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Discrimination of coatings on wooden materials using the gas sensor system

M. Maciejewska*, K. Kolodziejczak, A. Szczurek

Ecologistics and Atmosphere Protection Group, Institute of Environmental Protection Engineering, Wroclaw University of Technology, Pl. Grunwaldzki 9, 50-377 Wroclaw, Poland

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Abstract

The applicability of sensor system for the discrimination of sources of indoor pollution was investigated. As examples of indoor pollution sources, paint and lacquer coatings were considered. Commercially available preparations: Akrylux, Doamlux, Bejca and White Scandinavian were selected for headspace measurements using TGS sensor array. Following issues were investigated: (1) discrimination between water- and solvent-based coatings, (2) discrimination between one component coatings, and (3) discrimination between one component and two component coatings. Following data analysis methods were used: principal component analysis (PCA), linear discriminant analysis (LDA) and probabilistic neural network (PNN). Results showed that coatings could be discriminated successfully, provided the surface covered was solid wood (0–1.8% error). The interference of fibreboard volatiles in sensor measurements of coatings was most likely encountered. It could have significantly impaired discrimination of coatings on fibreboard (2.8–5.6% error) as compared to wood. Worst results were obtained for the discrimination of coatings on unknown material(12.5–28.7% error).

Keywords: Volatile compounds; Headspace; Sensor measurement; PCA; LDA; PNN

1. Introduction

In developed countries people spend about 90% of their time in indoor environment. The air quality in these places is quite different from that in outdoors. Researchers indicate that the air within houses and other buildings can be more seriously polluted than outdoors. Thus, for many people the risk to health may be greater due to exposure to air pollution indoors than outdoors [1].

The presence of airborne contaminants in building interior produces health effects and undesirable symptoms known as the sick building syndrome (SBS). A number of well identified illnesses, e.g. asthma, hypersensitivity pneumonitis have been directly traced to specific building problems. The SBS is characterised by complaints from the building occupants, such as: dry or burning mucous membranes in the nose, eyes and throat, sneezing, stuffy or running nose, fatigue or lethargy, headache, dizziness, nausea, irritability and forgetfulness [1–4].

The building's space is polluted by a large number of volatile substances emitted from many different sources. The full understanding of a given indoor air quality situation requires specific knowledge about emission sources. Usually they are separated into three groups, according to their origin: (1) outdoor air, (2) man and his activities (body odors, energy production, smoking, household activities and hobby products), (3) materials and equipment (building and renovation materials, furnishings). The most important sources with regard to materials and equipment are: adhesives, lacquers, paints, caulks, floor coverings, floor sealants, insulation materials, furniture, office machines [2,5].

When indoor sources of air pollutants are present, the emission of these compounds should be characterised. Measurements provide extremely valuable information in that respect. Different methods and techniques are available for

* Corresponding author. Tel.: +48 713202500; fax: +48 713203599.
E-mail address: monika.maciejewska@pwr.wroc.pl (M. Maciejewska).

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making such measurements. The most reliable approach is to set up an in situ or laboratory system to determine emission from a source directly. Of course the problem of extrapolating results obtained in laboratory measurements to full-scale building is critical, as pollutants may decay or interact under real life conditions. Therefore, the in situ techniques are preferred in indoor analysis. It would be very fruitful to apply instruments based on gas sensors in this field of measurements. Their primary advantages are relative simplicity, low cost, ability to be used in a continuous analysis. The disadvantage of gas sensor is a poor selectivity.

The question was asked concerning applicability of gas sensor system for the discrimination of sources of indoor pollution, which are coatings adequate for wooden materials, applicable for furnishing, floor and wall coverings. Several diagnostic tasks that the sensor system could perform were identified. These were: (1) discrimination between coatings made of solvent-based preparation and coatings made of water-based preparations, (2) differentiation of individual preparations used for surface coating, (3) discrimination between single layer coatings and two layer coating. Considering that TGS sensors, which we used were designed and were shown to well respond to reductive gases [6,7,8] and also to water vapour [6,9,10] the investigation of the issue these sensors appeared justified.

2. Experimental

2.1. Materials

Solvent- and water-based preparations were considered. Each group of preparations was represented by a commercially available lacquer and paint. The general description of preparations used in the experiment is presented in Table 1.

Preparations were applied to solid wood and to fibreboard, which is a wood-based material. Coatings consisting of one preparation (Akrylux, Domalux, Bejca and White Scandinavian) and coatings consisting of two preparations (Bejca/Domalux) were considered. Coatings were painted on small pieces of wood and fireboard (5 cm × 2 cm) in order to match our measurement system. Samples were stored in open glass jars placed in dry, well ventilated shed. This arrangement was intended to imitate conditions of coating drying process.

Table 1
Commercially available preparations, which were used in the experiment for coatings preparation

Name	Description
Akrylux	Paint, based on acrylic dispersion in water
Domalux	Lacquer, based on polyurethane dispersion in water
Bejca	Paint, based on alkyd resin dispersion in white spirit
White Scandinavian	Lacquer, based on organic solvents (solvents details unknown)

Sensor array measurements were performed 1 h, 1 day, 2, 3, 4 and 5 days after applying the coating. Three replicates of each combination surface-preparation were considered.

2.2. Measurement system

The layout of measurement system is shown in Fig. 1. The main part of system was a sensor cell (5) with TGS800, TGS822, TGS824, TGS825, TGS880 and TGS883 sensor array (4) inside. The pump (12) was used to flush out pure air through the measurement system. The air was provided by Horiba calibrator (8). The air stream could be directed with valves (6) either straight to the sensor cell (5) and then to the outlet from the system or it could be directed via the flask with the sample (9) prior to reaching sensor cell. The rotameter (11) was used for gas flow control. Sensors in the sensor cell were constantly heated with a power supply unit (10). The signal from sensor array was transferred via the connector (3) to the signal conditioning and multiplexer module of Data acquisition/switch unit Agilent 34970A (2). The connector (3) is a transition between sensor array wiring and standardises electric wiring of Agilent 34970A. Measurement data was filed on the computer with HP BenchLink Datalogger software (1).

The measurement procedure consisted of two phases. It started with flushing out the whole system with a stream of pure air for about 20 min. It removed any volatile compounds, which could have remained in the system after the measurement phase. In the second phase, the air stream (1 dm^3/min) from calibrator was directed through the flask with a sample (9) in order to carry volatile compounds from sample headspace to the measurement cell (5). In that phase the actual measurement was performed.

During measurement phase sensors operated in thermo stimulation mode. While the measured gas stream was flowing through the sensor cell following voltage sequence was applied to sensor heaters: 5 V over 3 min, followed by 3 V over 2 min, and finally 5 V over 1 min. Due to the contact with reductive gases and/or water vapour coming from coatings, resistance of sensors changed. The value of resistance of all sensors was monitored during full thermo stimulation cycle.

2.3. Data pre-processing

An example of time response of gas sensors during thermo stimulated measurement phase is shown in Fig. 2.

Two points from thermo stimulated response curve of each sensor were further considered. The value of sensor response at the end of first heating period 3 min (5 V), which is further denoted by $r_{i,1,j}$ and the value of sensor response at the end of second heating period 2 min (3 V), denoted by $r_{i,2,j}$. Based on values $r_{i,1,j}$ and $r_{i,2,j}$, data pre-processing was proposed, which enabled most efficient discrimination between

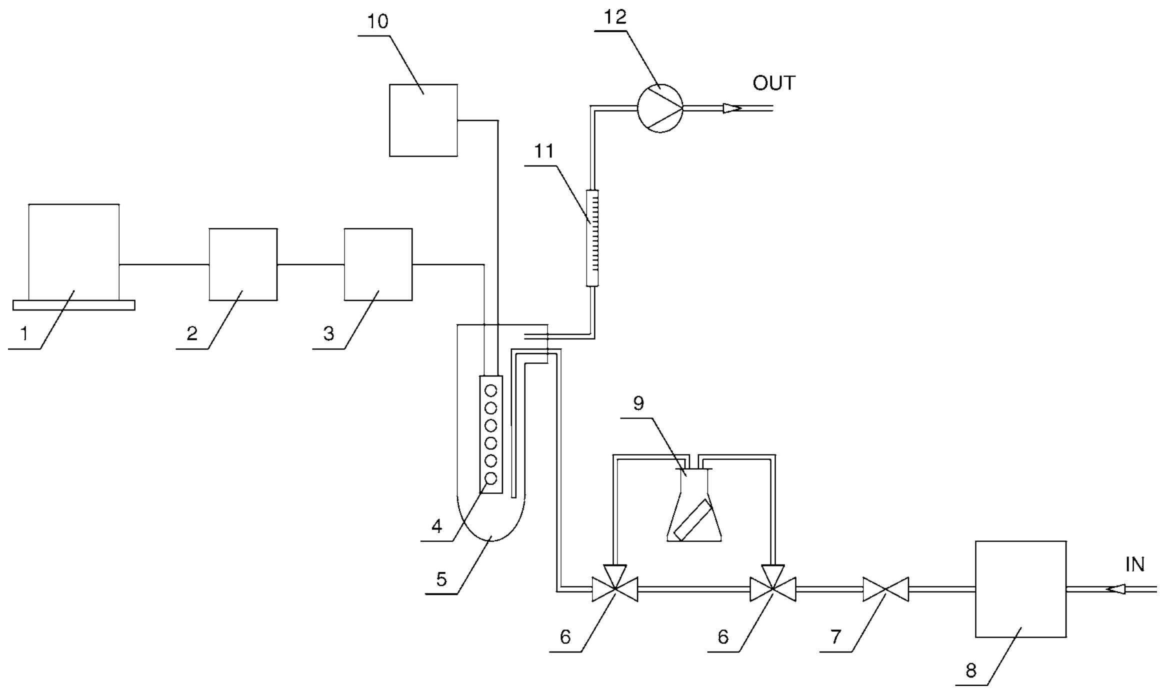

Fig. 1. The structure of measurement system.

coatings afterwards. The pre-processing formula is given by

$$R_{i,j} = \frac{r_{i,1,j} - \overline{(r_{i,1,1},\ \ldots,\ r_{i,1,n},\ r_{i,2,1},\ \ldots,\ r_{i,2,n})}}{\delta(r_{i,1,1},\ \ldots,\ r_{i,1,n},\ r_{i,2,1},\ \ldots,\ r_{i,2,n})} \qquad (1)$$

where $i = 1, \ldots, 6$ is the sensor number, $j = 1, \ldots, n$, n indicates the number of all measurements dedicated to a particular combination surface-coating. The motivation of using this pre-processing formula was to take into account changes of sensor array responses to a preparation due to the weathering process as the additional indicator of coating. Only scaled responses ($R_{i,j}$) of gas sensors were further considered as the basis for discrimination between coatings.

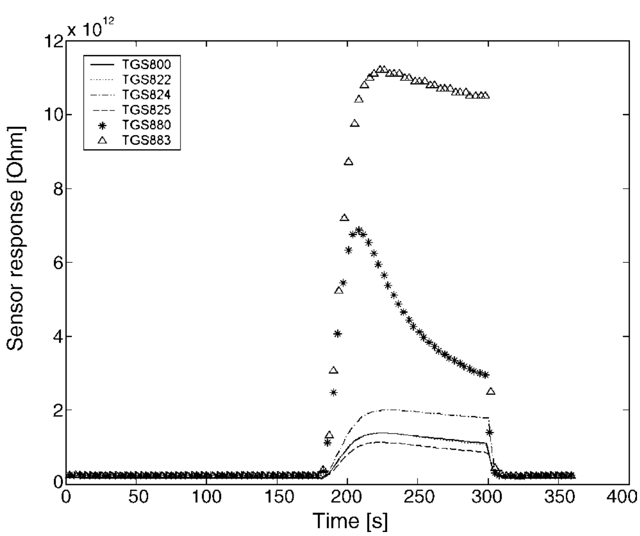

Fig. 2. Time response curves of gas sensors during thermo stimulated measurement cycle (5 V for 3 min, 3 V for 2 min, 5 V for 1 min). The sample measured was Bejca on solid wood, 5 days after the coating was made.

3. Results and discussion

3.1. Examination of patterns in data – principal component analysis (PCA)

PCA is a set of mathematical transformations performed on the multidimensional space of variables. A set of new variables, called principal components results from the transformation given by

$$\begin{bmatrix} R_{1,1} & \ldots & R_{1,d} \\ \vdots & \ddots & \vdots \\ R_{n,1} & \ldots & R_{n,d} \end{bmatrix} = \begin{bmatrix} S_{1,1} & \cdots & S_{1,d} \\ \vdots & \ddots & \vdots \\ S_{n,1} & \ldots & S_{n,d} \end{bmatrix} \cdot \begin{bmatrix} L_{1,1} & \ldots & L_{1,p} \\ \vdots & \ddots & \vdots \\ L_{d,1} & \ldots & L_{n,p} \end{bmatrix} \qquad (2)$$

where d, number of original variables ($i = 1, \ldots, d$); n, number of data vectors ($j = 1, \ldots, n$); p, number of principal components ($k = 1, \ldots, p$); $R_{i,j}$, single measurement result, element of measurement data matrix, $S_{i,j}$, single score, element of principal components matrix; $L_{i,k}$, single loading, element of loadings matrix [11]. The number of variables prior to transformation and after it is the same ($d = p$). New variables are orthogonal and ordered in a way that the first variable carries most of the variance of original variables, second carries less variance and so on. New variables preserve all the information content of original variables, but usually only first two or three PCs are important in data examination. Due to the fact that most of original data variance is usually preserved in first two or three components, PCA is often applied as a data dimensionality reduction technique. It also makes PCA a good method of data examination with respect to the appearance of patterns characteristic for measured objects. They appear in two or three dimensional plots of first, most significant PCs [11].

3.2. Linear data grouping – linear discriminant analysis (LDA)

LDA is a technique of linear discrimination between groups of data vectors. It is a supervised technique. Prior to the analysis a correct assignment of data vectors in training data set to groups must be known. The basic idea underlying discriminant analysis is to determine whether groups of data vectors differ with regard to the mean of any variable, and then to use that variable for predicting group membership. Usually, all measured variables are considered in order to see which ones contribute to the discrimination between groups. LDA maximizes the ratio of between-group variance to the within-group variance in any particular data set thereby guaranteeing maximal separation between groups. With respect to variables selection the discriminant analysis is very similar to multidimensional analysis of variance (MANOVA), which also allows to see which variables have significantly different means across groups [12,13].

In course of LDA discriminant functions are calculated, which are also called canonical variables. These are weighted sums of original variables, which contribute to between group discrimination, as given by

$$\mathrm{DF}_i = a_{1,i}x_1 + a_{2,i}x_2 + \cdots + a_{p,i}x_p \qquad (3)$$

where p, number of original variables; $i = 1, \ldots, k-1$; k, number of groups in the data. Discriminant functions are optimal combination of variables so that the first function provides the most overall discrimination between groups, second provides less discrimination, and so on. Discriminant functions are *orthogonal*, which means their contributions to the discrimination between groups do not overlap. The maximum number of functions is equal to the number of groups minus one, or the number of variables in the analysis, whichever is smaller. Original data vectors transformed into the space of canonical variables produce scores. The scores plot reveals how discriminative functions discriminate data set. Frequently, only first two or three canonical variables are significant and sufficient to obtain required data grouping.

Next to discriminant functions classification functions are calculated, as follows

$$\mathrm{CF}_i = b_0 + b_{1,i}x_1 + b_{2,i}x_2 + \cdots + b_{p,i}x_p \qquad (4)$$

where p, number of original variables; $i = 1, \ldots, k$; k, number of groups in the data. The number of classification functions equals the number of groups in the data set. With those functions classification scores can be computed for each data vector and for each group. The highest score obtained for a considered data vector indicates which group the vector belongs to. The performance of data classification by means of classification functions is an indication of quality of discriminant power of discriminant functions.

3.3. Nonlinear data grouping – probabilistic neural network (PNN)

PNN is a type of neural network suitable for solving classification problems [14]. It has a two-layer architecture. The first layer consists of neurons with radial basis transfer functions. The number of neurons in that layer equals the number of input vectors in a training set. Second layer of PNN is a competitive layer. The number of neurons in that layer equals the number of classes. Therefore, the design of PNN is straightforward and does not depend on training. The structure of neural network is shown in Fig. 3.

The class assignment of training vectors, is known from input-output pairs in the training data set. It is assumed that there are q input vector/target vector pairs and each target vector has k elements. One of these elements is 1 and the rest is 0. Thus, each input vector is associated with one of k classes.

The first layer of neural network computes distances from input vector I_0, which is subject to classification, to each training vector in the input data matrix. It is possible because initial weights of the first layer I_1W are set to the transpose

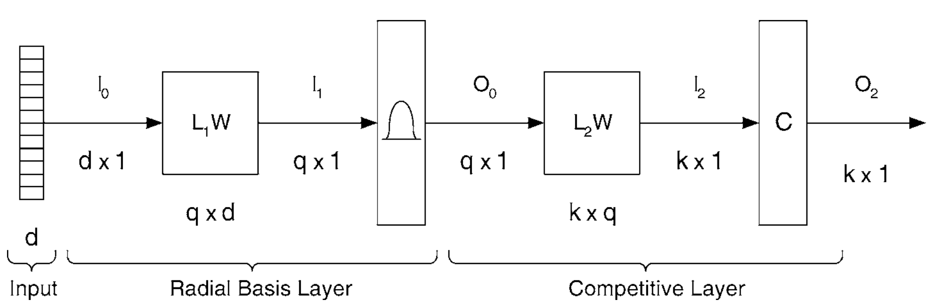

Fig. 3. The structure of probabilistic neural network; d, size of input vector; q, number of input/target pairs (number of neurons in layer 1); k, number of classes of input data (number of neurons in layer 2) [15].

of the training data matrix. Distances calculated I_1 are than multiplied by bias and transformed with a radial basis transfer function. Elements of resulting vector O_1 indicate how close the input vector is to input vectors in training data set. In case input vector is close to a training vector a value near 1 is obtained from radial basis function. If an input vector is close to several training vectors of a single class, several values close to 1 will be obtained.

The second layer of the network uses values calculated by the first layer O_1 to sum contributions from input vector to each class. In order to do this second layer weights are set to the matrix target vectors. The multiplication of I_2W by O_1 sums the elements of O_1 within each of the k classes. As a result a vector of probabilities of class membership is produced. Finally, a compete transfer function on the output of the second layer I_2 picks the maximum of these probabilities, and produces 1 for the class of vector assignment and 0 for the other classes.

PNN guarantees convergence to a Bayesian classifier on the condition that it is provided with enough training data. These networks show good generalization properties [14,15].

3.4. Discrimination between one preparation coatings

In the first part of analysis one preparation coatings were investigated. The discrimination between water-based coatings (Domalux, Akrylux) and solvent-based coatings (Bejca, White Scandinavian) was examined, as well as discrimination between four individual coatings. Coatings applied to solid wood, fibreboard and also irrespective of surface covered were examined separately. Discrimination was based on sensor array measurements performed at any time between 1 h and 5 days from applying the coating.

Based on comparison of PCA results, shown in figures Figs. 4 and 5, one could see that patterns occurring in the analysed data were strongly dependent on the surface covered. There is a notable shift in clusters position in Fig. 5 with respect to Fig. 4, which is an indication of the presence of some volatiles, others than ingredients of preparations. These volatiles could possibly emanate from fibreboard.

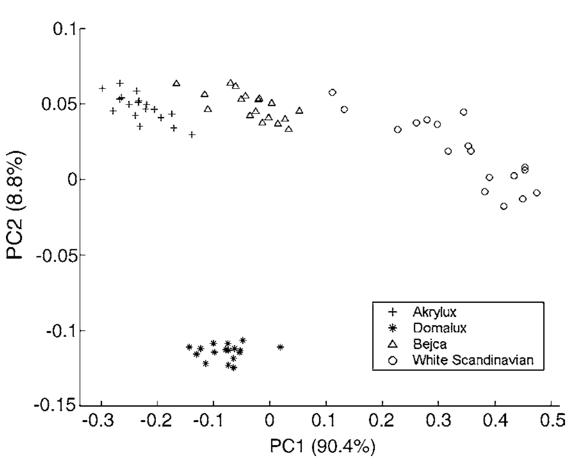

Fig. 4. The plot of scores resulting from PCA of sensor array measurements of one preparation coatings on solid wood, in coordinates of first two principal components.

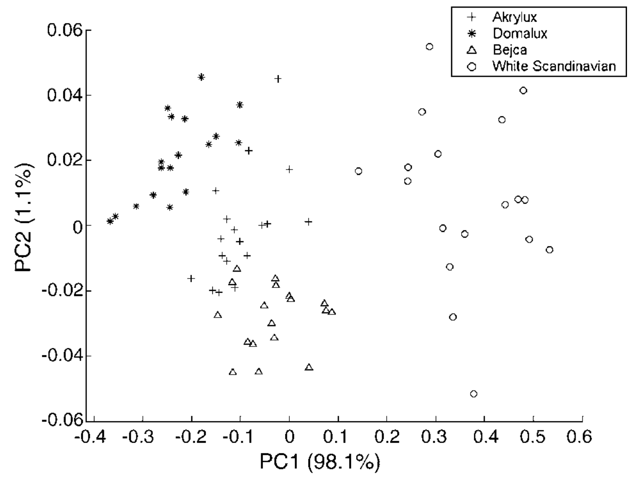

Fig. 5. The plot of scores resulting from PCA of sensor array measurements of one preparation coatings on fibreboard, in coordinates of first two principal components.

Separate groups of points representing each individual coating on solid wood could be identified in the PCA plot (Fig. 4). Patterns occurring in measurement data were clearly not determined by the difference between solvent- and water-based coatings. Nevertheless, good linear discrimination of water-based (Akrylux, Domalux) and solvent-based (Bejca, White Scandinavian) coatings on solid wood could be anticipated from the points layout in Fig. 4.

As it is shown in the PCA plot for coatings on fibreboard (Fig. 5) groupings formed by points representing individual coatings are less distinct than in case of solid wood. However, based on points layout in Fig. 5, one could expect significant degree of linear discrimination between water-based coatings (Akrylux, Domalux) and solvent-based coatings (Bejca, White Scandinavian) as well as between four individual coatings.

As it is displayed in Fig. 6, PCA of sensor array measurements of coatings irrespective of surface covered result in distinctive clusters for Akrylux and Domalux on solid wood. Points corresponding to Akrylux on fibreboard and points corresponding to Domalux on fibreboard are mixed with the others and form together a separate grouping. Data patterns which occur in Fig. 6 do not indicate linear discrimination between individual coatings and also between water- and solvent-based coatings.

In order to investigate the discrimination of coatings, based on sensor array measurements, supervised techniques of data analysis were applied. Linear discriminant analysis represented linear data discrimination methods and probabilistic neural network was employed as a non-linear data classifier. The discriminative power of both methods was tested in leave one out mode, so that the actual efficiency of coatings recognition, based on data vectors excluded from

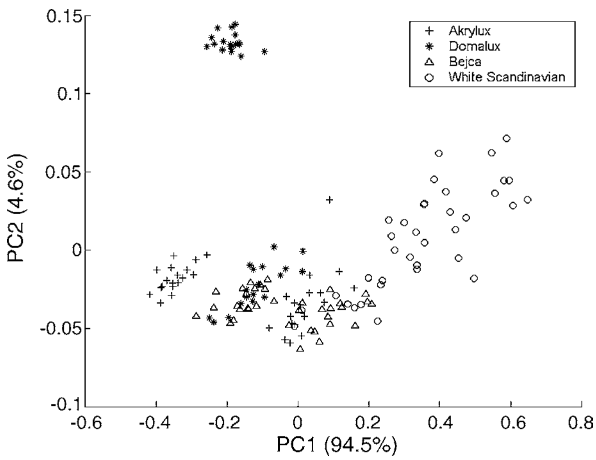

Fig. 6. The plot of scores resulting from PCA of sensor array measurements of one preparation coatings irrespective of the surface coated, in coordinates of first two principal components.

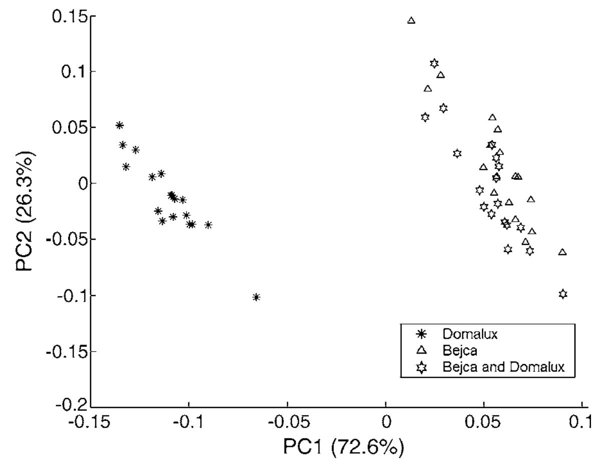

Fig. 7. The plot of scores resulting from PCA of sensor array measurements of Bejca, Domalux and Bejca/Domalux coatings on solid wood, in coordinates of first two principal components.

training data set, could be estimated. Percentages of faulty recognitions encountered during LDA and PPN testing are shown in Table 2.

Based on result shown in Table 2 one can see that water-based coatings are successfully discriminated from solvent-based coatings and also each individual coating is recognised successfully, provided that this coating is applied to solid wood. In case of fibreboard surface 3% recognition error had to be accepted irrespective of discrimination method applied. The most difficult task was to recognise coatings without prior knowledge of the surface covered. In that case PNN performed better than LDA. An error of 13% was associated with recognition of individual coatings, while 8% error was related to the discrimination between water- and solvent-based coatings.

The cause of difficulties in discrimination between coatings without prior knowledge of surface covered (Table 2) could be that fibreboard, as opposed to wood, was itself a source of volatile substances, which induced response of sensors. Probably these substances came from the binding medium in fibreboard. They could be emitted in the process of coating drying, when the material coated is subject to a ‘sponge effect’ as discussed in [16,17].

The results shown in Fig. 5 could imply that sensor array response to native fibreboard substances was very similar to sensor response to solvent-based preparations. Therefore,

Table 2
Percentage [%] of faulty classifications of single layer coatings in ‘leave one out’ model testing mode

Surface	Discrimination between water- and solvent-based coatings		Discrimination between individual coatings	
	LDA	PNN	LDA	PNN
Solid wood	1.4	0	0	0
Fibreboard	2.8	2.8	2.8	6.9
Wood or fibreboard	12.5	8.3	21.5	12.5

no separate clusters for solvent-based coatings on wood and fibreboard were formed. Contrary, sensor array response to native fibreboard substances was different from the response to water-based preparations. Therefore, the surface coated introduced significant discriminative factor in case of water-based coatings.

3.5. *Discrimination between water-based coating, solvent-based coating and mixed coating*

In the second part of analysis single preparation coatings and two preparation coatings were investigated. A discrimination between solvent-based coating (Bejca), water-based coating (Domalux) and two preparation coating (Bejca layer covered with Domalux layer) was examined. Discrimination of coatings on solid wood, on fibreboard, and irrespective of surface was investigated based on sensor measurement performed at any time between 1 h and 5 days from applying the coating.

Based on comparison of Figs. 7 and 8, patterns which occur in measurement data are dependent on the surface covered. There is a notable shift in clusters position in Fig. 8 with respect to Fig. 7, which is an indication of the presence of some volatiles, others than ingredients of preparations. These volatiles could possibly emanate from fibreboard.

In case of solid wood, good separation between water- and solvent-based coating together with a two preparation coating was observed (Fig. 7). This type of pattern appears also in Fig. 8, which shows PCA scores for fibreboard coatings.

Provided that surface covered was unknown, PCA resulted in one data grouping for water-based coating on solid wood and second grouping of all the other measurement results (Fig. 9).

LDA and PNN were applied to investigate the possibility of discrimination between water-based coating (Domalux), solvent-based coating (Bejca) and two layer coating (Bejca/Domalux). Discriminative power of both methods was

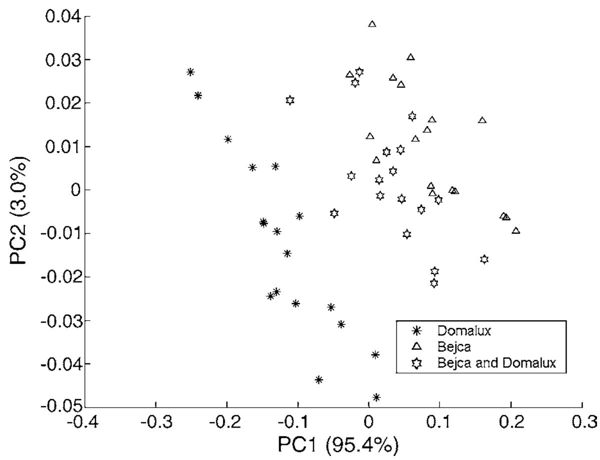

Fig. 8. The plot of scores resulting from PCA of sensor array measurements of Bejca, Domalux and Bejca/Domalux coatings on fibreboard, in coordinates of first two principal components.

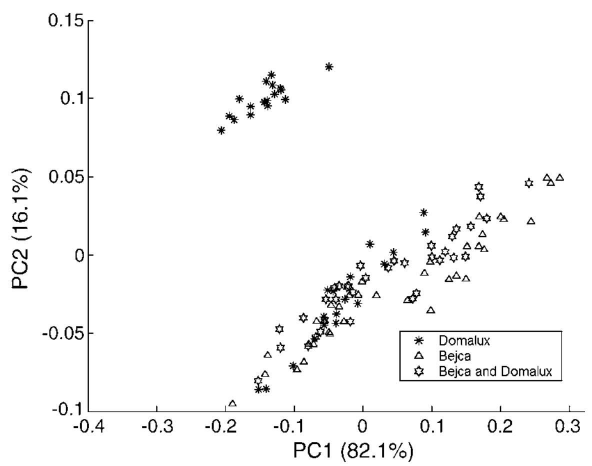

Fig. 9. The plot of scores resulting from PCA of sensor array measurements of Bejca, Domalux and Bejca/Domalux coatings irrespective of the surface covered, in coordinates of first two principal components.

tested in leave one out mode, so that the actual efficiency of coatings recognition, based on data vectors excluded from training data set, could be estimated. Percentages of faulty recognitions encountered during LDA and PPN testing are shown in Table 3.

Based on results, which are presented in Table 3, linear technique performs significantly better in classification of coatings than non linear technique. This result was unexpected. In particular, we could not find an explanation of 20% error in case of PNN discrimination of coatings on solid wood. Coatings recognition efficiency was acceptable

Table 3
Percentage [%] of faulty classifications of single preparation coatings and two layer coating in 'leave one out' model testing mode

Surface	LDA	PNN
Solid wood	1.8	20.4
Fibreboard	5.6	9.3
Wood or fibreboard	28.7	28.7

on the condition that surface covered was known. Coatings on solid wood were recognised by linear discriminator with 1.8% error and on fibreboard with 5.6% error (Table 3). Most difficult was discrimination between Domalux, Bejca and Domalux/Bejca coatings provided the surface covered was unknown. Neither linear nor nonlinear classifier could solve this task successfully.

Nevertheless the analysis of classification results revealed that Domalux coating (water based) was recognised without error in case the surface covered was known. Provided the surface was unknown Domalux on wood was never misclassified. This result could be anticipated, based on Fig. 9, and considering previous observations based on Fig. 6. Provided sensor array responds not only to preparations, but also to fibreboard originating volatiles and responds to them similarly as to solvent-based preparations, the interference from fibreboard could have explained good separation of Domalux on wood from Domalux on fibreboard, Bejca and Domalux/Bejca on any surface.

4. Conclusions

Coatings of wooden surfaces were subject to discrimination based on measurements of volatile substances present in their headspace. In our experiment, solvent- and water-based preparations were used as coating agents. We considered two types of wooden surfaces and these were solid wood and fibreboard. Measurements with TGS sensor array and subsequent data processing with PCA, LDA and PNN were aimed at discrimination between coatings.

A pattern was noticed in PCs plots of measurement data (Figs. 6 and 9), which represented separation between water-based coatings applied to wood and other coatings on any material. This pattern indicated the possibility of interference from fibreboard originating volatiles in sensor measurements of coatings. The interference was especially detrimental for coatings recognition when surface covered was unknown. It resulted in 12.5% classification error for one component coatings and 28.7% classification error for mixed coatings.

Patterns revealed in sensor measurements by PCA (Figs. 7–9) pointed at consistent similarity between solvent-based coating and mixed coating composed of water- and solvent-based preparations. Based on it one could suppose that sensor response to mixed coating was determined by its solvent-based layer. Coatings similarity, did not hinder their recognition in case of solid wood (1.8% error). Unfortunately, in case of fibreboard the similarity effect plus the interference form fibreboard resulted in 5.6% recognition error, which increased to 28.7% if the material was unknown.

Regardless of discussed misclassifications water-based coating were recognised without error provided the material covered was known in advance. Otherwise only water coating applied to wood was recognised with 100% accuracy.

Coatings discrimination on solid wood was most successful. It was less efficient in case of fibreboard and worst classification results were obtained if painted material was unknown.

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