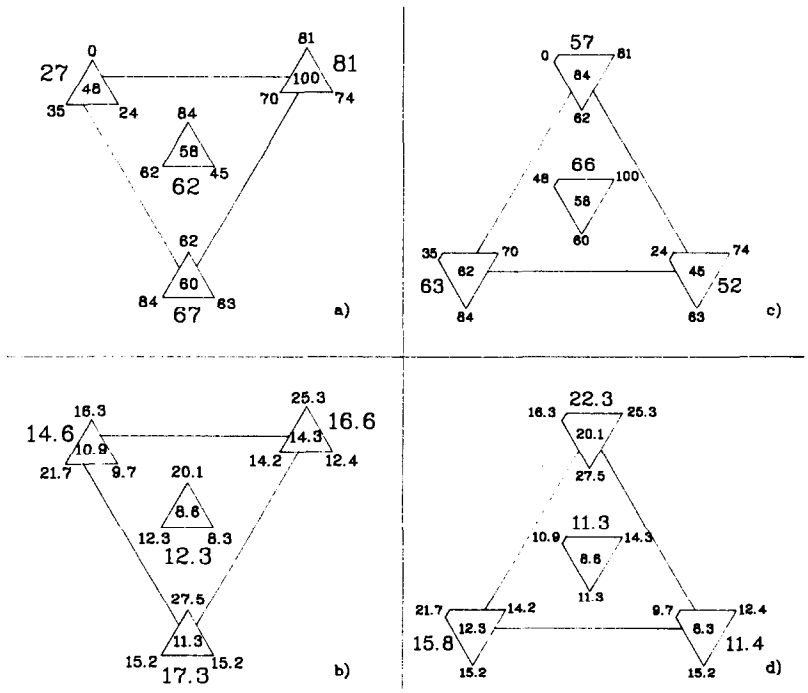

Fig. 6. Experimental responses in the space of the major components (a: score on varivector 1; b: wear) and of the lubricants (c: score on varivector 1; d: wear): small numbers for the responses of the single experiments, large numbers for the average of the responses of the experiments having the same proportions of major components (a, b) and of lubricants (c, d).

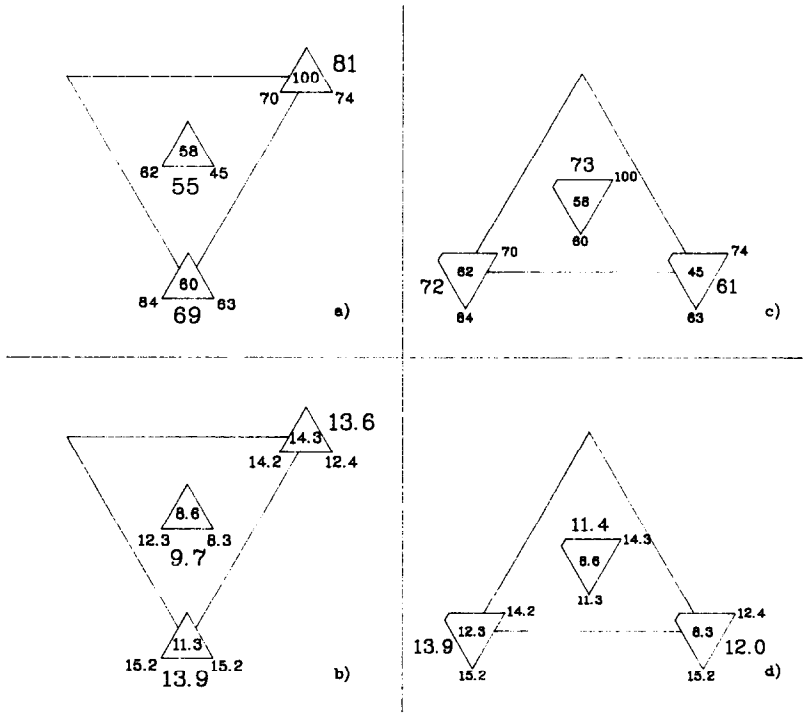

Fig. 7. Experimental responses in the space of the major components (a: score on varivector 1; b: wear) and of the lubricants (c: score on varivector 1; d: wear): small numbers for the responses of the single experiments, large numbers for the average of the responses of the experiments having the same proportions of major components (a, b) and of lubricants (c, d).

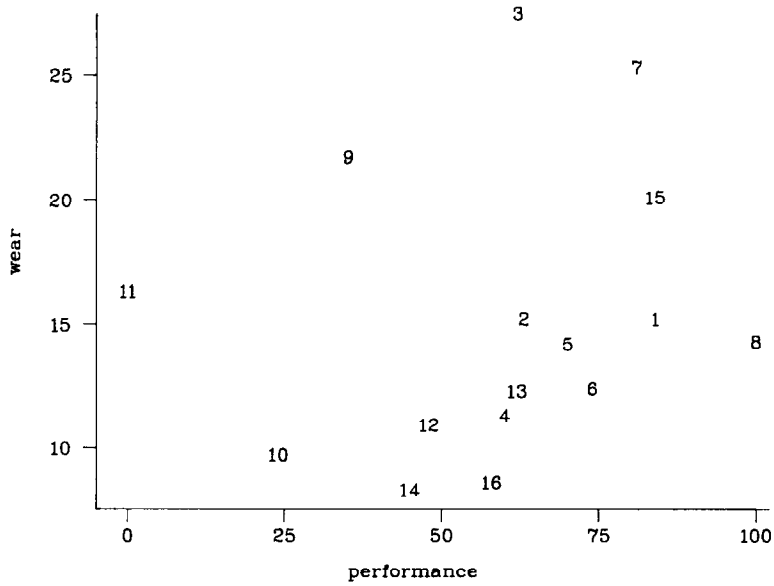

Fig. 8. Plot of the experimental points in the plane of the two responses.

Table 8
Summary of new formulations

Experiment	Abrasives (%)	Lubricants (%)	Metals (%)	L1 (%)	L2 (%)	L3 (%)	Score on varivector 1	Wear (g)
17	44	28	28	30	–	70	93	14.3
18	66	17	17	30	–	70	88	24.2

most suitable method is therefore the Pareto approach [9]; according to it, three points are optimal (8, 14 and 16), but only point 8 has an acceptable performance.

On the basis of these results, new formulations in the direction of increased amounts of abrasives were tested. The original simplex of the major components was reflected with respect to the edge delimited by the two best vertices, and two experiments (the centre and the new vertex of this simplex) were performed at the lowest percentage of lubricants L1 and L2 (Fig. 9 and Table 8).

The experiment at the centre of the new simplex (17) gave very good results for both responses; the experiment at the new vertex (18) gave good performances, but a too high value of wear. After moving the experimental domain towards increasing abrasives, it is confirmed that high proportions of this component allow to obtain good formulations. It is important to note that also amounts larger than 33% (value initially proposed as upper limit) can be successfully used. However, when the amount is too large, the value of wear becomes unacceptable.

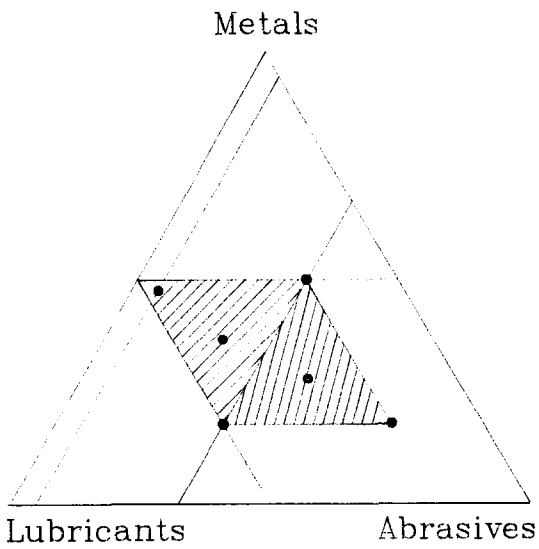

Fig. 9. Enlarged experimental domain with the new experimental points.

As a consequence of the results, experimental work is in progress in two directions:

- a new definition of the range for abrasives (both lower and upper limits will be set to higher values);
- the study of the effect of different proportions of the 3 metals and of the 3 abrasives, following the same procedure described for lubricants.

Actually, the approach followed here (i.e. the study of one major component at a time) does not allow to estimate the interactions: if one wants to know how the responses change when different proportions of each major component and each minor component are used, their amounts must be changed simultaneously. Such a strategy requires a very high number of experiments, so the loss of information about the interactions is the price we have to pay for reducing the experiments to an acceptable number.

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