

Software for the system of European water monitoring

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Abstract

A lack of clean water is becoming a problem in Europe. There is a need to develop easily accessible, cheap and reliable microsystems for water pollution monitoring and early warning, which could be used in most water resources on our continent. The objective of the FP5 project SEWING is to create a system of continuous water pollution monitoring in real time. Small and cheap smart sensors are being developed, which, when inserted in many locations of water resources, will collect data about pollution with a number of selected non-organic ions. CHEMFETs are used as sensors. Data processing, coding, storing and transmitting circuits are on the same chip. The system needs quite elaborate software; some of the results of its preparation are presented in this paper. The models simulating the CHEMFETs are of a behavioural type, but based on physical phenomena in water solutions of cations and anions, and in the ion-sensitive and ion-selective membranes deposited on the FET insulator. These models, after identification of their parameters through measurements, are used to prepare data fusion algorithms which give as a result the activity of particular ions in the solution. These results are stored in RAM integrated with the smart sensor construction. As the next part of software for the system, algorithms for data coding, transmission through the GSM network and final visualisation are completed. They are very flexible and can be used in different ways according to user demand.

1. Introduction

The Fifth Framework Programme for European Research and Development (FP5) strongly emphasised the problem of environment protection against pollution. This could be seen in almost all the Thematic Programmes, but particularly in the second: Information Society Technology (IST), and in the fourth: Energy, Environment and Sustainable Development (EESD). This is why 9 institutions from 7 European countries made a project application "System for European Water monitorING" (SEWING) [http 2002a] in 2000. The project was accepted, and started on September 1, 2001 with a duration of 36 months. The following institutions are taking part in the project:

- Politechnika Warszawska, Warsaw, Poland – co-ordinator
- Institute of Electron Technology Warsaw, Poland
- Technical University of Lodz , Lodz, Poland
- Valtion Teknillinen Tutkimuskeskus (VTT), Espoo, Finland
- Centre National de la Recherche Scientifique (LAAS –CNRS), Toulouse, France
- MICROSENS S.A., Neuchatel, Switzerland
- Universitat Polytechnica de Catalunya Barcelona, Spain
- Institut fuer Wasservorsorge, Gewaesserekologie und Abfallwirtschaft (IWGA) Vienna, Austria
- Systea, Rome, Italy

The project is interdisciplinary, involving specialists in chemistry, environmental engineering, computer science, electronics, and semiconductor technology. The crucial problem of CHEMFET chemical sensors has been solved after 1½ years of work. The smart sensors with several ion-selective CHEMFETs include quite sophisticated data processing, coding and transmitting circuits, and software had to be made for their operation. This paper briefly presents the main ideas used in creating this software.

2. General description of the system

The problem of environment sustainability has been approached by many research and industrial institutions all over the world. Water is one of the most polluted media on our planet, and there are many solutions concerning its maintenance, management and cleaning. Each approach to water quality needs measurement, or, more generally, monitoring. The equipment available so far is mostly of the laboratory type, and measures water samples inserted into measuring devices. More and more efforts are being made to create methods enabling water quality monitoring in the field and in real time.

The elements of the system are known and accessible. Many different kinds of sensors for water pollution monitoring have been invented, based on chemical, optical, electrical and biological principles [Wilson 2001]. Also, methods of data acquisition, processing and visualisation can be found in the literature. Looking through specialised literature, one can see how many approaches have been used, and there are still many efforts to create better, easily accessible and flexible systems of water monitoring.

On the basis of this state-of-the-art, the SEWING project is going to create a very flexible and user-friendly system for broad and general water pollution monitoring, cheap enough to have hundreds of measuring probes in a large area and collecting data in real time in a central computer. To do this a set of problems have to be solved, among them design and fabrication of sensors, selective for a given range of pollutants, their mathematical modelling, necessary for data aggregation and fusion, software for data coding, decoding and visualisation, and finally hardware realisation of the system. This is an interdisciplinary problem and its aim is to create a fully accessible and complete system for water monitoring.

The system will help in the possibility of a general European or regional policy in water management. It will be implemented and verified by end-users, and prepared for industrial implementation. The block diagram of the system is shown in Fig 1.

The probe has several chemical sensors of the ISFET type, each being selectively sensitive for different ions; their gate area is in contact with the water to be monitored. In Fig 2 two solutions of such sensors, developed at LAAS-CNRS in Toulouse, France, are shown. The voltage obtained from sensors is, in a range, proportional to the logarithm of ion activity.

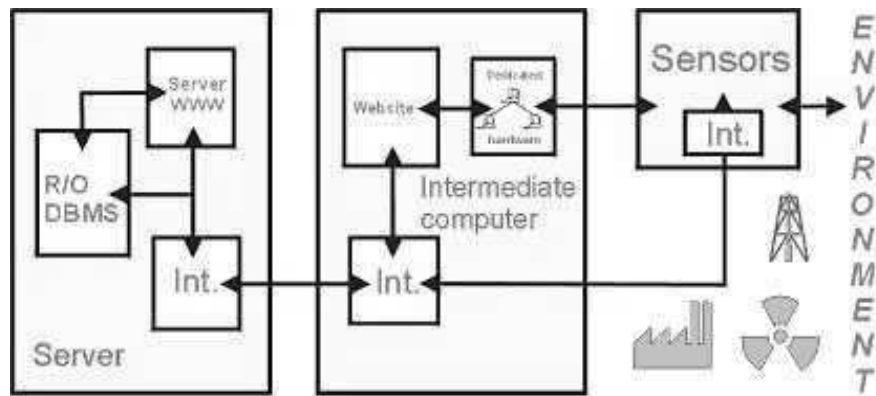


Fig. 1 Block diagram of the environment monitoring system

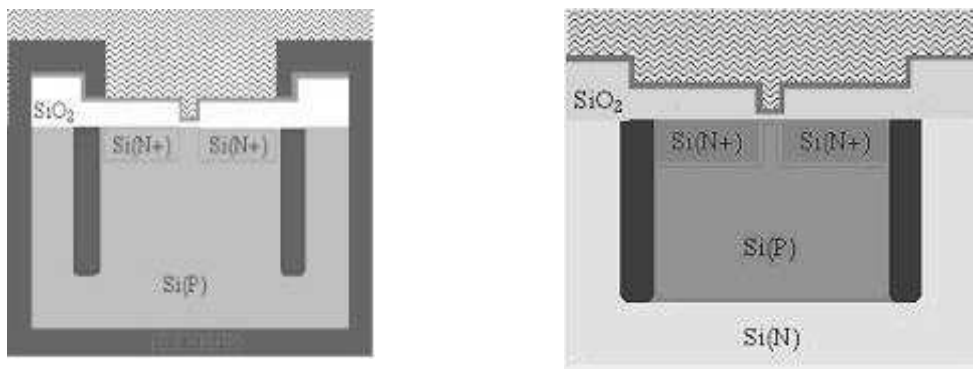


Fig.2 Two types of the ISFET construction.

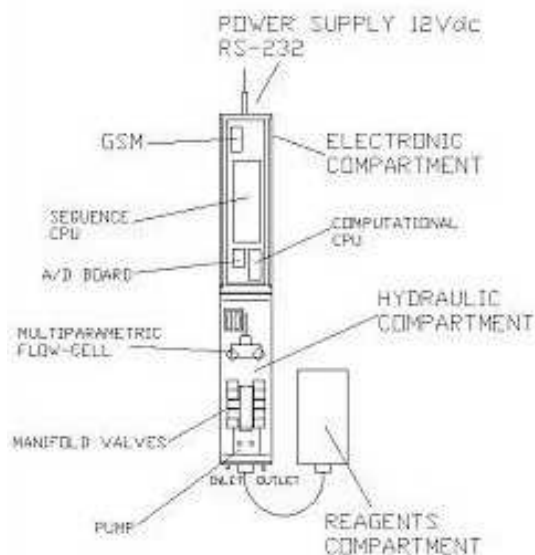


Fig 3 The construction of the probe

PREPROCESSOR	<ul style="list-style-type: none"> - Detect sensors that operate improperly and ignore them - Correct measurement results, taking into account the impact of the aging effect
MAIN PROCESSING MODULE	<ul style="list-style-type: none"> - Extract ion concentration from the measurement result
POSTPROCESSOR	<ul style="list-style-type: none"> - Reduce the output stream taking into account the relations specific to the measured phenomena – physical (intelligent) data reduction, selecting the most interesting data - Initial result post-processing (to possibly more human-friendly form)
CODING AND COMPRESSION	<ul style="list-style-type: none"> -Reduce the amount of stored (sent) bytes trough a raw byte stream compression (strictly mathematical compression, without data interpreting)
STORAGE AND/OR TRANSMISSION	<ul style="list-style-type: none"> -Store the post-processed results - Or arrange the data to be transmitted in large packets (store them in the output buffer) and transmit them as the buffer becomes full

Fig 4 Data processing algorithm

Interface (Int.) consists of analogue and digital circuits extracting pertinent information about water pollution. This information, after appropriate coding, is stored in a RAM. A GSM transmitting system sends it to the intermediate and server computers, where it can be visualised. The concept of the smart sensor, elaborated by SYSTEA in Rome, Italy, is shown in Fig 3. Software to be used in the system is being done by several partners. In Fig 4 the block diagram of the data processing algorithm, proposed by Technical University of Lodz, Poland, is shown. In the next sections of this paper software for SEWING project, prepared by the Authors at Warsaw University of Technology, is presented.

3. Software for CHEMFET sensor systems modelling

This section outlines an original mixed-domain model of CHEMFET devices. The model not only correctly senses ionic activity in a tested aqueous solution at a fixed operating point. It can also closely approximate the influence of the operating point on CHEMFET sensor responses. CHEMFET devices are built upon the silicon structure of a gateless MOSFET gate, as sketched in Fig 5. The gate insulator is directly covered with a hydrogel, containing an aqueous solution of the internal electrolyte. The hydrogel is, in turn, covered with a special ion-selective membrane, which directly contacts the aqueous test solution. Ideally a chemical sensor should be specific, i.e. it should respond to a strictly specified (main) ion activity only.

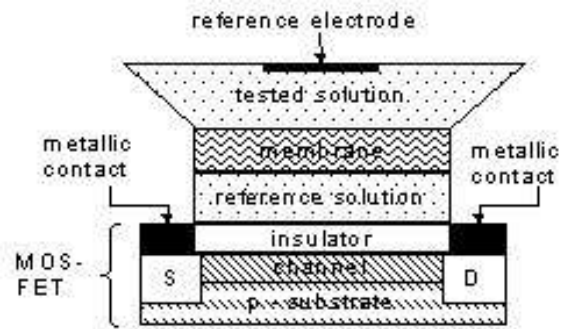


Fig. 5 Schematic CHEMFET structure

3.1. Computer model of the CHEMFET chemo-electrical transducer

In reality the potential drop between each electrolyte and the membrane surfaces also depends on the activity of other (interfering) ions. This effect, modelled by Van den Berg, [Berg 1988, Ogrodzki 2000] can be approximated in practice by the well-known Nikolsky-

Eisenman (NE) model [Nikolsky 1937] $E_B = \psi_0 \ln(C_1)$, where $C_1 = \sum_{j=1}^m \beta_j k_{Cj} a_{Cj}$ depends

on the tested ions activity a_{C1} , interfering ions activities a_{Cj} , division constants for cations k_{Cj} , electro-thermal potential $\psi_0 = n_i kT / q$ (with Boltzmann constant k , Kelvin temperature T , electron charge q and nonideality index n_i) and complexation coefficients β_j . Recently, the appearance of more accurate measuring systems encouraged us to improve the accuracy of the Nikolsky model to a super-Nikolsky-Eisenman (SNE) model [Ogrodzki 2002, Ogrodzki 2002a], with a lipophylic salt influence consideration:

$$E_B = \psi_0 \ln(e_B),$$

where:

$$e_B = C_1 \left[\frac{\bar{a}_{Ltot}}{2\bar{a}_{Ytot}} \left(1 + \sqrt{1 + 4\bar{a}_{Ytot} \frac{C_2}{C_1}} \right) - 1 \right], \quad C_2 = \sum_{j=1}^m K_{aj} \beta_j k_{Cj} a_{Cj},$$

\bar{a}_{Ltot} is the total activity of ligands in the membrane, \bar{a}_{Ytot} is the total activity of the lipophylic salt, and K_{aj} are association constants. The quality of the SNE and NE models is shown in Fig. 6.

A more accurate SNE well explains the small and medium ion concentration range, with accuracy w.r.t. the Van den Berg (VDB) model better than 0.3% of the range. The accuracy of the NE approximation in this region is worse, since the error level is about 1.5% and increases when the ratio C_2/C_1 , i.e. the contrast between association constants, increases. The high-concentration knee seen in Fig 6 in practice lies above the maximum level for useful activity, and so is not observable.

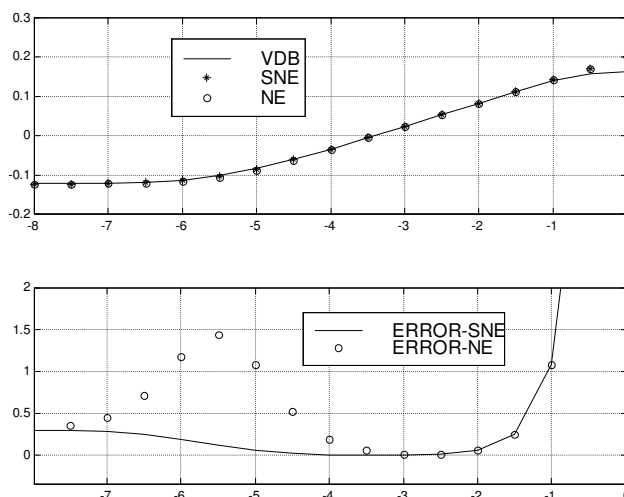


Fig.6. Comparison of NE, SNE and VDB models

3.2. Computer model of the internal FET

To reduce influence of the internal MOSFET properties on the CHEMFET sensor response, the operating point is typically fixed by a biasing circuit of the sensor, i.e. the drain-source voltage UDS and the drain current ID are stabilized. For such a mode of operation the CHEMFET device can be represented by a mixed-domain (chemo-electrical) model where the chemical part is not dependent on the electrical part, so the gate-less MOSFET device can be viewed as separable from the transducer, which deals with the conversion of ionic activities in the tested solution into a voltage drop VGR between the gate insulator surface and the reference electrode terminal. Typically, the dependence of VGR on ionic activity follows the NE or SNE equations. However, for rational design of a CHEMFET based sensor, it was necessary to develop a CHEMFET device model [Opalski 2002a] which does not place such constraints on biasing.

A simple SPICE LEVEL=1 model exhibits 6 to 10% of maximum relative error (of the drain current), especially for higher levels of ionic activity (>0.01 mol/l), making the model unsuitable for further investigations. A poor fit for low URS values could be attributed to the depletion type construction of the internal gate-less MOSFET. Unfortunately, precise modelling of such a device is very complicated [Klaassen 1986, Tol 1991]. Taking simplicity as our main concern, a depletion MOSFET model described by Merckel [Merckel 1977] was adapted instead. It contains 7 parameters.

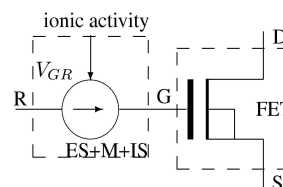


Fig. 7. A separable CHEMFET model

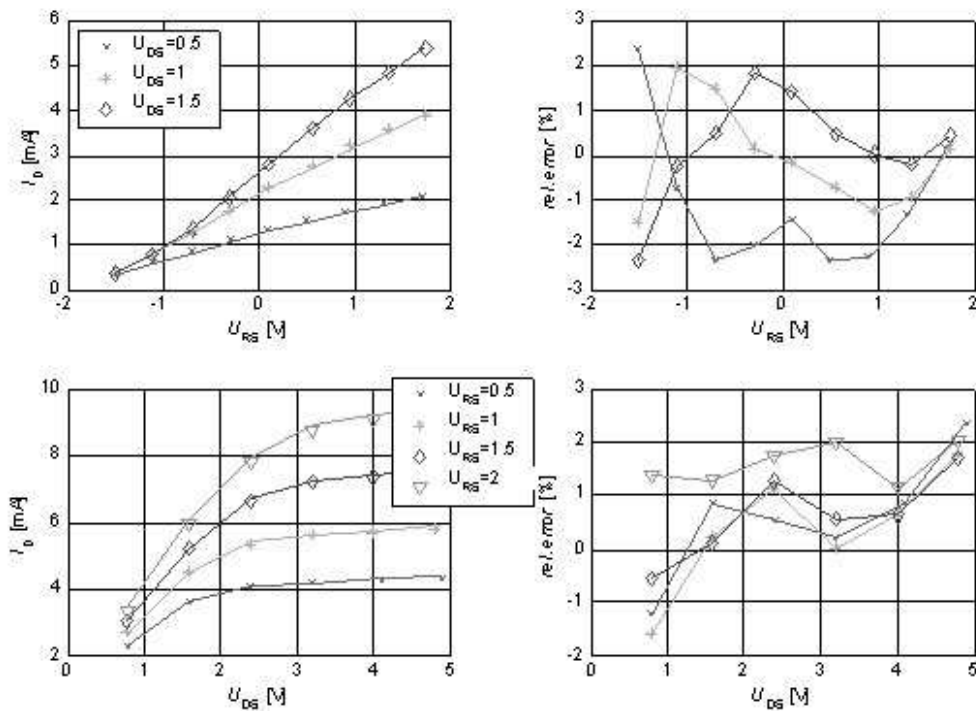


Fig.8. The chemo-electrical CHEMFET model fitting (left) and accuracy plots (right)

For use as a separable CHEMFET model one should replace UGS with $URS + VGR$, where URS is the reference electrode-source voltage and VGR models the transducer activity. A separable CHEMFET model composed of the Merckel MOSFET model with the NE type membrane model shown in Fig. 7 did quite well for modelling measurements with variable ionic activity and operating point of the device. Joint approximation of transfer and output electrical characteristics for seven activity levels of the main ion variable in the range 10^{-6} up to 10^{-1} mol/l can be observed in Fig. 8, where the Merckel model (solid lines) is fitted to measurements (at markers). The plots correspond to a concentration of potassium ions equal to 10^{-6} mol/l, and a concentration of interfering sodium ions = 0.1 mol/l. The composite model exhibits a maximum relative error of 4.9% for the whole range of concentration variations – which is only slightly larger than for a single activity level fit (4.6%). For the narrower range 10^{-6} up to 10^{-2} mol/l the error was only 3%. The model has been positively verified for a depletion mode MOSFET-based CHEMFET device. The subtle dependence of a sensor response on the operating point was observed [Opalski 2002b] and is considered in more accurate behavioural models. Implementation of this effect enables a designer to optimise the sensor bias point on the CHEMFET system design stage.

4. Software for model identification and measurements database management

4.1. Automatic measuring system software and measured data processing software

For quick measurement of a large number of CHEMFET sensors, dedicated automatic equipment has been constructed. This equipment consists of a hydraulic system for automatic dosage of chemical compounds, a measuring head including sockets for a few sensors and programmable analogue-digital equipment for supplying sensors, measuring their currents

and voltages. The set is controlled by the LabView PC-based application which generates XML data files. For storing and quick access to the measured data a relational data base has been designed. At the moment it is arranged in the form of an MS ACCESS database operated from a MATLAB database controller application via the Database Toolbox. This controller application enables: a) importing XML measurement files and generation of SQL instructions, b) manual, automatic or mixed data filtering for elimination of noisy and faulty data, c) determination of chemical characteristics of sensors using filtered chemical-electrical measurements (transfer and output characteristics for different concentration levels of maximum 3 ions), d) identification of sensor model parameters by means of a MATLAB based special purpose characterization tool, e) statistical analysis and visualization of sensor parameters, f) storage of both the results of the data processing operations and also of processing algorithms (filtering, optimization). Saving parameters of algorithms may enable tuning of data processing algorithms for maximum efficiency and accuracy.

4.2. Model parameters extraction software

Software for CHEMFET sensors model identification consists of the following two main applications dedicated to: a) initial rough pre-extraction of model parameters from chemical and electrical characteristics, b) sensitivity analysis of the identification task and c) exact optimisation-based extraction. Sensitivity analysis of the sensor model shows that the identification task is well conditioned when primary model parameters are transformed into secondary parameters [Ogrodzki 2002]. Essential among these are those strongly influencing the output sensor voltage according to the sensitivity analysis performed in [Ogrodzki 2002] (relative sensitivities given in parentheses): selectivity ($\sim 10^{-2}$), offset voltage ($\sim 10^{-1}$), nonideality index, association constants ($\sim 10^{-2}$), concentration of ionophore ($\sim 10^{-2}$), concentration of a lipophylic salt ($\sim 10^{-2}$), normalized division constants for anions ($\sim 10^{-2}$). Other parameters have less influence.

Chemical characteristics provide a quick rough identification of the following parameters:

- Nonideality index available from the slope of $U_{GS}(a_{C1})$ in the linear region.
- Offset voltage E_{offs} estimable from a point selected in the linear region $E^1 = U_{GS}(a_{C1}^1)$.
- Association constants obtainable from the plot $d \exp(-U_{GS}) / da_{C1}$.
- Interfering cations contribution $w = \sum_{j=2,m} K_j a_{Cj}$ estimable from points chosen on the

low concentration knee.

Exact identification, starting from the results of an initial identification, was performed by means of an optimization procedure fitting the drain current. As an objective function we used a sum of squares over all measured points of electrical and chemical characteristics. Minimization was done by the MATLAB optimization toolbox.

5. "Data fusion" software for chemical information extraction from electrical data

Much effort has been expended in the field of analytical chemistry towards the development of selective sensors, which respond only to one analyte while ignoring all interferents that are present in the samples [Forster 1991, Fung 1986]. Obviously, real sensors suffer from some degree of nonspecificity - thus allowing some inaccuracy of measurement. There are two ways to reduce the inaccuracy: 1) through further development of more selective sensors, and 2) through appropriate processing of raw signals coming from a collection (array) of sensors which are selective to different ions - so called "data fusion"

[Opalski 2002]. In what follows we assume that the ionic content of the electrolyte is to be determined from the responses of N sensors, each one selective to a different ion. The dependence of response E_i of the i -th sensor on activities a_j of M ions in the solution can be modelled approximately by the NE equation. Its offset voltages are determined in the course of a calibration process. Electrolyte characterization, based on N separate measurements, is affected by the systematic error due to nonideal sensor specificity.

The error can be reduced by reduction of selectivity coefficients and an increase in sensor stability - through development in sensor technology. We are interested in accuracy improvement derived from joint processing of the N raw measurements. Note, that if activities of all interferences were known for the i -th sensor then, ideally, exact value of a_i could be derived from sensor equation.

The electrolyte characterization problem can be, ideally, considered as a solution of a set of nonlinear equations relating activities of ions \mathbf{a} and responses of sensors \mathbf{E} . Only ions of the same charge sign are taken into account, i.e. for an anion-selective type of sensor, only ions with a negative sign. An “exponential” form of sensor matrix equations is particularly interesting for $M = N$, when all ions are of the same charge for $i = 1, \dots, N$, because the equations take the linear form: $\mathbf{v} = \mathbf{K} \cdot \mathbf{a}$. If we knew \mathbf{v} exactly, i.e. the responses of the sensors \mathbf{E} , offsets \mathbf{E}_0 , temperature T , and selectivity coefficients of the sensors collected in the $N \times M$ matrix \mathbf{K} , then estimates of ion activities \mathbf{a} could be determined exactly. In other words, responses observed in a solution of known composition can be perfectly decoupled into contributions arising from analytes and interferences.

If the NE model is accurate then $\mathbf{a} = \mathbf{K}^{-1} \cdot \mathbf{v}$ defines a “data fusion” technique which is free from the systematic characterization error due to interferences. Taking into account interferences and also voltage measurement errors, it is seen that the “data fusion” always leads to a smaller inaccuracy, although the advantage is reduced with an increase of voltage measurement errors.

In the above analysis voltage error can in fact express not only pure voltage measurement error, but also the error (inadequacy) of the idealized sensor model and the inaccuracy of the offset parameter in the sensor model. Evaluation of the characterization error due to inaccuracy of the selectivity coefficient matrix requires a separate analysis, though.

To make presentation clearer let us only consider the perturbation of a single off-diagonal entry K_{ij} of the \mathbf{K} matrix. The perturbation of K_{ij} does not affect the accuracy of \mathbf{a} at all. Unfortunately, error analysis for \mathbf{a} is more complicated. For high selectivity sensors the total characterization error is determined by the voltage measurement error.

The “data fusion” concept has been shown to increase the accuracy of estimates of ion activities in a solution of known ionic contents, especially for larger selectivity coefficients and large concentrations of interferences.

If the number of ions (with proper charge sign) M is larger than the number of sensors N it is not possible to determine ion activities uniquely, although it is possible to put a bound on inaccuracy for a particular characterization method. In such a case \mathbf{a} contains an additional systematic error component due to the extra $N - M$ ion activities, but the inaccuracy is smaller than the respective systematic error in \mathbf{a} .

6. Software for information storing, coding and transmission.

Today's solutions in the newest (2nd, 3rd) generations of mobile phones has enabled three main ways of communication: CSD (Circuit Switched Data), GPRS (General Packet Radio Service) or point to point communication between wireless terminals. They are included in all SMS/MMS messages and other Push-Pull technologies. In short, internet access or other "machine" access by mobile phone and WAP is similar to WWW and radio wireless model communication (see Fig. 9).

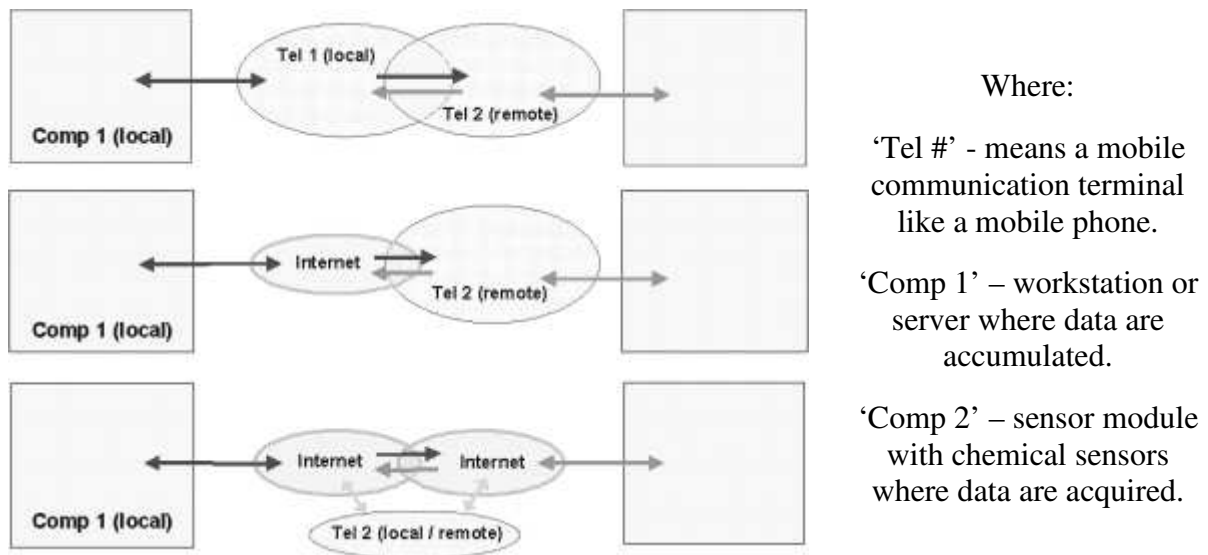


Fig. 9 Communication model: GSM and Internet application.

These three types of connection are representative for any implementation. In this way we can establish any required link (hard, semi-hard or on demand e.g. dial-up) between parts of the system. In practice, to establish a link, three main steps are to be taken. The first step is the choice of GSM operator. In this case the GSM, ERA operator was chosen [http 2001]. The next, mobile phones used as terminals (for tests one can use the mobile hand phone, M2M – Machine to Machine module or GSM module). And finally, to use an appropriate setup [Goralski 2002] (for both: GSM phones and computer), in order to establish a GSM wireless link and access to the Internet. All tests were successful in this project and proved that the first approach to the design was correct.

Standard WAP solutions, available in basic configurations, are not flexible enough for some purposes. The use of graphical data visualisation is more efficient and clear in comparison to raw data. Using tables and charts, a very versatile and useful, smart and short application can be constructed, which can be implemented or modified by different developers in a similar way as the RAD method application development [http 2002].

The WAP technology and PHP/ASP scripts give interesting choices for data visualisation on a mobile phone display (Fig. 10). In the next generation of phones the colour display will be standard. The scripts made before can easily be modified for colour visualisation.

Finally, by creating a WAP source-code, a working pre-prototype application with data transfer simulation was done. On that test stage, we can transmit data between the

terminals by using the FTP protocol, which means that the data can be transmitted through the links in a bidirectional way between the sensor side and e.g. centre database server.

During the tests, M2M (Machine to Machine) technology was also recognised for target use in the SEWING project. The M2M module is a very valuable GSM mobile phone for some professional use. It is usually without classical display and keyboard, but of course it is suitable for practical use e.g. with stronger waterproof casing.

The tests identified all required steps predicted in the first design approach, and real implementation of a measuring system working in the natural environment: 1) software and hardware, 2) configuration, 3) additional requirements and tests, 4) elimination of incorrect ideas in development studies, and 5) cognitive interdisciplinary studies.

The development method in building the solution was as follows: 1) integration and simplicity of structure, application and coding, 2) checking the results at every point during the development stage, 3) freeware software use: e.g. GSM Phone Simulators.

The following elements were used and integrated in the pre-prototype: 1) graphical user interface, 2) miscellaneous components for data visualization, 3) other languages support (PHP/ASP – data and databases processing), 4) GSM connectivity (CSD/GPRS - APN), 5) R&D works for Push-Pull Technology, Mobile SVG etc. as well as for suitability, 6) an encapsulation data transfer protocol in WAP code in GPRS connectivity, 7) verification of delays for ODBC and wireless GPRS connections for the designed application, 8) analysis of the cost of tests, ergonomic rules, and implementation.

In the next generation of GSM networks the colour display will be used most often. The possibility of colour visualization was tested by techniques like those mentioned below. Future browsers implemented in mobile phones will be similar to those used for WWW creation (static and dynamic mode): 1) XHTML, 2) Java embedded application, 3) MMS for additional control, and 4) Mobile SVG.

The application of wireless GSM networks as a medium of connection in a heterogeneous and territorially distributed network system for measurement is both suitable and proper for technical and economical reasons.

All tests performed fulfilled the expected requirements. Because of this, all the achieved results will be implemented fully in the SEWING system project, in the near future.

The next steps to be taken for the development of the current results and achieved solutions will be M2M and Bluetooth modules application in real environment implementation. Only a flexible use of M2M modules and classic hand mobile phones gives

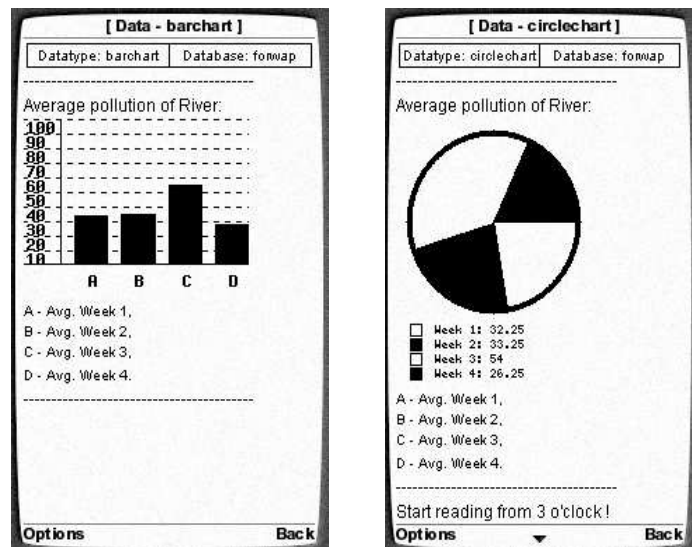


Fig. 10 Screenshots: Bar Chart and Circle Chart view.

us an advantage in administering, data acquiring and supervising system activity, and data transfer on demand or alarm mode.

7. Conclusions

The SEWING project solves many problems in the field of effective water pollution monitoring. The first of these is the technology for creating ion-selective sensors. Other problems are software resolvable and they have been outlined in this paper. The project required development of the following efficient software.

- 1) **Software for automatic sensor measurement, mathematical modelling, and parameter identification for CAD.** An important conclusion from this part of the work was that great attention has to be paid to careful realisation of chemical measurements, since problems with the flow of fluids, air bells, and imprecise temperature stabilisation, to a quick measuring process in comparison with the time constants of chemical and hydraulic phenomena leads to quite unusable data for model identification.
- 2) **Software for chemical data fusion from a matrix of sensors, monitoring different ions, dedicated to chemical data extraction.** great attention has to be paid to smart reduction of interfering ions influence. Dedicated methods have to be developed.
- 3) **Software for transfer, storage and user-friendly visualisation of monitoring results.** Standard WAP solutions, available in basic configurations, are not flexible enough for some purposes. Investigation done in this project brings an important conclusion that the use of graphical data visualisation is more efficient and clear in comparison to raw data. Using tables and charts a very versatile and useful, short and smart and application can be constructed, which can be implemented or modified by different developers in a similar way to the RAD method application development [http 2002].

These elements create a flexible, accessible, open and easy to use system, which will help management of water quality at national, regional and local level.

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