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Artificial intelligence/fuzzy logic method for analysis of combined signals from heavy metal chemical sensors

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ABSTRACT

The cross-sensitivity of chemical sensors for several metal ions resembles in a way the overlapping sensitivity of some biological sensors, like the optical colour receptors of human retinal cone cells. While it is difficult to assign crisp classification values to measurands based on complex overlapping sensory signals, fuzzy logic offers a possibility to mathematically model such systems. Current work goes into the direction of mixed heavy metal solutions and the combination of fuzzy logic with heavy metal-sensitive, silicon-based chemical sensors for training scenarios of arbitrary sensor/probe combinations in terms of an electronic tongue. Heavy metals play an important role in environmental analysis. As trace elements as well as water impurities released from industrial processes they occur in the environment. In this work, the development of a new fuzzy logic method based on potentiometric measurements performed with three different miniaturised chalcogenide glass sensors in different heavy metal solutions will be presented. The critical validation of the developed fuzzy logic program will be demonstrated by means of measurements in unknown single- and multi-component heavy metal solutions. Limitations of this program and a comparison between calculated and expected values in terms of analyte composition and heavy metal ion concentration will be shown and discussed.

1. Introduction

The detection of heavy metal ions in aqueous solutions plays an important role in the field of environmental and industrial water analysis. Heavy metals e.g., copper, lead and silver, are not biodegradable and consumption of small amounts of them over a long period or intake of large amounts over a short period can lead to chronic or acute diseases, respectively. Here, essential enzymes and thus biochemical processes in the human body are inhibited [1]. In recent years, it could be demonstrated that miniaturised silicon-based chalcogenide glass sensors can be very well applied for the detection of heavy metals. Those sensors showed good stability in liquid media, high long-term stability in operation, low detection limit and compatibility to silicon technology [2–12]. However, in multi-component solutions potentiometric chalcogenide glass chemical sensors show cross-sensitivities towards other/interfering ions in the test sample due to the nature of the complex sensing material. Based on cross-sensitivities of chemical sensors in combination with intelligent data analysis software, different electronic tongues for the detection of e.g., heavy metals, different kinds of wines, beverages and even tomatoes, have been developed [13–25]. However, such electronic tongues consist of either pattern or complex recognition tools, e.g., artificial neural networks (ANN), principal component analysis (PCA), partial least squares (PLS) regression and soft-independent modelling of class analogy (SIMCA), and/or of a huge number of sensors. The calibration of these electronic tongues results in a complex, time-consuming and laborious procedure. In order to meet these problems fuzzy logic as intelligent (while different to many other techniques that mimic human intelligence, transparent and with short calculation times) data recognition software together with miniaturised silicon-based chalcogenide glass sensors can offer an innovative, relatively "simple" and fast approach for qualitative and quantitative detection of multi-component heavy metal solutions.

The concept of fuzzy logic was introduced more than 40 years ago by Zadeh [26]. Lying dormant for many years, it has been rediscovered in the mid-1980s for regulation in micro-electronics, automatic process regulation or in operation research. In general,

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fuzzy logic is an artificial intelligence (AI) technique that tries to emulate human decision processes, which are usually based on comparative estimation rather than on fixed thresholds. Whenever various measures with complex interrelationship have to be considered, static decision rules often fail. This is where fuzzy logic can help.

By now, the fuzzy set theory has many applications in a large variety of different domains. It has, for example, been used for segmentation of molecular surfaces by means of physico-chemical potentials [27]. In combination with other AI techniques, like neural networks, the benefits of both methods can lead to fast learning self-supervised decision-making systems [28]. Since the field is quite complex and in development, the basics of fuzzy logic cannot be discussed fully in this paper. For detailed representation refer to [29,30]. Here, only those concepts which are (more or less) directly used for the cross-sensitive sensor analysis are presented.

- *Fuzzy set theory* may be seen as a generalisation of classical set theory, each element of a fuzzy set $\tilde{A}$ being defined by a function value x in definition space X together with its degree of membership to $\tilde{A}$. The latter is defined by a membership function $\mu_{\tilde{A}(x)}$, whose values lie normally within a range $0 \le \mu_{\tilde{A}(x)} \le 1$ between zero and complete membership, respectively:

$$\tilde{A} = \{x, \mu_{\tilde{A}(x)} | x \in X\}. \tag{1}$$

In classical (crisp) sets $\mu_{\tilde{A}(x)}$ can only be 0 or 1, while fuzzy logic allows almost any type of function for membership definitions.

- One of the most important tools in applications of fuzzy set theory is the *concept of linguistic variables* (LV) [31]. These are groups of fuzzy sets with (partially) overlapping membership functions over a common (crisp) basic variable x. In order to represent several classes within a LV the membership functions should cover all the relevant definition space of the basic variable x with membership function values $0 \le \mu_{\tilde{A}(x)} \le 1$ (see Fig. 1). Values of 0 or 1 are assigned to the rest of the definition space in all membership functions. The overlap of these functions defines the fuzziness. Generally, a linguistic variable $\mathfrak{L}$, classified by n fuzzy sets $\tilde{A}_i$, can be defined as

$$\mathfrak{L} = \{\tilde{A}_1, \ldots, \tilde{A}_n\} \tag{2}$$

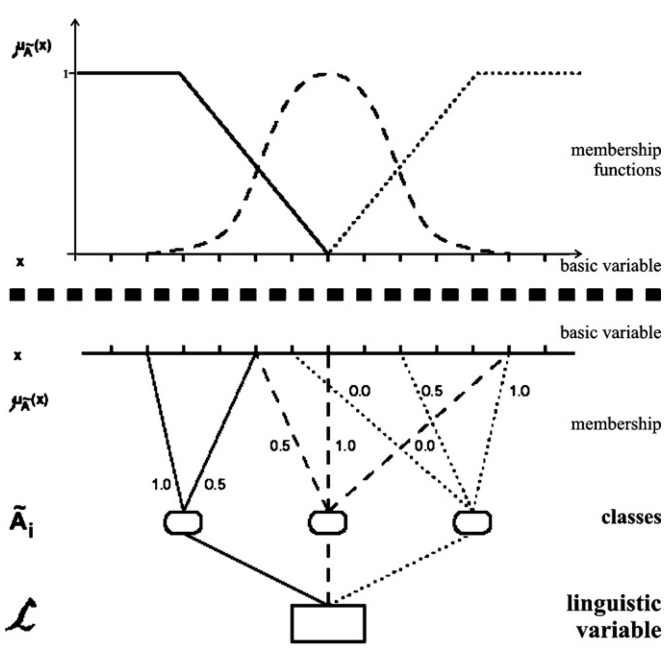

Fig. 1. Schematic of linguistic variables.

or, together with Eq. (1):

$$\mathfrak{L} = \{\{x, \mu_{\tilde{A}1(x)}\}, \ldots, \{x, \mu_{\tilde{A}n(x)}\}\}. \tag{3}$$

- Usually, the information of a decision (*decision making in fuzzy environments*) should be based upon, is given by crisp function values; for sensor signal analysis, this means e.g., the voltage output of an electrochemical sensor. Also the decision itself shall again lead to a crisp value: in this case, the binary decision about how to interpret the signal tuple of the applied sensor array in terms of ion type and concentration. However, in order to apply fuzzy logic tools to a problem, it has to be defined by linguistic variables. Thus, decision making requires three steps:
  1. fuzzification (the conversion of crisp input data into fuzzy sets and formulation of LV for further calculations),
  2. fuzzy inference (the application of fuzzy operators for the mathematical evaluation of the LV, thereby producing new fuzzy sets),
  3. defuzzification (the conversion of fuzzy sets resulting from the inference step into evaluable concrete crisp output data).

The details of these steps are discussed with the specific application patterns as far as necessary. For further details, see reference [30].

Aiming at the development of artificial sensors, like an “electronic tongue”, it might be helpful to have a look on biological sensors which have to cope with very similar problems (e.g., in particular cross-sensitivity of specific sensor cells for physical or chemical stimuli). Nature has been working on this problem for millions of years—and has solved it quite impressively in a multitude of organisms with highly accurate as well as broad-range perceptive abilities.

A well-known example is the reception of colour by the human visual system, where light is absorbed by three different receptor proteins in the retinal cone cells. These sensor cells then emit an electric signal according to the wavelength of the absorbed light, which finally leads to a cognitive impression of colour. The optical sensors have absorption maxima at different wavelengths, but with overlapping sensitivity curves (Fig. 2). These curves strikingly resemble the membership functions of a linguistic variable, which led to the idea to develop a software system based on fuzzy logic for the interpretation of signals derived from a set of cross-sensitive artificial sensors.

This work demonstrates a new concept of a recognition method for the qualitative and quantitative detection of different heavy

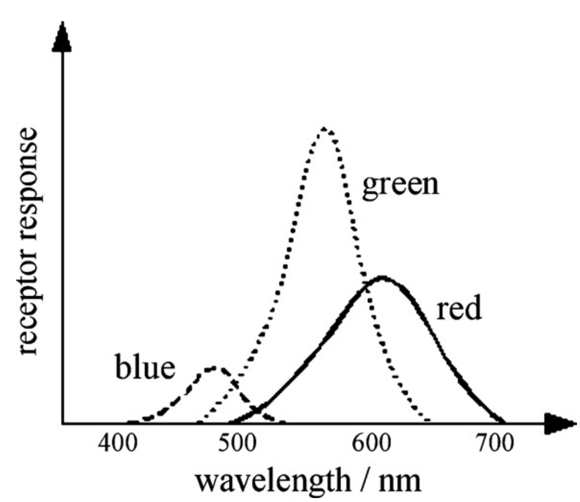

Fig. 2. Human colour perception: overlapping sensitivity curves of cone cell colour receptors (blue—dashed line; green—dotted line; red—continuous line); scheme after an image in [32].

metal ions in unknown mixed solutions by means of a miniaturised chalcogenide glass multi-sensor system. The method is based on fuzzy logic as an AI technique. The critical evaluation and validation of this fuzzy logic program will be presented by means of measurements in unknown single- and multi-component heavy metal solutions. Limitations of the fuzzy logic program due to the input data and a comparison between calculated and expected values in terms of analyte composition and heavy metal ion concentration will be shown and discussed.

2. Experimental and computations

2.1. Chemical sensors and measurement set-up

The miniaturised chalcogenide glass multi-sensor system consists of an Ag-, Cu- and Pb-selective sensor. Each sensor is made of a p-doped Si layer (specific resistance >1000 Ω cm) with a 500 nm thick SiO_2 layer for electrical insulation and a metal contact consisting of 15 nm Ti, 30 nm Pt and 250 nm Au on the sensor substrate. In order to realise the miniaturised Ag-, Cu- and Pb-sensors, complex chalcogenide glass material systems of AgIAsSe, CuAgAsSe, $PbI_2Ag_2SAs_2S_3$, have been used, respectively. The deposition of these chalcogenide glass materials in a thin-film state onto the substrates has been performed by means of pulsed laser deposition (PLD) technique. The sensing area is approx. 40 mm^2. For more detailed information of sensor fabrication, see [4,33,34].

The (ion-selective) potentiometry was presented to characterise the electrochemical behaviour of the chalcogenide glass sensors in single- and multi-component heavy metal solutions. Fig. 3 demonstrates schematically the measurement set-up, including the multi-sensor system (Ag-, Cu-, Pb-sensor) and a conventional double-liquid junction Ag/AgCl reference electrode which are immersed in the analyte solution and connected via a highly ohmic multimeter (2700, Keithley) to close the electrical circuit. The outer and inner electrolyte of the reference electrode is 10^{-1} mol/l KNO_3 and 10^{-1} mol/l KCl, respectively.

Three different stock solutions have been prepared: 10^{-2} mol/l $Ag(NO_3)$, 10^{-2} mol/l $Cu(NO_3)_2$ and 10^{-2} mol/l $Pb(NO_3)_2$. Every two stock solutions have been mixed in ratios of 100%:0%, 80%:20%, 60%:40%, 40%:60%, 20%:80% and 0%:100%, in order to realise different single- and multi-component analyte solutions with different ratios of heavy metal ions. As background solution 10^{-1} mol/l KNO_3 with 10^{-3} mol/l HNO_3 solution has been applied.

The calibration measurements as well as the potentiometric measurements in unknown heavy metal solutions have been performed in the background solution increasing the heavy metal ion concentration from 10^{-6} mol/l to 10^{-3} mol/l by using the standard addition method.

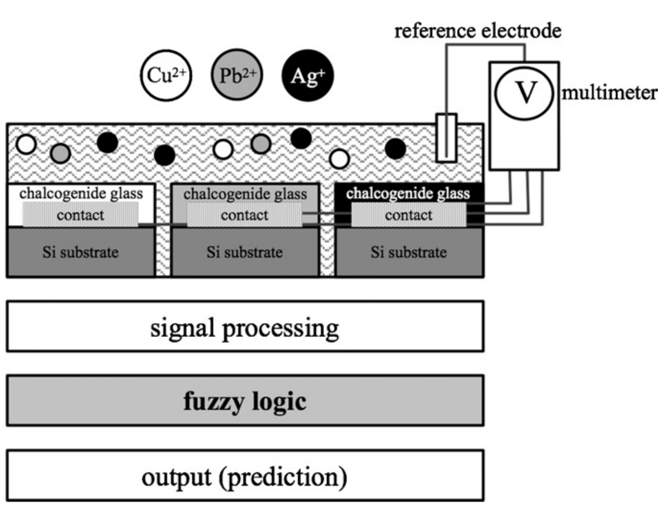

Fig. 3. Schematic of the measurement set-up for the development of a fuzzy logic program.

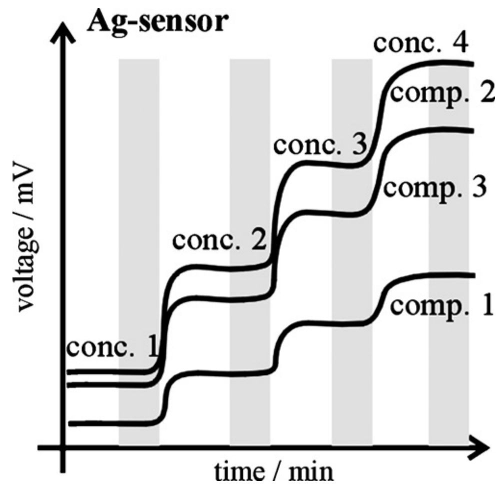

Fig. 4. Measurement curve from potentiometric measurements in three different heavy metal ion compositions (in diagram “comp. 1” to “comp. 3”) in concentration range “conc. 1” to “conc. 4” performed with an Ag-selective sensor. The measured potential values, which are underlined grey, are used as a priori knowledge for fuzzy logic calculations.

2.2. Development of fuzzy logic software

For the development of the fuzzy logic software, the calibration measurements performed in different heavy metal solutions were imported into a database. The program extracted the information of relevant measurements. Here, relevant information was after 1.5 min of a measurement period in a certain ion concentration of the solution (Fig. 4, grey areas), because of the response time of the heavy metal sensors. Relevant information was used as a priori knowledge for further calculations. These measurements were assigned as characteristic curves to appropriate sensors. A characteristic curve consists of an average value of the calibration measurements in each heavy metal ion concentration of a solution (Fig. 5a, solid line) and the corresponding standard deviation (Fig. 5a, shade). All three sensors were associated with all calibration measurements resulting in a total number of 18 characteristic curves for each sensor, as 18 different calibration solutions for the single- and multi-component measurements have been investigated. Applying the sensor set to a heavy metal solution of unknown composition and concentration, from the relevant measured potentials (a priori knowledge) for each sensor a constant mean potential was calculated.

Fuzzy logic was used to combine the constant mean value of each sensor with the characteristic curves of the particular sensor to an intersection line. From the intersections of the measured constant mean potential with every characteristic curve a fuzzy set was calculated. An intersection was assigned as the highest membership value (1) if the constant mean potential cuts exactly the average of the curve and as the lowest membership value (0) if the measured mean potential was outside the standard deviation of the characteristic curve. Between the average value and its standard deviation the membership value was interpolated linearly. Fig. 5b shows exemplarily the membership function of the Ag-selective sensor from the intersection of the measured average potential with the characteristic curve from composition 2 (from Fig. 5a).

Three fuzzy sets were summarised to a linguistic variable, due to the number of constant mean potentials in a solution, and thus

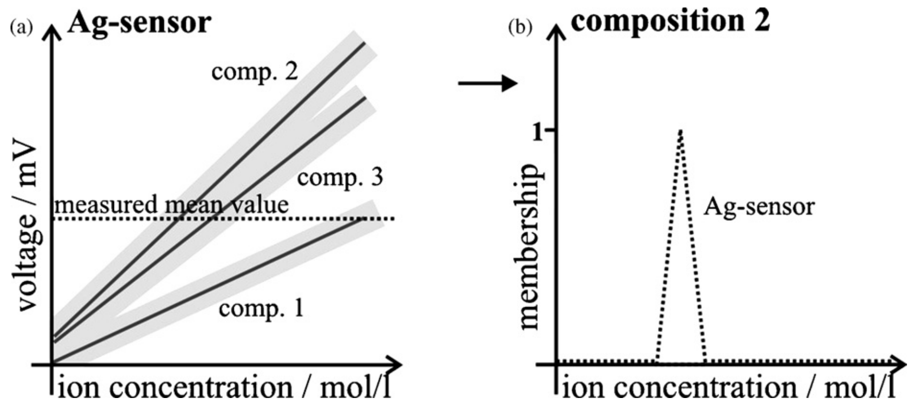

Fig. 5. (a) Characteristic curves of a Ag-selective sensor in three different heavy metal ion compositions with their mean value (solid line), their respective standard deviation (shade) and the constant mean potential measured in an unknown heavy metal ion solution by the Ag-selective sensor. (b) Fuzzy set based on the intersection between the measured constant mean value and the characteristic curve of composition 2 (see (a)).

to the number of applied sensors. This resulted in 18 different linguistic variables, according to the number of used calibration solutions.

For the prediction of the measured unknown heavy metal solution a fuzzy intersection operator (fuzzy AND connective) was applied to the three fuzzy sets of a linguistic variable through their membership functions. This fuzzy AND connective constituted a membership function for the unknown solution to a known solution represented by the linguistic variable (see Eq. (4)).

$$\mu_{\mathrm{AND}(x)} = \min\{\mu_{\text{Ag-sensor}(x)}, \mu_{\text{Pb-sensor}(x)}, \mu_{\text{Cu-sensor}(x)}\}, \quad x \in X \qquad (4)$$

Through an analysis of all linguistic variables a likely conclusion on the composition of the unknown heavy metal solution was made. The most likely composition was the linguistic variable with the highest membership value of its fuzzy AND connective membership function.

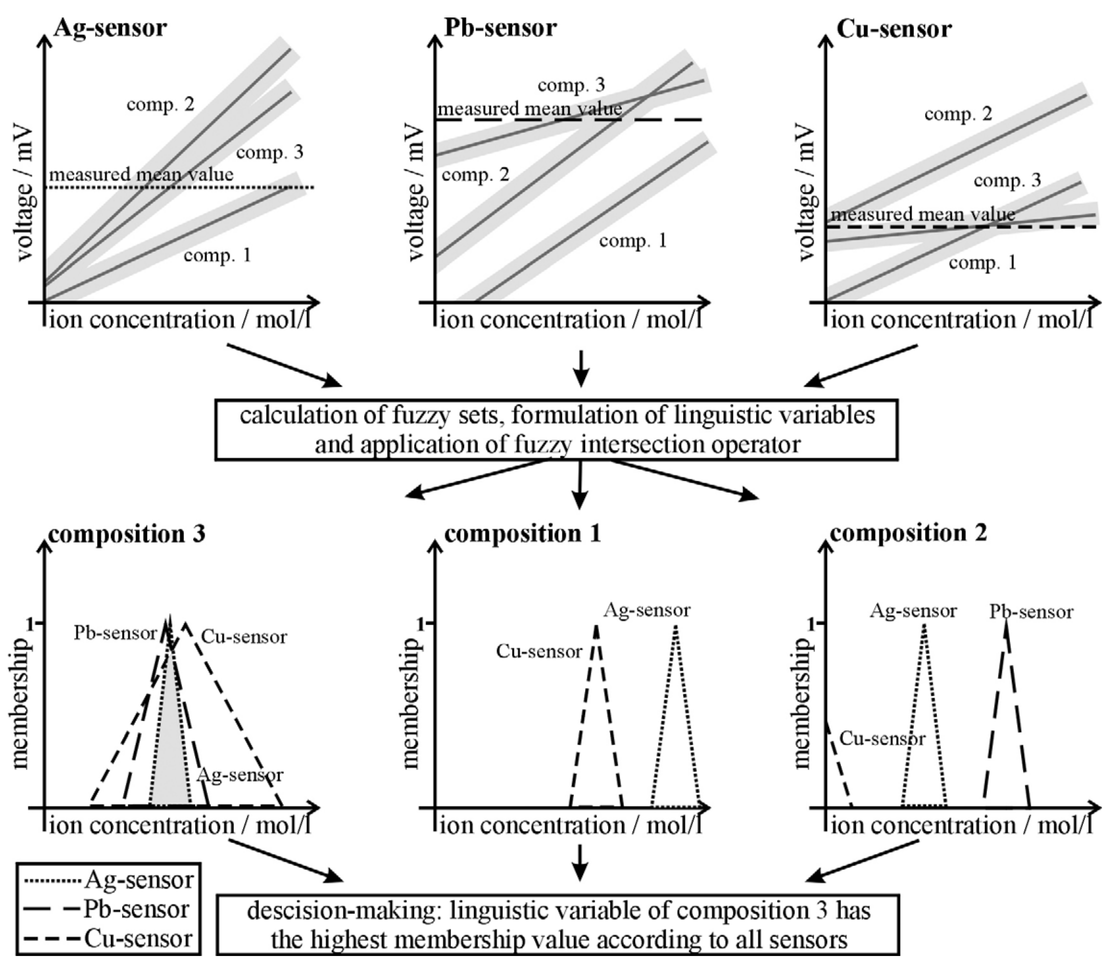

Fig. 6. Schematic of the procedure of the fuzzy logic program based on three chalcogenide glass sensors (Ag-, Pb- and Cu-selective sensor), three calibration compositions and the measured constant mean value in an unknown heavy metal solution. The “grey area” of the composition 3 represents the intersection membership function of the three fuzzy sets (see Eq. (4)).

An overview of the fuzzy logic program procedure is exemplarily shown in Fig. 6. Each one of the three sensors has measured a signal in an unknown heavy metal solution. The program calculated fuzzy sets based on the determined constant mean potential values and characteristic curves of known ion type and concentration combinations, that are compositions 1–3. In each case, three fuzzy sets, according to the three different sensors used, were formulated to a linguistic variable for each composition. On each linguistic variable the fuzzy intersection operator (Eq. (4)) was applied. The linguistic variable of compositions 1 or 2 did not have any intersection areas, thus the unknown heavy metal ion composition was neither composition 1 nor 2. However, composition 3 showed an intersection area of all electrodes (see "grey area" in Fig. 6), resulting in that the unknown measured heavy metal composition was most probably the composition 3 with the ion concentration where the highest membership value of the intersection area was found.

3. Results and discussion

3.1. Characterisation of the sensors

In order to illustrate important sensor parameters in terms of electronic tongue behaviour, sensitivity, cross-sensitivity, response time and linear measuring range will be discussed exemplarily. Fig. 7 demonstrates calibration measurements with the miniaturised chalcogenide glass-based Ag-sensor in three different analyte solutions: Ag:Cu with 100%:0%, Ag:Pb with 0%:100% and Cu:Pb with 60%:40%. The measurement in analyte solution containing only Ag^+-ions represents the sensor behaviour towards its primary ion. The Ag^+-ion sensitivity was about 60.5 mV/dec. and correlates well with the expected Nernstian response for monovalent ions. The response time was around several seconds and the linear measuring range was extended over four decades down to 10^{-6} mol/l Ag^+-ions. The measurements in 100% Pb^{2+}-ion and 60%:40% of Cu^{2+}:Pb^{2+}-ion solution demonstrate the cross-sensitivity properties of the chalcogenide glass-based Ag-sensor. The cross-sensitivity towards Pb^{2+}-ions and the Cu^{2+}:Pb^{2+}-ion mixture was about 38.0 mV/dec. and 48.1 mV/dec., respectively. The measurement time increased to several tenths of seconds, the linear range changed from 10^{-4} mol/l to 10^{-3} mol/l for measurements in 100% Pb^{2+}-ion and remained from 10^{-6} mol/l to 10^{-3} mol/l for Cu^{2+}:Pb^{2+}-ion mixture solution. The calibration measurements in Fig. 7 represent three characteristic examples from a set of 18 measurements that have been performed with different composition mixtures, as described in the experimental part (see Section 2.1).

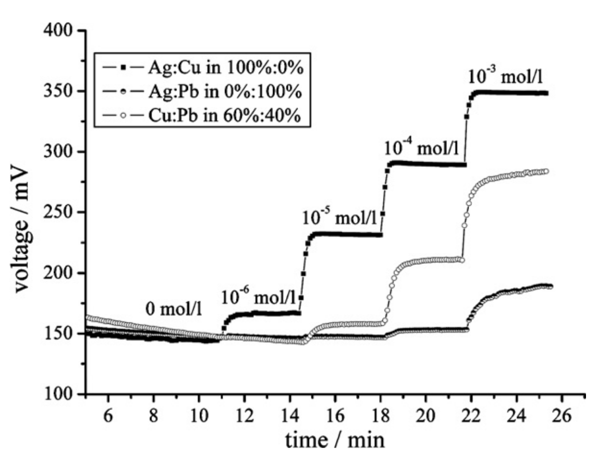

Fig. 7. Measurement curves from potentiometric measurements in different heavy metal ion compositions in the concentration range from 10^{-6} mol/l to 10^{-3} mol/l with the miniaturised chalcogenide glass Ag-selective sensor.

In order to feed the developed fuzzy logic program with calibration data, this procedure has been repeated with the Cu- and Pb-sensor in an analogue way, varying the mixture of the heavy metal compositions. The resulting cross-sensitivities and linear ranges have been determined (not shown here). With regard to its primary ion solution, the Cu-sensor had an average sensitivity of 30.0 ± 4.5 mV/pCu; thus a near-Nernstian response, which is in good agreement with previous published sensitivity values, has been achieved [2]. The Pb-sensor showed an average sensitivity of 20.0 ± 0.7 mV/pPb which is around 6–9 mV/pPb less than the expected value [2,5,8]. The decreased sensitivity (that has been constant during the experiment) is probably due to the already "aged" sensor chip used in this experiment.

3.2. Evaluation of the fuzzy logic program for heavy metal measurements

In order to validate the functionality of the developed program, a self-test was made. Here, to illustrate the results more clearly, a diagram representation was chosen (see Fig. 8). On the *x*-axis the sequence of the input data as unknown composition and on the *y*-axis the range of the proposable/known compositions are presented. Instead of calculating proposed composition proportions continuously, the suggestion of the software is assigned to a set of finite concentration ratios, namely Ag:Cu with 0%:100%, 100%:0%, 80%:20%, 20%:80%, 60%:40% and 40%:60% ratios from the lowest to the highest ion concentration followed by compositions Pb:Ag and Pb:Cu (same sequence of ratio and ion concentration) from bottom to top. Each proposal calculated by the fuzzy logic program is represented by a small rectangle. The colour of those rectangles represents the probability: black boxes mean high membership and thus high probability, while grey boxes mean minor membership (decreasing with increasing brightness).

In order to evaluate the limits of the fuzzy logic software, known heavy metal ion compositions were used as unknown data input. These input data are the same data as used for the calculation of the characteristic curves in the fuzzy logic program. The sequence of the input data in Fig. 8 on the *x*-axis (from left to right) is assigned in the same order as for the *y*-axis from bottom to top. The proposals of the fuzzy logic program for the unknown input data should ideally result in black rectangles on a diagonal. Further proposals for the same data input are shown as rectangles out of the diagonal, with the same *x*-value (input data) but with different *y*-values (output data).

Fig. 8a exemplarily shows the proposition results of the fuzzy logic program for the input data as described earlier. The diagonal of black rectangles illustrates that the highest probability was assigned to the correct compositions and concentrations by the program. However, for some compositions additional proposals with a lower probability have been calculated, too (additional grey boxes at the same *x*-value). These additional proposals of the fuzzy logic program were mainly due to unknown heavy metal solutions with a low ionic concentration. Without taking into account data from heavy metal ion concentrations of 10^{-6} mol/l, the fuzzy logic method recognised the unknown solutions with 100% probability (see Fig. 8b).

In order to demonstrate the limits of the fuzzy logic program for an acceptable analyte estimation, propositions of heavy metal ion composition and concentration equal to data from the calibration measurements but shifted with an offset of 5 mV have been calculated. The program still resulted in a relatively acceptable hit rate of approximately 80% (see Fig. 8c). Increasing the offset to 10 mV, the fuzzy logic program resulted in a larger number of different grey boxes and thus, for the estimation of the analyte solution less correct statements could be made. In some cases there were even erroneous proposals about the composition and concentration of

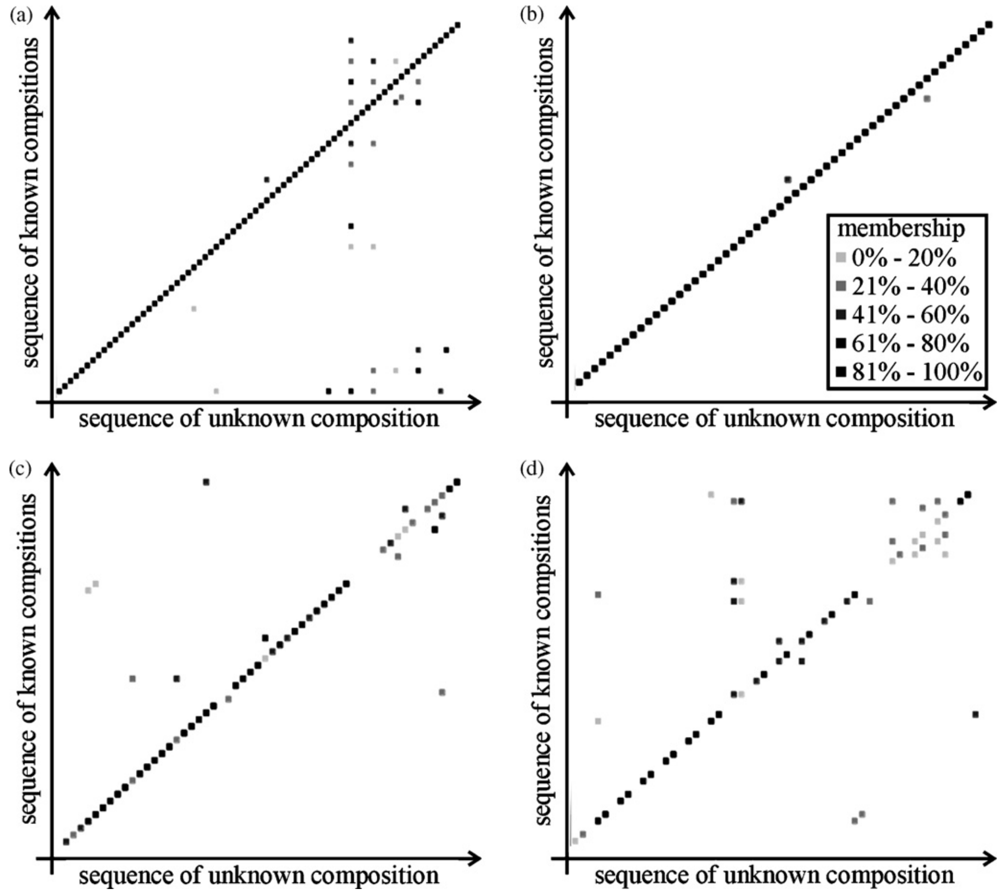

Fig. 8. Mathematical diagram of the prediction of potentiometric measurements in unknown heavy metal solution performed by the fuzzy logic program: (a) in unknown heavy metal solution with identical absolute potentials compared to calibration measurements; (b) in unknown heavy metal solution with identical absolute potentials compared to calibration measurements excluding heavy metal ion concentration of 10^{-6} mol/l; (c) in unknown heavy metal solution with an offset of 5 mV to the absolute potentials; (d) in unknown heavy metal solution with an offset of 10 mV to the absolute potentials.

the heavy metal solution (see Fig. 8d). Consequently, for the application of the developed fuzzy logic method it can be assumed that if the absolute input values of the unknown solutions differ less than $\pm$5 mV in comparison to the characteristic data used for calibration of the fuzzy program, a correct proposal about the composition and concentration of the heavy metal solution with a high probability is possible.

4. Conclusions

In this work, the development of a fuzzy logic method for analysis of combined signals from heavy metal chemical sensors has been demonstrated. The number of calibration data (database) for the fuzzy logic program could be minimised to 18 measurements for one- and two-component solutions of Ag^+-, Pb^{2+}- and Cu^{2+}-ions and is expected to be 24 for application of this program for one-, two- and three-component heavy metal solutions. Potentiometric measurements in unknown single- and multi-component heavy metal solutions have validated the property of this AI/fuzzy logic method. The proposal of composition and concentration of heavy metal ions (Ag^+-, Pb^{2+}- and Cu^{2+}-ions) in single- and two-component solutions have shown up to 80% hit rate when the measured absolute potentials in unknown solutions differ up to 5 mV from the absolute values of the respective calibration potentials. Thus, such a combination of miniaturised silicon-based chalcogenide glass sensors showing cross-sensitivities with combinatory analysis using fuzzy logic offers a relatively "simple" and fast approach towards an electronic tongue-type sensor system for heavy metal detection in environmental and industrial analysis.

In future, the developed fuzzy logic program has to be validated by means of "real" unknown heavy metal solutions e.g., waste water analysis. Hereby, the sensed absolute potential values could vary more than $\pm$5 mV from the calibration values and therefore uncertain or false statements could result from the fuzzy logic program. Thus, an advancement of the artificial intelligence program should be considered. For a further increase of the prediction certainty of unknown multi-component solutions, more a priori knowledge in terms of sensor sensitivity and weighting factors for measured potentials of a certain concentration should be included. These additional factors might improve the correctness of proposals on the unknown compositions and allow an analysis not only predominantly based on absolute values. Additionally, the quantity of sensors could be slightly enhanced in order to explore the application of this electronic tongue arrangement towards real sample solutions for environmental and food analysis such as industrial waste water analysis or the analysis of luxury foodstuffs, like wine and juice towards their originality.

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