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Prediction of Product Quality from Spectral Data Using the Partial Least-Squares Method

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Received August 8, 1983

Complex regression models for product quality control are calculated by partial least squares with latent variables. Predictor variables from different spectral sources are treated as separate blocks, combinations of which, according to a preset pathway, give prediction models for quality-control variables of an industrial product.

INTRODUCTION

Quality control of raw materials, intermediates, and final products is one of the crucial points in industrial processes. It is the task of analytical chemistry to provide sufficient information on which to base decisions allowing correction of the processes. There are a broad variety of data processing methods applied to quality-control problems in analytical chemistry.[1]

In the 1970s, H. Wold developed a method[2] commonly called partial least squares or PLS referring to a solution for the regression model. This method was developed for application in social sciences but, in the last few years, has found application in chemistry as well.[3-7] PLS is primarily designed for causal-predictive analysis of complex problems that are rich in data but scarce in theoretical knowledge. The basic idea is to separate predictor variables, coming from different influence sectors or sources, into blocks and describe each block by a set of latent variables. These latent variables are linear combinations of the original predictor variables, mutually orthogonal in the same block, and the ones from different blocks correlate to each other according to a preset causal path model.

In the following, a part of a complex study aimed at building predictive models by PLS for product quality control and process control problems is discussed. Our purpose is to point out the potential of the PLS method for use in quality-control problems and its advantages over other multivariate statistical methods. The requirements of proprietary confidentiality do not allow a description of all the data in detail. However, this is not a limitation to understanding the philosophy of the PLS method, illustrating its performance, and discussing the advantages of its applicability in quality control.

METHOD

Multiple linear regression by ordinary least squares is a well-known solution to the problem of describing the variance of a response variable (e.g., a quality-control variable) by a linear combination of predictor variables.[8] The ordinary least-squares algorithms, however, handle only one block of predictor variables and one response variable. In the actual practice of quality control, it is possible to measure predictor variables from different sources or different influence sectors. In this case, different chemical measurements should be handled separately in different blocks or matrices. Another version of the regression problem is the case where there is not only one but several response variables to be described and predicted together by the model. In case of an underdetermined system (more predictor variables than samples), the ordinary least-squares solution incorporates only a limited number of predictor variables in the regression model. The PLS method was developed for the case where several influence sectors are present, each represented by a separate block of variables and connected by a predetermined causal pathway. This method can handle several predictor blocks, multiple response variables, and underdetermined systems. In the following, three special cases of PLS used in this study are described.

(A) PLS Regression. One special case of PLS is distinguished by having one set of predictor variables and only one dependent variable. This is a normal multiple regression case but solved by the partial least-squares algorithm.[9] This algorithm is analogous to principal component regression as it attempts to span the block of the original predictor variables by orthogonal latent variables, which are linear combinations of the original variables. However, there is a difference in the criteria for calculating these latent variables; namely, they describe only the amount of variance in the original variables predictive for the response variable. The response variable is then modeled by a linear combination of these orthogonal latent variables. The maximum number of the latent variables NL_{max}, similar to principal components, is the number of the original variables NV. The optimal NL is found by cross validation,[10] a method that compares the predictive power of the models and chooses the one with the optimal number of latent variables for prediction. Due to the orthogonality of latent variables, the method gives a solution to the collinearity

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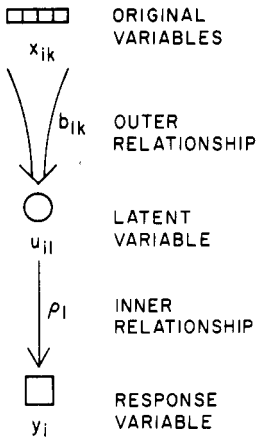

Figure 1. PLS regression.

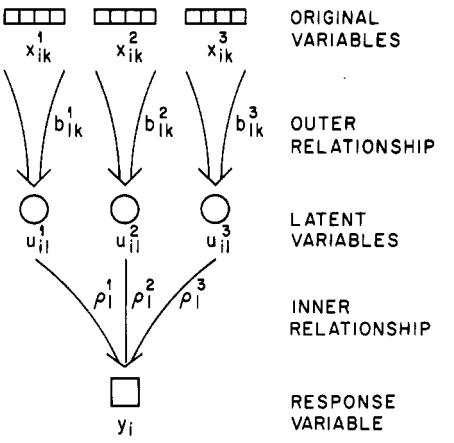

Figure 2. PLS multiblock regression.

problem, i.e., for the instability of estimated regression coefficients due to the correlation among the predictor variables. Also, this algorithm gives better prediction results than the normal least-squares solution when a model without the last latent variables describing only noise is used.

When the kth predictor variable for the ith sample is denoted x_{ik} and the response variable for the ith sample is y_i, the linear model is

$$y_i = \sum_{k=1}^{NV} \beta_k x_{ik} + \epsilon_i \qquad i = 1, ..., NP;\ k = 1, ..., NV \qquad (1)$$

where β_k's are the regression coefficients and ϵ_i is the residual error in y_i not described by the model. The variance of the predictor block associated with the response variable is described by orthogonal latent variables u_{il}, in the so-called outer relationship:

$$x_{ik} = \sum_{l=1}^{NL} u_{il} b_{lk} + e_{ik} \qquad (2)$$

where l is the index of the latent variables, b_{lk}'s are called the loadings, and e_{ik} is the residual error in x_{ik} not described by the model of NL latent variables. The response variable is also a linear combination of latent variables:

$$y_i = \sum_{l=1}^{NL} \rho_l u_{il} + \Delta_i \qquad (3)$$

where the ρ_l's are the regression coefficients for the latent variables and Δ_i is the residual of y_i. This equation is called the inner relationship. The more latent variables included in the model (NL approaches NV), the smaller the e_{ik}'s become. The scheme for the PLS regression algorithm is shown in Figure 1. The steps of the algorithm are in the Appendix.

(B) PLS Multiblock Regression. Predictor variables coming from different types of measurements (e.g., different spectra) can be represented by different influence blocks. For each of the NB blocks, a separate set of latent variables is calculated and related to the response variable. One possible scheme for this model is shown in Figure 2.

The algorithm is similar to that of PLS regression, except that latent variables and loadings are calculated for each block separately as (for notation see eq 2)

$$x_{ik}^j = \sum_{l=1}^{NL} u_{il}^j b_{lk}^j + e_{ik}^j \qquad j = 1, ..., NB \qquad (13)$$

The inner relationship in this case, analogous to eq 3, leads to linear regression between latent variables u^j and the response variable:

$$y_i = \sum_{l=1}^{NL} \sum_{j=1}^{NB} \rho_l^j u_{il}^j + \Delta_i \qquad (14)$$

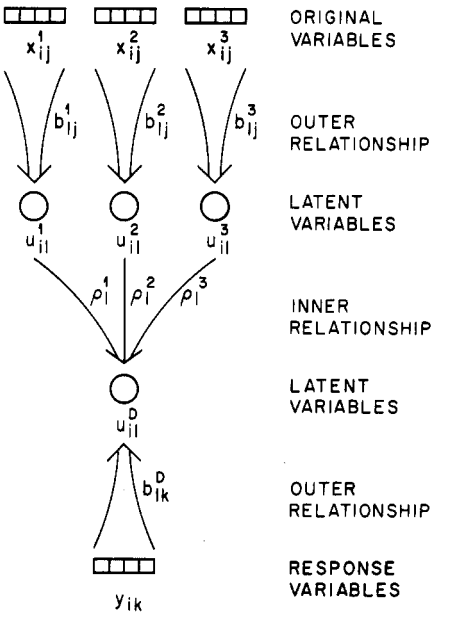

Figure 3. PLS general multiblock regression.

(C) PLS Regression with Multiple Response Variables. The algorithm can also be extended to the case when there is more than one response variable in the dependent variable block, which, similar to other blocks, can be described by a set of orthogonal latent variables u_{il}^D:

$$y_{ik} = \sum_{l=1}^{NL} u_{il}^D b_{lk}^D + e_{ik}^D \qquad (15)$$

In the inner relationship a latent variable of the dependent block D is described by a linear combination of the latent variables from the NB predictor blocks:

$$u_{il}^D = \sum_{j=1}^{NB} \rho_l^j u_{il}^j + d_{il} \qquad l = 1, ..., NL \qquad (16)$$

The scheme for this algorithm is shown in Figure 3.

In the Appendix, an algorithm is described for the case when there is only one predictor block ($X = [x_{ij}]$; $i = 1, ..., NP$; $j = 1, ..., NV1$) and the response block ($Y = [y_{ik}]$; $i = 1, ..., NP$; $k = 1, ..., NV2$). The models can be extended ad infinitum to considerably more complex systems.

EXPERIMENTAL DETAILS

Two sets of different spectral measurements were recorded from 3M adhesive tape samples. The IR spectra were measured between 600 and 4000 cm^{-1} and stored in digital form in 219 increments. The sonic spectra, representative of a typical adhesive, were recorded from 20 to 20 000 kHz in 310

Table I. Multiple Regression of Quality Variable A on IR Spectra

method	selected variables (wavelengths)	sum of absolute errors: training set	sum of absolute errors: test set
STEP	208	0.979	1.006
PLS (1)	208	1.008 (1.001)	1.005 (0.999)
STEP	208, 61	0.559	0.786
PLS (1)	208, 61	0.564 (0.560)	0.784 (0.743)
STEP	208, 61, 36	0.485	0.717
PLS (2)	208, 61, 36	0.571 (0.543)	0.652 (0.616)
STEP	208, 61, 36, 216	0.384	0.669
PLS (2)	208, 61, 36, 216	0.456 (0.466)	0.658 (0.611)
STEP	208, 61, 36, 216, 71	0.310	0.651
PLS (4)	208, 61, 36, 216, 71	0.330 (0.389)	0.639 (0.590)

Table II. Multiple Regression of Quality Variable B on IR Spectra

method	selected variables (wavelengths)	sum of absolute errors: training set	sum of absolute errors: test set
STEP	210	2.620	2.625
PLS (1)	210	2.621 (2.620)	2.623 (2.605)
STEP	210, 32	2.460	2.464
PLS (1)	210, 32	2.503 (2.498)	2.457 (2.442)
STEP	210, 32, 100	1.931	2.302
PLS (3)	210, 32, 100	1.935 (1.940)	2.172 (2.170)
STEP	210, 32, 100, 4	1.782	2.307
PLS (2)	210, 32, 100, 4	2.121 (2.120)	2.194 (2.187)

Table III. Regression on Two Predictor Blocks

method	selected variables: IR	selected variables: sonic	sum of absolute errors: training set	sum of absolute errors: test set
		Quality Variable A		
PLS (1)	208	132	0.979 (0.970)	0.956 (0.933)
PLS (2)	208, 61	132	0.581 (0.580)	0.635 (0.636)
PLS (2)	208, 61	132, 227	0.578 (0.579)	0.611 (0.610)
PLS (2)	208, 61, 36	132	0.557 (0.582)	0.649 (0.603)
PLS (2)	208, 61, 36	132, 227	0.579 (0.570)	0.640 (0.605)
		Quality Variable B		
PLS (1)	210	134	2.586 (2.581)	2.583 (2.580)
PLS (2)	210, 32	134	2.475 (2.476)	2.451 (2.445)
PLS (3)	210, 32, 100	134	1.940 (1.940)	2.144 (2.143)
PLS (1)	210, 32, 100	134, 108	2.214 (2.205)	2.205 (2.191)

increments. Two quality-control parameters were obtained on each of the 34 samples. These parameters are strongly related to the performance of the product. In the following discussion, they will be referred to as quality variable A and quality variable B. These were determined from measurements taken after the product was made. Besides these two variables, two other variables were recorded that are closely related to the process parameters; they will be referred to as process variable C and process variable D. These two variables represent data taken during the manufacturing process. Data on 34 samples were recorded and were separated randomly into a training set and a test set, each with 17 samples. The whole data set was autoscaled; i.e., each variable was scaled to zero mean and unit variance.

All calculations were done by using a PLS Fortran IV program developed by the Laboratory for Chemometrics, and computations were performed on the Department of Chemistry VAX 11/780 computer. The data were provided by 3M Engineering Systems and Technology Laboratory.

RESULTS AND DISCUSSION

The main purpose of this study was to develop an accurate mathematical relationship between on-line spectral measurements and process variables and the final quality of a product. This would allow much faster feedback for the manufacturing personnel and, hence, reduce waste and increase productivity.

To build the best possible predictive linear model, several combinations of the data blocks were examined. There are two predictor blocks, the IR spectra with 219 variables and the sonic spectra with 310 variables. The dependent block can contain either one or both of the quality variables.

First, a straightforward PLS regression was performed. Quality variable A was regressed on the IR spectra, which showed a higher correlation to the quality variables than the sonic spectra. The results are compared to those from stepwise regression (STEP) in Table I. The wavelengths used in regression models were selected by a stepwise regression algorithm available in ARTHUR program package.[11] The numbers in parentheses after PLS show the optimal number of latent variables picked by cross validation. Most of the time, the number of latent variables is less than the number of predictor variables, and the prediction results of PLS are slightly better than STEP. The prediction on the test set is seen to improve for selected wavelengths. After inclusion of more variables in the model, its predictive capability decreased. Results from the same regression models for quality variable B are reported in Table II.

In the PLS calculation, the same wavelengths were used as those which were selected by STEP. The values reported under training set and test set are the sums of the absolute error in prediction in training set and test set. These values will be referred to as results. The results clearly show that quality variable A is better predicted than quality variable B. It was found that a slightly better prediction can be achieved if individual wavelengths are not selected but, instead, are replaced by the average spectral intensity over a range of 4 wavelength units. These results are shown in Table I and II; the sum of absolute error values are given in parentheses in the training set and test set columns. It is important to note that the PLS results are always poorer on the training set used to develop the models (worse fit) but always better at prediction, thereby providing a better predictive model. This is due to its resistance to fitting noise in the measurements.

By extension of the PLS algorithm, both predictor blocks, i.e., the IR and the sonic spectra, could be included in the model. The results with STEP-selected wavelengths and averaged wavelengths (those in parentheses) for quality variables A and B are shown in Table III. Only a slight improvement was obtained by adding information from sonic spectra, which means that by this measurement selection method, the variance in quality variables described by the sonic spectra is almost a subset of the variance already described by IR data. Increasing the number of selected variables did not improve the predictive capability of the models, although there is definitely more informaion stored in the full spectra than just in a few selected variables. Therefore, a measurement selection method was needed that, while reducing the number of variables, would still conserve the amount of information of the full spectra. Using a minimum number of measurements in the model is important for on-line industrial applications where small computers are employed.

The method used was to calculate principal components of the spectra and use them as variables in the two predictor blocks. The best results for quality variables A and B are shown in Table IV. The predictive capability of the model increased significantly when principal components were used instead of the measurements in the case of quality variable A. The best prediction was achieved by using the first six principal components from the IR spectra and the first three principal components from the sonic spectra, although incorporation of the sonic spectra into the model yielded only small improvement.

In comparison of the models of principal-component predictor variables with the models of the original predictor variables, it is clear that the former models give better pre-

Table IV. Feature Selection by Calculating Principal Components

method	no. of eigenvectors IR	no. of eigenvectors sonic	sum of absolute errors training set	sum of absolute errors test set
		Quality Variable A		
PLS (1)	6	0	0.476	0.433
PLS (1)	6	1	0.476	0.431
PLS (1)	6	2	0.475	0.430
PLS (1)	6	3	0.471	0.424
		Quality Variable B		
PLS (1)	6	0	2.410	2.621
PLS (1)	6	1	2.408	2.618
PLS (1)	6	2	2.408	2.615
PLS (1)	6	3	2.405	2.615

Table V. Predictions from the Whole Spectra for Quality Variable A

method	selected no. of latent variables	sum of absolute errors training set	sum of absolute errors test set
PLS	4 from IR	0.450	0.503
PLS	5 from IR	0.366	0.617
PLS	6 from IR	0.209	0.731
PLS	4 from IR and sonic	0.316	0.566
PLS	5 from IR and sonic	0.219	0.555

diction, although the fit is better by the latter models. In Table V we have the results for quality variable A from different PLS models on the basis of the whole spectra.

Calculating principal components as predictor variables for quality variable B did not improve the prediction compared to the model using selected original variables. By examination of the four correlation coefficients between quality variable B and the first four principal components with the largest eigenvalues each from IR and sonic spectra, it becomes clear that most of the variance of the spectra is not related to the quality variable B. This is true also for the quality variable A–sonic spectra relationship and is the reason why involvement of sonic spectra as a second block in the model did not decrease the prediction error significantly. This hypothesis is supported also by the small regression coefficients for the sonic features.

It was suspected that the sonic spectra are correlated not only to the quality variables but also to the process variables. To improve the former relationship, the variance due to the process variables was removed by subtracting one of the process variables from each of the channels of sonic spectra after autoscaling. There is a large negative correlation between the two process variables; therefore, subtracting both of them would cause an overcorrection. In Table VI the best prediction results on the basis of one or two predictor blocks for quality variables A and B are summarized. It is clear that combination of the information from IR spectra and sonic spectra, free from the variance due to process variable C or D, improves the prediction, especially in the case of quality variable B. For both quality variables, the best prediction was achieved by a combination of six principal components and five sonic principal components free of variance due to process variable C. After the quality variables were unscaled the average absolute relative errors (calculated as $\sum_{i=1}^{NP}$[(true − predicted)/true]/NP) are reported in Table VII. In comparison of the relative errors of prediction by the best STEP and PLS models, it is clear that significant improvement can be achieved by the latter model. PLS gave twice as good prediction as the stepwise model used before, which significantly improves the quality control of this product.

As a last step, prediction of both quality variables at the same time was attempted. The model consisted of two blocks: the predictor block of IR spectra and the dependent block of the two quality variables. Principal components with the largest eigenvalues were extracted from IR spectra and used as predictor variables. The results, given in Table VIII, indicate that the prediction errors in this case were slightly higher than those from models predicting each quality variable separately. If there were enough samples to estimate accurately the multidimensional distribution, this two-block model would give better prediction for both quality variables. The high correlation between quality variables A and B would stabilize the model and correct the bias in the latent variables of the predictor block. However, in our case, due to the small number of samples, this model failed to improve the prediction.

Table VI. Prediction with Sonic Spectra Free of Variance due to Process Variables

method	no. of eigenvectors IR	no. of eigenvectors sonic − process variable	sum of absolute errors training set	sum of absolute errors test set
		Quality Variable A		
PLS (1)	6		0.476	0.433
PLS (1)		6 − C	0.533	0.592
PLS (1)		6 − D	0.535	0.603
PLS (1)	5	5 − C	0.413	0.578
PLS (1)	6	5 − C	0.361	0.391
PLS (1)	6	6 − C	0.363	0.394
PLS (1)	5	5 − D	0.415	0.577
PLS (1)	6	5 − D	0.360	0.397
PLS (1)	6	6 − D	0.370	0.401
		Quality Variable B		
PLS (1)	6		2.410	2.621
PLS (1)		6 − C	1.944	2.694
PLS (1)		6 − D	2.529	2.880
PLS (2)	5	5 − C	1.213	1.331
PLS (3)	6	5 − C	1.111	1.319
PLS (3)	6	6 − C	1.042	1.545
PLS (2)	5	5 − D	1.363	1.924
PLS (2)	6	5 − D	1.355	1.899
PLS (2)	6	6 − D	1.363	1.783

Table VII. Prediction Errors by the Best PLS Model and Stepwise Regression Model for Quality Variables A and B

	sum of absolute errors by STEP training set (%)	sum of absolute errors by STEP test set (%)	sum of absolute errors by PLS training set (%)	sum of absolute errors by PLS test set (%)
quality variable A	2.9	3.2	1.2	1.3
quality variable B	7.5	8.8	4.3	4.7

Table VIII. Prediction of Both Quality Variables Together

no. of principal components from IR spectra	sum of absolute errors training set A	sum of absolute errors training set B	sum of absolute errors test set A	sum of absolute errors test set B
4	0.535	2.676	0.595	2.905
5	0.503	2.685	0.594	2.891
6	0.490	2.573	0.458	2.836

CONCLUSIONS

In this paper, linear regression models were developed by PLS method according to different causal paths to describe and predict quality variables of a 3M product. Several kinds of preprocessing were investigated to find the best features for predictor blocks. The prediction was improved significantly by using principal components as orthogonal features in the predictor blocks of IR and sonic spectra. A further decrease in the prediction errors was achieved by subtracting the variance from sonic spectra due to process variables.

The results discussed here represent a part of a complex study to characterize and predict the quality of raw materials

and final products directly on the process stream. Our goal was 2-fold. From a methodological point of view, we wanted to introduce the philosophy of the PLS method, to describe in detail three algorithms solving different causal paths, and to point out the advantages of PLS over other linear regression methods. From the applications point of view, we showed how to calculate PLS models in a quality-control problem and how to improve the prediction power of the PLS model by various feature selections by incorporating more information (spectral or process variables). Our intention was to attract the attention of chemists involved in quality-control problems to PLS and to demonstrate its usefulness by a successful application.

APPENDIX

Algorithm for PLS regression.

(1) Autoscale X and Y: subtract from each variable their means and divide by their standard deviation.

(2) Set $l = 1$, $\beta_k = 0$, $k = 1, \ldots, \mathrm{NV}$, and $Z(\mathrm{NV}^*\mathrm{NV}) =$ identity matrix.

(3) Weights:

$$w_{kl} = \sum_{i=1}^{\mathrm{NP}} x_{ik} y_i \tag{4}$$

(4) Normalize weight vector to unit length.

(5) Latent variables (outer relationship):

$$u_{il} = \sum_{k=1}^{\mathrm{NV}} x_{ik} w_{kl} \tag{5}$$

(6) Correlation coefficients (inner relationship):

$$\rho_l = \sum_{i=1}^{\mathrm{NP}} y_i u_{il} \tag{6}$$

(7) Loadings:

$$b_{kl} = \sum_{i=1}^{\mathrm{NP}} u_{il} x_{ik} \rho_l \tag{7}$$

(8) Regression coefficients:

$$\beta_j = \beta_j + \rho_l \sum_{k=1}^{\mathrm{NV}} w_{kl} z_{kj} \tag{8}$$

(9) Update Z matrix:

$$Z = Z - ZW_l B'_l \tag{9}$$

(10) Residuals in X matrix:

$$x_{ik} = x_{ik} - u_{il} b_{lk} \rho_l \tag{10}$$

(11) Residuals in Y vector:

$$y_i = y_i - \rho_l u_{il} \tag{11}$$

(12) $l = l + 1$. Check significance of the next latent variable by cross validation (10). If it was found significant, go back to step 3; otherwise, continue with step 13.

(13) Prediction:

$$y_i = \sum_{k=1}^{\mathrm{NV}} x_{ik} \beta_k \tag{12}$$

Algorithm for two-block PLS.

(1) Autoscale X and Y as in PLS regression.

(2) $l = 1$.

(3) To start the iteration, set $u_{il}^2 = y_{i1}$.

(4) Weights for block I:

$$w_{jl} = \sum_{i=1}^{\mathrm{NP}} x_{ij} u_{il}^2 \tag{17}$$

(5) Normalize the weight vector to unit length.

(6) Latent variables for block I (outer relationship):

$$u_{il}^1 = \sum_{j=1}^{\mathrm{NV1}} x_{ij} w_{jl} \tag{18}$$

(7) Weights for block II:

$$w_{kl} = \sum_{i=1}^{\mathrm{NP}} y_{ik} u_{il}^1 \tag{19}$$

(8) Normalize the weight vector to unit length.

(9) Latent variables for block II (outer relationship):

$$u_{il}^{2'} = \sum_{k=1}^{\mathrm{NV2}} y_{ik} w_{kl} \tag{20}$$

(10) Convergence check: If $|u_{il}^{2'} - u_{il}^2| < \delta$, convergence is reached and continue with step 11. Otherwise, go back to step 4 with $u_{il}^2 = u_{il}^{2'}$.

(11) Inner relationship:

$$\rho_l = \sum_{i=1}^{\mathrm{NP}} u_{il}^1 u_{il}^2 \tag{21}$$

(12) Loadings for block I:

$$b_{jl} = \sum_{i=1}^{\mathrm{NP}} u_{il}^1 x_{ij} \rho_l \tag{22}$$

(13) Residuals in X matrix:

$$x_{ij} = x_{ij} - u_{il}^1 b_{lj} \rho_l \tag{23}$$

(14) Residuals in Y matrix:

$$y_{ik} = y_{ik} - \rho_l u_{il}^1 w_{kl} \tag{24}$$

(15) $l = l + 1$. Check significance of the next latent variables by cross validation. If significant, go back to step 3; otherwise, prediction of block II.

ACKNOWLEDGMENT

This work was supported in part by the Office of Naval Research.

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