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# 30 YEARS WITH CARBON PASTE ELECTRODES AT THE UNIVERSITY OF PARDUBICE

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*In this review (with 425 Refs), three decades of the electrochemistry and electroanalysis with carbon paste electrodes characterising the research activities of the electroanalytical group at the University of Pardubice (EAG UPa) are summarised, when presenting all the achievements and, where applicable, also highlighting research outputs throughout the time. In more detail, reminded are the very beginnings in the mid-1980s and the early era at the University of Chemical Technology. The article contains the complete list of publications plus a great majority of conference presentations associated with the field and having been prepared by the EAG UPa in the period 1987-2016.*

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## Introduction

Carbon paste electrodes (CPEs), known for nearly six decades [1], are now being classified as a special kind of carbonaceous electrodes having already achieved the status of "traditional" or even "classical" type of indicatory / sensing / detection unit in electrochemical and/or electroanalytical measurements. Consensually, CPEs are one of the most popular type of electrodes / sensors thanks to their valuable physicochemical and electrochemical properties, together with the fact that they can be made practically in every laboratory; often, at minimal expenses and *via* simple procedures.

The development of CPEs in the 1960s is forever associated with the name of Ralph N. "Buzz" Adams, their inventor [1] and great propagator in organic electrochemistry in the 1960s and early-1970s whose pioneering and fundamental compilations are sorted to the classic files in electrochemistry [2,3]. Also, Adams's farewell to CPEs, the extensive article on their specific reaction kinetics [4], belongs amongst the most cited paper in the field. After his era, largely highlighted in the electroanalytical literature (see e.g. Refs [5,6]), carbon paste-based electrodes, sensors, and detectors had gradually attracted attention of scientists throughout the world. This is well documented on a series of reviews surveying the individual periods and the respective applications in various areas of pure and applied electrochemistry [7-19].

● Based on the Thomson Reuter's electronic databases, namely Web of Knowledge® and its sub-portal Web of Science® (WoS [20]), as well as on the deep literature search utilised in the first monograph on CPEs [21], it can be estimated that CPEs and related configurations are the subject of about 3,500 scientific reports, with a continuing growth of 100-150 articles each year. By paraphrasing introductory words in the latter source [21], one can state that "*There is scarcely another type of the electrode whose employment would illustrate more faithfully the overall progress, the individual movements, and trends in electrochemical and electroanalytical measurements over the half a century than carbon paste-based electrodes…*"

## Carbon Paste Electrodes at the University of Pardubice

The entitled thirty years of using carbon paste electrodes by the electroanalytical group at the University of Pardubice (further abbreviated as EAG UPa) are reflected in the following collection of scientific outputs:

- *monograph* [21], being already mentioned above;
- *book chapters* [22-30], of which three appeared in special textbooks [22,24,25], one in an encyclopaedia [23], and other five in compilation

monographs [26-30];

- *reviews* or related articles [31-59], comprising general as well as specialised texts;
- *patent* (concerning the construction of carbon paste electrode holders) [60];
- *unpublished results* (archived by the first author) [61];
- *standard publications* [62-235], gathering (i) *original papers* published in impacted international journals, (ii) *original papers* from peer-reviewed local periodicals, plus (iii) *conference contributions and related articles* written in the full-text format;
- *very first report* appearing within EAG UPa [236];
- *diploma*, *dissertation*, and *habilitation theses* [237-311], where carbon paste was the central topic or, at least, one of the main topics;
- *presentations* on conferences and seminars in the form of oral contributions, or longer plenary lectures, and as posters; all being gathered in one block of citations [312-412].

● In a brief estimate, the publications surveyed above are cited in 3,000 articles registered at the WoS, when some articles prepared with cooperating scientists have already achieved the outstanding citation profiles[2] : Ref. [36] … 464×, Ref. [43] … 228× , and Ref. [50] … 161× .

## The Very Beginnings of Carbon Paste at the University of Chemical Technology

In the mid-1980s, scientific orientation of the EaG UPa had been divided into three different areas of electrochemical experimentation having, at that time, profiled the following research and pedagogical activities:

(1) *potentiometry of pharmaceuticals and surfactants*; typically, with the so-called *coated-wire electrodes* (see e.g. Refs [413,414] and Refs therein);
(2) *advanced studies on chemical equilibria* by computer-assisted treatment and evaluation of electrochemical data (e.g. from potentiometric titrations [415,416]);
(3) *occasional analyses* of real samples performed upon request and employing *faradic (current-flow) measurements* [417].

The experimental work within the third category had also included an effort to develop "own" working electrodes for voltammetric and coulometric measurements, which could lower the dependence on commercial offer within the

[2] Data from mid-2015 (according to the first-author's archives [61])

East-Block provenience, where such a portfolio was limited and the quality of accessible electrodes not always satisfactory.

During the respective investigations, two similar configurations had been of interest: (i) *carbon composite electrode* [418] and a (ii) *carbon paste electrode* [236,237]. The first type was made by mixing the powdered graphite (with particles in the μm-scale) with a monomer, when the respective mass had been left to polymerise and solidify in narrow glass probe with inserted metallic wire. After hardening, the glass form had been broken, removed, and the resultant rod polished into an electrode of common pencil-like shape and with already integrated electric contact. The second heterogeneous mixture was a *carbon paste* prepared by manual homogenising of the same graphite powder with a liquid binder. A trio of possible binding agents had been tested and highly viscous silicone oil finally chosen; also, according to the carbon paste mixtures proposed and successfully used in the 1960s by Middle European electrochemists *Farsang* and *Monien* (see Ref. [21] and Refs therein).

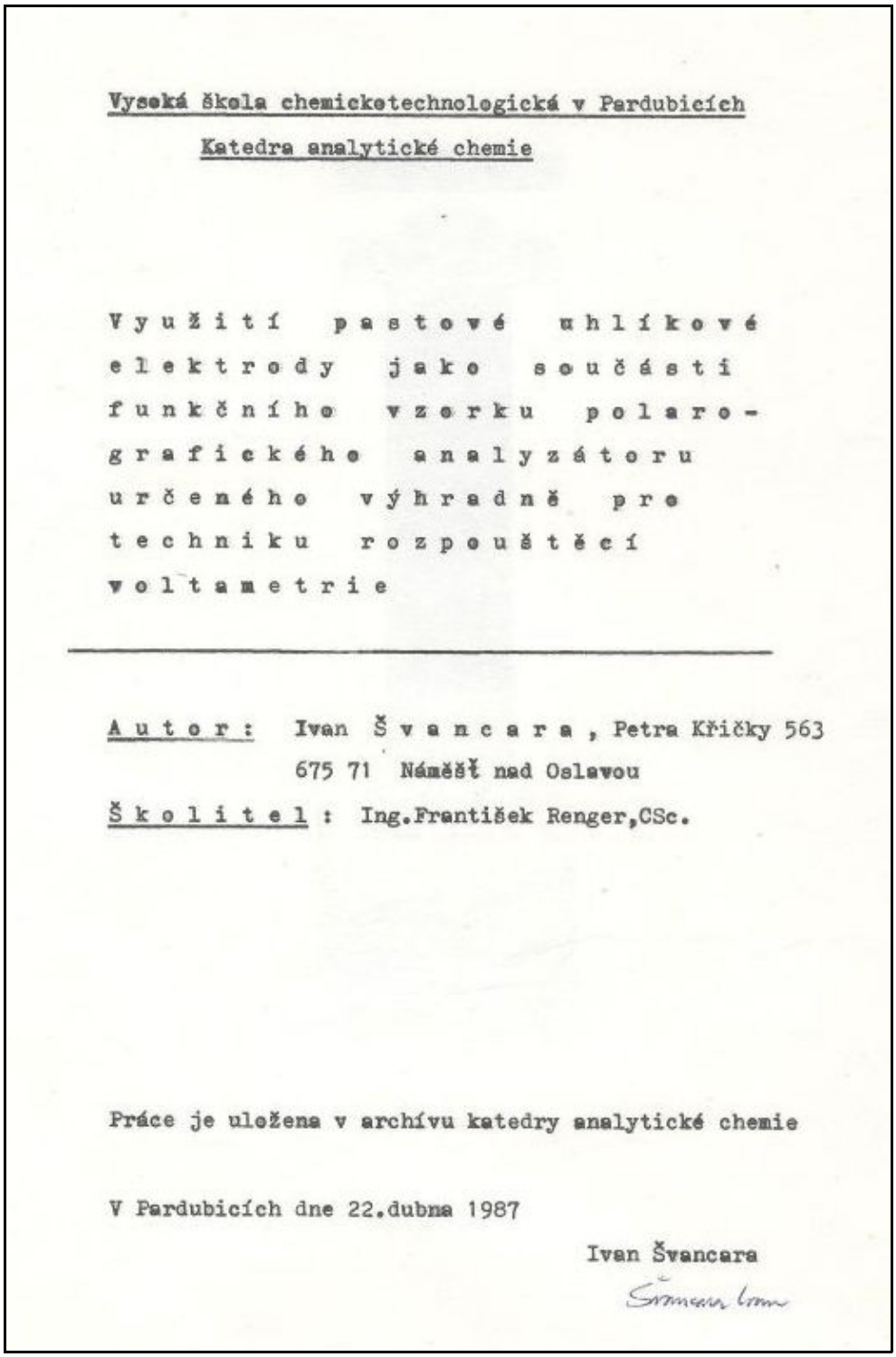
For both heterogeneous electrodes, the carbon moiety was identical and obtained from Tesla Lanškroun — a local manufacture in the Eastern Bohemia where such graphites had been used for printing of electrical multi-layered resistors. The binding agents was either (i) methyl *m*-acrylate with an alkyl-benzoyl peroxide as catalyst [418] or a (ii) silicone fluid obtained from Lučební závody Kolín [236,237] — chemical plants near Pardubice producing at that time various silicone fluids for industrial use under the trademark Lukoil®.

Both types of heterogeneous electrodes had then been subjected to the initial characterisation to define their performance in the configuration of *in-situ* operated mercury film electrode(s) for electrochemical stripping analysis of some heavy metal ions at the nanomolar concentration level; namely, $Cd^{2+}$, $Pb^{2+}$, $Bi^{3+}$, and $Cu^{2+}$. With respect to carbon paste-based variant, the pioneering experiments had been performed with a CPE with electrode body made of a plastic syringe, soon replaced by special piston-driven construction of own production [60,129] (and loosely inspired by the Monien's design).

All the initial experiments were described in two texts. The first one was a *contribution to the traditional students' competition* [236] — by the way continuing at the Faculty of Chemical Technology of the University of Pardubice up until now [221] — where the above-specified CPE had also been emphasised in the title and, in experimental work, served for all the measurements. The second text, having appeared a few weeks later, was a diploma work [237] reporting on a series of measurements with CPEs but not specifying the working electrode of choice in the official title. Retrospectively, the work submitted and defended in the students' competition[3] [236] can be regarded to represent really *the very first*

[3] In fact, this contribution did not achieve any notable success in the final confrontation of similar students' works from Czech and Slovak chemical universities held in Bratislava, in the spring of 1987

*report* dealing with a carbon paste electrode and published at the academic ground in the city of Pardubice; see also Fig. 1.



Vysoká škola chemicketechnologická v Pardubicích

Katedra analytické chemie

Využití pastové uhlíkové elektrody jako součásti funkčního vzorku polarografického analyzátoru určeného výhradně pro techniku rozpouštěcí voltametrie

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Práce je uložena v archívu katedry analytické chemie

V Pardubicích dne 22.dubna 1987

Ivan Švancara

Švancara Ivan

Fig. 1 Title page of the very first report on carbon paste electrodes published under the umbrella of University of Chemical Technology in Pardubice (see Ref. [236])

The premiere report itself [236] comprises a concisely commented collection of stripping voltammetric measurements, from which the prevailing part of experiments offers only average results, revealing little experience with heterogeneous electrode materials and therefore not properly optimised experimental conditions. (This, of course, can be understood as the work presented in this pioneering file had described truly and in pretty authentic way the very first steps in brand new field.)

On the other hand, there is one highlight included in and featuring excellent electrochemical stripping characteristics of bismuth — at that time yet unforeseen sign of future research orientation within the EAG UPa; see Fig. 2 and further text.

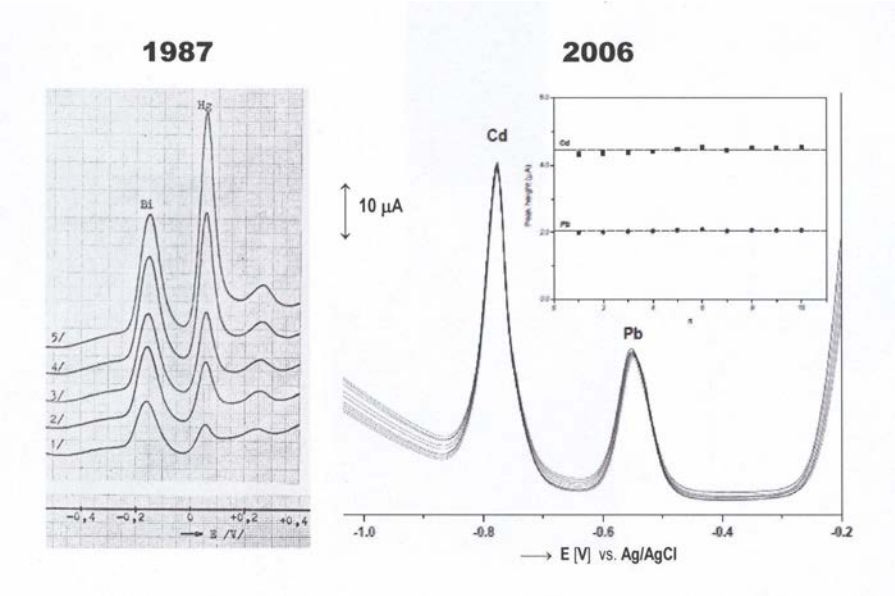


Fig. 2 Carbon paste electrodes *vs*. bismuth or else: Two experiments divided by two decades; left … Model calibration of $Bi^{III}$ species at the nanomolar concentration level detected with mercury-film plated carbon paste electrode in combination with anodic stripping voltammetry (taken from Ref. [236]); right … Voltammetric stripping analysis of $Cd^{II}$ + $Pb^{II}$ in model mixture at the ppb level ($\mu g l^{-1}$) and reproducibility test with bismuth-film plated carbon paste electrode (taken from Ref. [305])

## Experimentation with Carbon Paste Electrodes at the University of Pardubice: A Survey of Research Areas of Interest

Thirty years of more or less intensive experimental work with CPEs and related configurations has naturally been reflected in a widespread research that had covered a number of diverse areas across the electrochemistry and electroanalysis.

Below, the respective activities are surveyed, when the representative references cited chronologically are typically the principal contributions — usually initial reports — plus other closely associated items, e.g. monothematic reviews, the continuing studies and practically oriented papers. Finally, mentioned are also

some citations that contain the original drawings and images used herein to assembly collages in Figs 3-5 (see overleaf). Thus, the individual areas of interest given are as follows:

- *Basic and advanced characterisation of traditional carbon paste mixtures* by using standardised tests for recommended model substances and/or redox systems in combination with common voltammetric techniques (see e.g. Refs [32,63,89,91,106,130,156,243,275]);

- *Proposals, testing, and applications of new types of carbon paste mixtures* containing alternate and sometimes even brand new main components (e.g. Refs [28,64,71,167,178,214]);

- *Development, testing, and applications of specific configurations of carbon paste electrodes*, such as special modifications of carbon pastes or metallic-film plated carbon paste electrodes used in electrochemical stripping analysis for determination of various (heavy) metals ([40-42,47,67,74,98,110,112,116,128,136, 145,164,181,215] and also Fig. 3);

- *Construction and employment of new and/or further innovated CPE-holders and similar assemblies* mostly connected with subsequent manufacturing and production in mechanical workshops at the University of Pardubice [60,126,129, 144,153,238,276];

- *Special microscopic studies with carbon pastes*, either with numerous types of carbon pastes as such or with CPEs as substrates for metallic films and some special modifiers (see Refs [71,120,125,147,303,304] and also a collage in Fig. 4);

- *Basic and advanced characterisation of carbon-paste based indicatory electrodes for (equilibrium) potentiometry* (e.g. Refs [69,83,87,229,235,244,297]) and later also for stripping potentiometry (see Refs [86,100,106-108,113,155,298] and also Fig. 5, second row);

- *Development, preparation and applications of new and/or innovated types of carbon paste-based biosensors*; operated either as a batch arrangement in the amperometric hydrodynamic mode (HA [70,75,77,114,172,200]) or as detection units in flow injection analysis (FIA [78,153,203,282]) and for liquid chromatography (HPLC [13,72,149]);

- *Proposals, development, and testing of new and/or innovated methods of inorganic electroanalysis*, in a majority devoted to environmentally important ionic and molecular species (see Refs [62,63,66,73,76,79,80,94,95,99,102,108, 121,122,150,151,154,176,189,205] and also Fig. 5), but concerning also some precious metals in technical samples [65,103,137], (bio)essential elements in foodstuff [85,92,111,121] or pharmaceuticals [88,104,246].

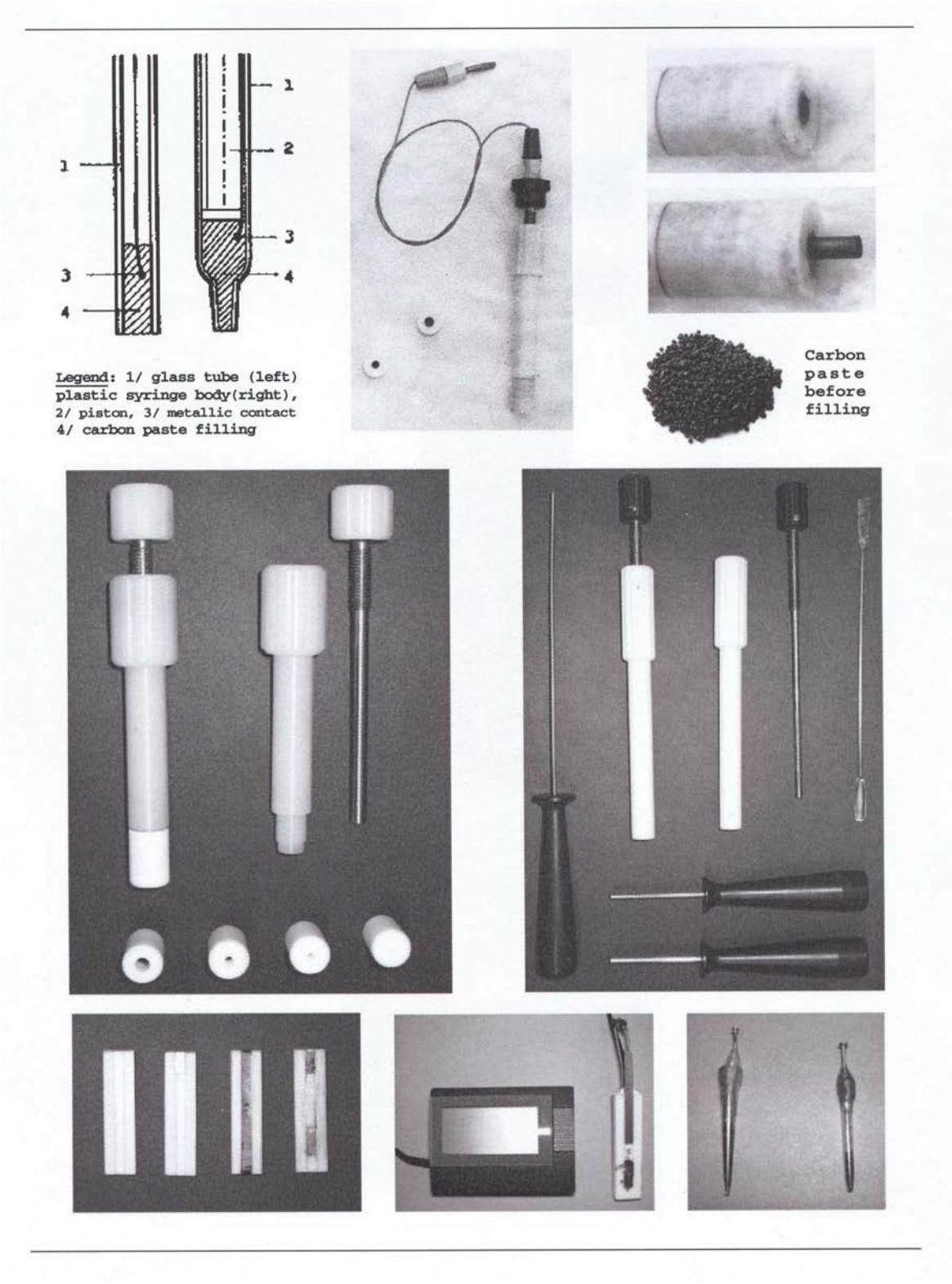


Fig. 3 Carbon paste electrode holders designed and manufactured in the workshops at the University of Chemical Technology (1987-1993) / University of Pardubice (1994-2016); upper row … First prototypes, with details of the electrode tip (used also on the cover of the first book on CPEs [21]); middle row … Two sets of the most successful designs and accessories; bottom row … special miniaturised configurations. (The individual images taken from Refs [21,60,126,129,144,160,238,310] and, in some cases, newly rearranged.)

- *New methods focused on* studies and determinations of *organic synthetic compounds* [124,142,212,225,240,241,291]) and *environmental pollutants* [158,174,176,180, 184,190];

- *New methods of clinical and pharmaceutical analysis* for biologically important compounds [75,81,104,114,149,172,191,198,202,206,216,223,231,234], pharmaceuticals [63,149,196,202,209,230] or various food stuff [194,201,210,211, 217,218,221,222,233];

- *Applicability of carbon-paste based electrodes to study interactions of organic and biological macromolecules* (e.g. polymeric films [70,75,172] and DNA [134,166]), or even some microorganisms (namely, bacteria and yeasts [27,175, 181,228,306]).

With regard to all the research areas surveyed above, experimental work with CPEs and related configurations reflects yet other aspects that are worth of emphasising:

- *The use of CPEs and their modified forms as an education tool for students at the Department of Analytical Chemistry*: either in advanced laboratory trainings [128], or as a long line of various themes in diploma [237-295] and dissertation [296-309] theses.

- *Start of new collaborations with allied research groups at other universities or research institutes*. Concerning (i) *national institutions* and their electroanalytical staff, there are mainly two representatives: Department of Analytical Chemistry, Faculty of Natural Sciences at the Charles University in Prague [27,43,52,55,89,149] and Department of Molecular Electrochemistry group at the Heyrovsky Institute of Physical Chemistry in Prague [142,156,167,178,183]. Besides, there have also been and still are numerous minor co-operations — or better, exchanges of the individual skills — that have resulted in a few reports; among others, with Palacký University in Olomouc [105]; Kovohutě Příbram and Safina Vestec [65]; Institute of Mineral Resources, Kutná Hora [66], or even such examples spawning no regular publications but being beneficial anyway (e.g., Inorganic institute, ASCR, Řež u Prahy [61]; Biophysical Institute, Brno [267], Masaryk Institute for Water Management and the University of Ostrava, both in Silesian metropole [84,311]).

Thirty years of existence of electrochemistry and electroanalysis had also led to the establishment of a number of bilateral collaborations on the international level; here, it should be added — also thanks to political changes on the verge of the 1980s and 1990s that had led to open borders and the unprecedented students' mobility in-between the involved institutions. Such venues, where such mobility had been realised and the research with CPEs had played a significant role, were as follows (in chronological order): (i) School of Chemistry, Dublin City

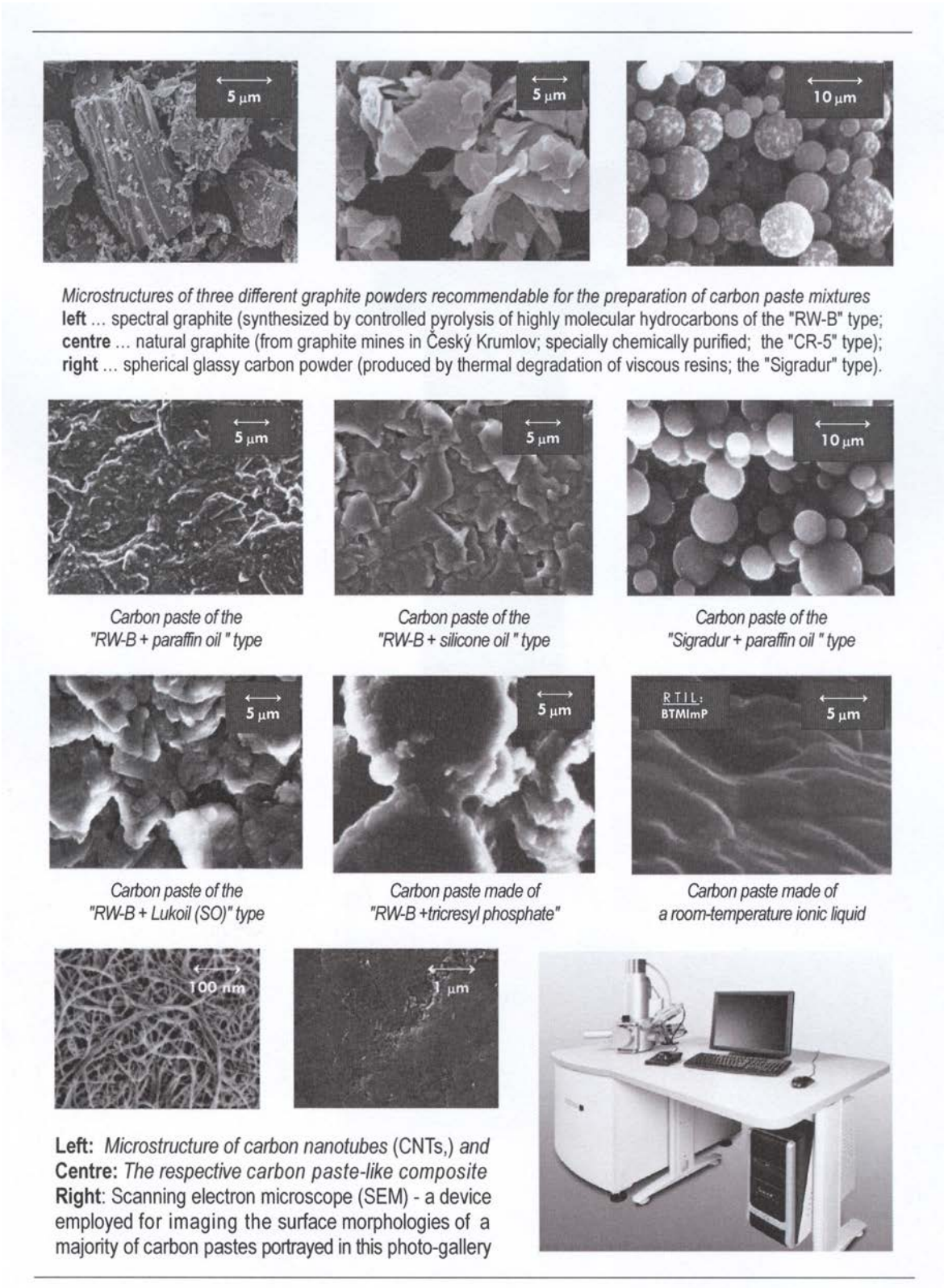


Fig. 4 Surface morphology of various carbon-paste based mixtures. Illustrative collage (The individual images withdrawn from [71,120,125,147,303,304] and the-first-author's archives; in most cases, being newly rearranged.)

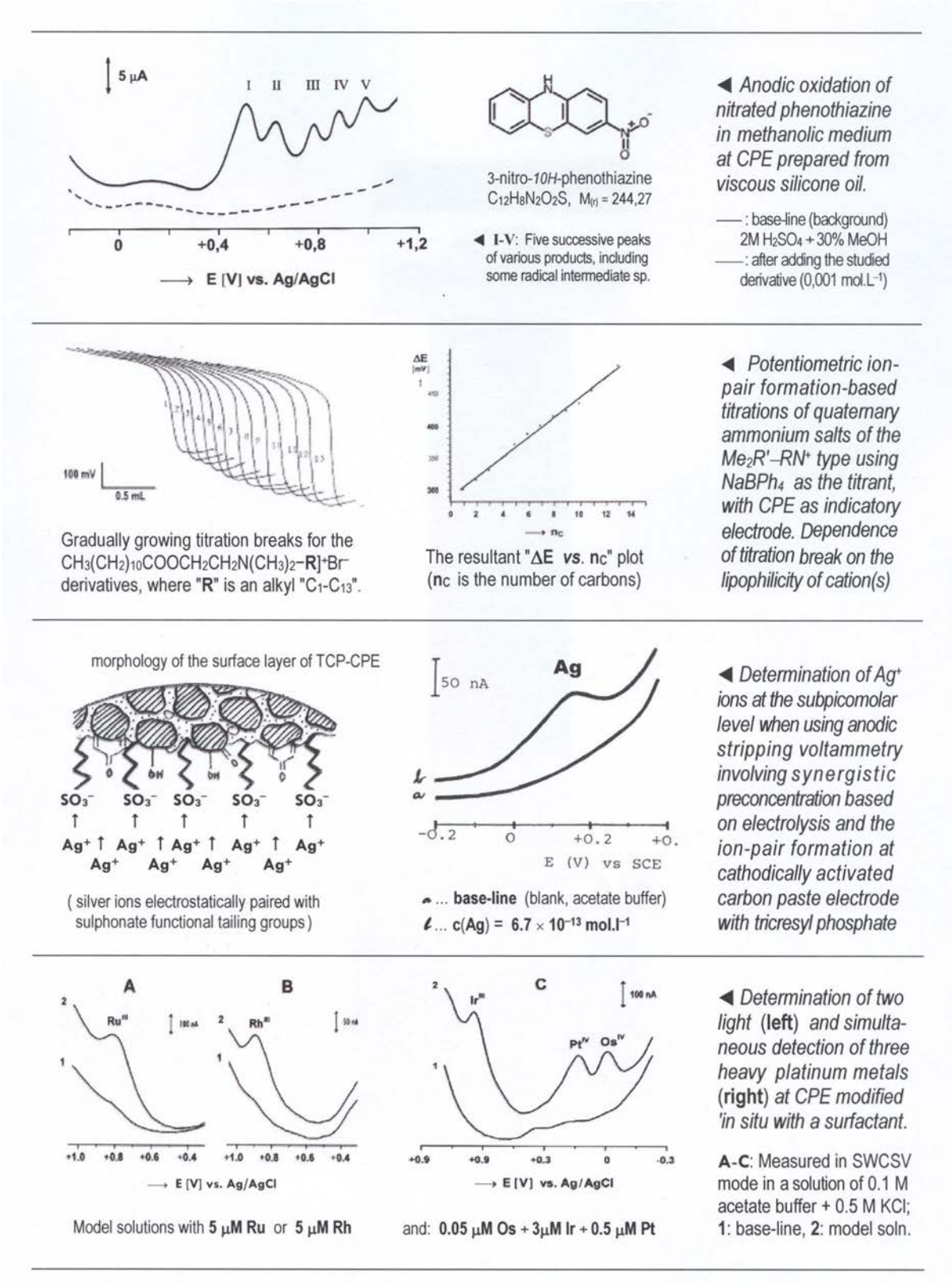


Fig. 5 Mosaic of some interesting measurements with carbon paste electrodes and related configurations (The individual images withdrawn from Refs [21,63,69,73,127,133,143] and eventually rearranged)

University (Ireland [61-63,70,75]); (ii) Institute of (Analytical) Chemistry, Karl-Franzens-University Graz (Austria, among others: Refs [68,73,77, 81,88,108,110, 156,172,200,216]); (iii) National Institute of Chemistry, Ljubljana (Slovenia, e.g. Refs [113,127,157,163,170,186]), (iv) Department of Building Materials, University of Mining and Metallurgy, Krakow (Poland e.g. Refs [98,108,110,112, 137,141]); (v) Department of Chemistry, Aristotle University of Thessaloniki (Greece, e.g. Refs [136,138,144,147,152,171,215]); (vi) Department of Chemistry, Norwegian University of Science and Technology, Trondheim (Norway [130,151, 154,159]); (vii) Department of Chemistry, University of Novi Sad (Serbia, e.g. Refs [158,174,184,207]).

Presentation activities about CPEs arisen from international collaboration represent more than a third of all the publications and hence, it is quite interesting to perform a little statistics within the respective databases featured by the following survey in Table I.

Table I Top-5 chart on publication activities of EAG UPa & cooperating institutions in abroad

| № | Institution (Abbreviation) Location (International code) | Heading scientists | Orig.p. + Conf.c.* | Review articles** | Total number |
|---|---|---|---|---|---|
| 1 | Karl-Franzens-University (KFU) Graz (AUT) | Kalcher K. | 27 | 15 | 42 |
| 2-3. | National Institute of Chemistry, Ljubljana (SLO) | Hočevar S.B., Ogorevc B. | 12 | 0 | 12 |
| 2-3. | Aristotle University (AUTH) Thessaloniki (GRE) | Economou E., Sotiropoulos S. | 12 | 0 | 12 |
| 4 | University of Novi Sad (UNS) Novi Sad (SRB) | Guzsvány V. | 10 | 1 | 11 |
| 5 | University of Mining & Metallurgy (AGH) Krakow (POL) | Bobrowski A. | 10 | 0 | 10 |

* Original papers and conference contributions in the full-text format plus selected presentations registered at WoS;

** Including one book and a series of chapters in monographs and special textbooks

● To complete the profile on the international cooperation within the EAG UPa, there is also a number of contributions by the visiting students and PhD aspirants, coming from Austria (e.g. Refs [79,114]), Poland [110,137], Lithuania [112,163, 208], Egypt (e.g. Refs [97,100,117,150]), Kuwait [119,122], Slovenia [127,145, 164], Serbia [176,184], Germany [165,212], Vietnam [175,209,306], and Iran [173,185,196,307]); when some contributions of these young scientists could already be gathered in Table I as the results of a wider collaboration.

## Some Contributions of the Primacy Character and Other Significant Achievements in the Field versus Some Unsuccessful Activities and Unrealised Ideas

Thirty years of intensive research with a long line of diverse applications across the electrochemistry and electroanalysis with carbon paste-based electrodes have given rise to a number of notable contributions into the field that can be remembered fondly and with certain pride. On the other hand, lengthy decades have also seen some dull moments and unfulfilled anticipations. From both sides, we can select:

- *Pioneering Studies and New Findings* … During a systematic characterisation of various carbon paste mixtures in the early 1990s, the research activities within EAG UPa have led to some outstanding results, presenting new facts and conclusions unknown at that time.

(i) *New Carbon Pastes with Liquid Organic Esters* [64,242]. Apparently, the first notable achievement was with CPEs containing organic esters as the alternate binding moiety instead of normally used paraffin and silicone oils. The most distinct representative, *tricresyl phosphate* (originally, liquid ion-exchanger for capillary chromatography) had manifested a very fine chemical activity and the respective TCP-CPE(s) could be utilised in a applications; see Refs [67,73,85, 104,162,174].

(ii) *Carbon Pastes More Resistant to Disintegration in Media with (Polar) Organic Solvents*. Practically in the same time, comparative studies with two types of standard carbon pastes, namely: paraffin-oil and silicone-oil based mixtures, had revealed a substantially better stability of the latter in aqueous solutions containing higher amounts of organic solvents tested (MeOH, EtOH, MeCN, DMFA and DMSO [61,243]). This new finding could then be utilised to prepare special carbon pastes from highly silicone fluids (with relative molecular weight *ca*. 10 000 g $mol^{-1}$) that had been stable and applicable in mixed media containing up to 50 % (v/v) MeOH [63].

(iii) *Metallic Film-Plated Carbon Paste Electrodes and Related Configurations* … The most significant achievement of EAG UPa into the electroanalysis with CPEs is associated with introduction and propagation of the carbon paste material as a support for metallic films and possible alternative to common solid electrodes employed for this purpose for a long time [40]. In this area, we have contributed several times — initially alone [66,237-239], later in collaboration with Middle European partners [67,74,98,110,145].

First, it was a mercury-film plated carbon paste electrode (MF-CPE [67]) whose properties in the role of a support had been found excellent despite the previous reports on unsuitability of carbon pastes for such purposes (see e.g. Ref. [40] and with Refs therein). The second type came shortly after as AuF-CPE,

followed by its fine applications in practical analysis [99,108,247]. Apparently, the most important step in the area of MeF-CPEs was a collective work on bismuth-film plated carbon paste electrodes (BiF-CPEs [110]), meaning a real entry of EAG UPa and their partners into the newly born discipline of electroanalysis with bismuth-based electrodes (see e.g. Ref. [319] and Refs therein). (Note: Both BiEs and BiFEs are now considered as a textbook case of (eco)electroanalytical measurements within massively popularised green analytical chemistry [420,421].) Regarding the BiF-CPE, they offer interesting employments; also, thanks to our own activities (see e.g. Refs [107,117,152,212]). Finally, there is also antimony-film plated configuration (SbF-CPE [319]) and hybrid BiF/SbF-CPE [197]; the former again with some nice applications [164,189,205,291].

As a certain complement to the above described configurations, also bismuth- or antimony-powder modified carbon pastes, Bi-CPE [127] and Sb-CPE [170] should not be omitted; also due to the fact that have given rise to other related electrodes, completing — along with HgO-CPE [98], $Bi_2O_3$-CPE and $Sb_2O_3$-CPE [112], $NH_4BiF_4$-CPE [163], SbOCl-CPE [168], or $BiF_3$-CPE [199] and $SbF_3$-CPE [179] — the large family of metal-modified CPEs introduced for the first time by the EAG UPa with cooperating partners.

(iv) *Microscopic Studies on the Surface Morphology of Carbon Pastes and Related Structures* ([71,120,125,147,243,303,304] and Fig. 4) … By following some pioneering studies carried out in the late 1980s in U.S.A. (see Ref. [71] and Refs therein), the EAG UPa in cooperation with the Joint Laboratory of Solid State Chemistry at the UPa and some permanent partners from abroad have also contributed to define the basic types of carbon paste with respect to their microstructure. Of principal importance was already the initial study [71,243], concerning for the first time the carbon paste mixtures made of glassy carbon microspheres. The unique structure of this special material had principally helped to feature the binding role of the liquid moiety in carbon pastes. The remaining microscopic studies were focused mainly on specific (micro)structures of mercury [243], bismuth [120,125,147, 303], and antimony [304]. Although the respective microlayers had usually been deposited onto carbon paste substrates and resulted also in some principal findings, their detailed commentary is already beyond the scope of this survey.

● *Other Notable Results and Achievements* … The above-highlighted contributions can be completed yet with some other items, characterising the research in the field within the EAG UPa and, again, typically as the actual output of bilateral collaboration(s). Among the activities worthy of surveying here, one can mention the following topics:

(v) *CPEs vs. Surfactants*, where quite extensive research had covered the use of surface-active substances as titrants and/or titrated compounds [38,69,213,220, 227,229,235,244,297]) or *in-situ* modifiers for highly selective determinations in

the electrochemical stripping analysis [90,92,121,133,143,213,252,269].

(vi) *CPEs vs. Stripping Potentiometry*, when EAG UPa belonged to one of the first teams having started to use CPEs in both PSA and CCSA systematically [86,100,106,107,113,161, 165,298]. Then, knowledge obtained with the new technique in combination with previous experience with CPEs could be exploited in some fine applications [95,103,108,111].

(vii) *Carbon Pastes as Biosensors*, when the EAG UPa joined forces with our closest partner at the KFUG, responding to an unprecedented boom in biosensorics during the1990s [10-12]. This research orientation has soon resulted in two directions: (vii-a) *propagation of a concept* utilising carbon pastes as transient elements for development and testing of carbon inks, representing a very similar material and basis of screen-printed (carbon) electrodes and (bio)sensors (SPEs; [23,115,128,299]); (vii-b) a series of reports on *CP-based (bio)sensors with* $MnO_2$ [77,78,114,300] acting as a mediator and cheaper alternative to platinum metal oxides, within which the popularised concept could also be demonstrated [45,79].

(viii) *Traditional Carbon Pastes vs. New Mixtures (from Alternate Components),* where the already discussed CPEs from glassy carbon microspheres — see par. (v) and Ref. [71] — had coincidentally represented one of the very first examples of the-so-called *new carbon pastes* whose era came a half a decade afterwards [71]. A few typical representatives of such new carbon pastes reported recently [167,178,183] were, in fact, "obligatory" contributions of the EAG UPa into the already established area of these carbon paste-like composites.

(ix) *Electroanalysis with CPEs in Conference Presentations*. In the overall survey of all the activities (see above), also oral contributions and posters at conferences and seminars were included, remembering — in a chronological order — most of such items that have been presented at various scientific events [312-412]. Among the selected items, some notable performances can be reminded. First, it is a (i) *very first presentation in abroad* (at a local conference in Scotland as a poster [312]; a (ii) *debut at a home event* (*via* short oral speech [313] and during traditional Czechoslovak / Czech meeting series organised under the moniker *'Modern Electrochemical Methods'* (*MEM* [419]) and, since the mid-2000s receiving the status of International Conference (see Refs [367] and [374]) and soon after entering into the WoS databases [385,386,390-392,396-399,401,404-407,409-412]. A trio of such memorable events can be completed by (iii) the *first plenary lecture* upon invitation [315].

Among conference presentations, where one or even more members of the EAG UPa had participated, regular contributions on large international meetings have prevailed. From the attendances at such events one can emphasise the following items (where the first numbers of each sequence with citations refer to the respective debut: the ESEAC series ([314] + [319,331,342,350,363,376,377]),

ISE, ISEC, and ISEAC meetings ([321 & 327 & 394] + [322, 339,345,371, 373,389,400,403]), or IMA conferences ([346] + [358]). Second, there are various regional conferences and seminars, emphasising the role of Central Europe ([323] + [359,408]) or Aegean [387], Baltic [325], Iberic [395], and Eurasian ([388] + [393]) regions. Some other regional or local meetings had been established on the platform(s) of join scientific programmes and organised regularly, such as the YISAC series (e.g. Refs [332, 355,364,369,378,383]) or as a special occasion to celebrate significant jubilees in electrochemistry and/or electroanalysis (e.g. MEM '99 [326] or Heyrovsky Lectures [379]). Third, the research work of the EAG UPa could be presented at some distant or even exotic venues, such as those in USA [339,343], Canada [371], Mexico [380], Brazil [345,373], China [327,328], South Korea [321], Japan [322], Turkey [393], Jordan [388], Syria [366], India [334,372, 394,403], Egypt [315,335,347], and South Africa [400]. Finally, there is also one really curious "contribution" — a submission of never presented lecture [340].

Last but not least, there are also some scientific events that had been established within the activities of the EAG UPa and where the contributions about CPEs in all possible forms were being appearing regularly. First, it is the already-mentioned (i) YISAC series started in 2000 and still existing. Then, it was a similar line of meetings known as (ii) 'Sensing in Electroanalysis', having spawned a set of full-text proceedings of the same name, with a premiere in 2005 (see e.g. Ref. [129]) and hitherto last issue in 2014 [216]. Finally, there is a series of seminars entitled (iii) 'Monitoring of Environmental Pollutants' (and organised mainly for Czech participants), founded in 1999 [41] and running up until now [234].

(x) *A Two-Decade Monopoly in Reviewing the Field (from 1993 up to 2013)*. Apart from a hardly accessible article from 1998 ([11]; in addition, published in Chinese), there was no-one else for lengthy twenty years who would review the electrochemistry and electroanalysis with CPEs in its entirety — except the EAG UPa and collaborating partners, preparing altogether twenty nine reviews [31-59] plus nine book-chapters [22-30] being, in fact, some kind of reviews, too. This hegemony was ended by a Spanish group in 2013 (see Ref. [18]).

Retrospectively, such publication potency is also one of remarkable outputs; moreover, objectively measurable via citation profiles of all principal reviews at Web of Science [20][4]: Ref. [36]: 485×, [43]: 260×, [50]: 271×, [51]: 115×, [52]: 103× ; each being filtrated from cross self-citations (neither author nor other co-authors may cite himself / themselves).

- Of course, during very diverse investigations within the field, there were also

[4] In contrast to small statistics given at the beginning of this text, these numbers are actual and thus, for some items, they may also illustrate *ca.* two-year growth in the corresponding citation indexes

some unsuccessful efforts and unrealised ideas. Among such disappointments, one can reveal:

(α) *Experiments with Carbon Paste Mixtures Prepared from Halogenated Hydrocarbons* [61]. In contrast to repeated recommendations by Adams [1-3], our mixtures of such composition had exhibited unstable consistency and unfavourable properties in overall. In addition, the examined substances had been found very aggressive against the Silon® plastic material used for machining the CPE-holders at the workshops of UPa [59,129].

(β) *Studies on Applicability of CPEs for Determination of Plant Hormones* [105]. The actual tests had been carried out with 6-benzylaminopurine and despite certain expectations, the results and observations obtained gave no chance to develop the desired method for trace determination of this compound and, apparently, of related derivatives as well.

(γ) *The Use of $NaBiO_3$ as a Precursor for Preparation of New Bismuth-Modified CPE* [61]. During a rapid progress of (eco)electroanalysis with bismuth-modified carbon paste electrodes [420,421], this new area saw a wide palette of various configurations; however, none of them had employed a compound of pentavalent bismuth. Because it was believed that its abrupt electrode reduction, $Bi^V + 5\ e^- \rightarrow Bi^0$, might proceed in a particular pathway yielding bismuth layers with unusual properties, the most common compound of $Bi^V$, sodium bismuthate, was tested for this purpose. However, its high reactivity and overall instability hindered the use of this chemical as a precursor or, eventually, as an *in-situ* modifier.

(δ) *Electrically-Heated Corpus with Carbon Paste* [160,276]. By further adaptation of previously developed planar CP-holders [126,153], an assembly for measurements at higher temperatures was constructed. Because the subsequent experiments revealed problems with its functioning, further tests were abandoned and originally plans unrealised.

(ε) *Carbon Pastes from Ionic Liquids* [178]. Similarly, carbon ionic liquid electrodes (CILEs), massively popularised in the 2000s, had not convinced of their benefits and the just-initiated research on these new types of CPEs was stopped.

## Conclusion

At the University of Pardubice, the electrochemistry and electroanalysis with carbon-paste based electrodes, celebrating this year a round jubilee, represents undoubtedly one of the most distinct signs characterising the scientific activities at the Department of Analytical Chemistry during its whole existence.

In the previous sections, it was documented clearly that the activities of the EAG UPa within the field have enriched the scientific and educational life at the

department when taking into account the overall evaluation. And one can even conclude that all the results and publication outputs have also contributed to the good name and constantly high reputation of the Czech electrochemistry in the widest point of view.

With respect to the field itself, it is not yet exhausted — not at all — and despite its almost classical status, it is still very vital and offering a full compatibility with the latest trends. This can be documented in all last reviews [18,30,51-55], as well as in newest electroanalytical literature (see e.g. Ref. [422]). At the end, we can add a very interesting info connecting the prenatal period of the field with the present. It concerns the Adams's original idea on the dropping carbon electrode (DCE) operated through a suspense of graphite powder in readily flowing medium [3,50]. Although the devised prototype had not been successful, its importance is principal as the respective experiments led to the discovery of a new electrode material of soft consistency — the carbon paste [1,2]. (Recently, it was a great honour for us that the scientist who, as a young lad, had been involved in the pioneering experiments with DCE and carbon paste prototypes agreed to write a foreword for our book on CPEs [21,399].) The concept of DCE, regularly reminded in our retrospective texts [36,43,49,50], was resurrected a few year ago — in elegant studies of Japanese scientists headed by Tatsumi who have introduced two new models of their own dropping carbon fluid electrode (DCFE, see Refs [423-425]).

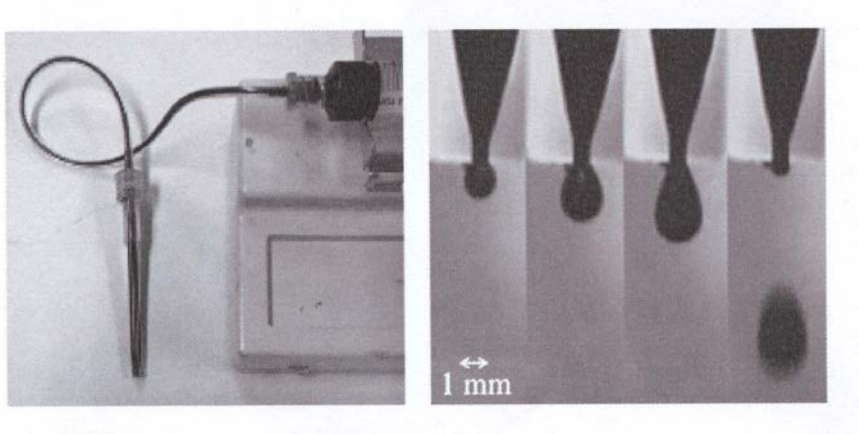


Fig. 6 Dropping carbon fluid electrode; left: overall design; right detail (of the tip and rising drop). Taken from Ref. [424], reproduced with permission

The first construction, described in first two reports (Refs [423,424] and Fig. 6), employed a ternary mixture with binder and an ionic liquid, the second [425] was a simpler configuration based on two-component fluid from a special paraffin oil. Regardless of the fact that both variants have required rather atypical conditions for proper functioning, they represent the real fulfilment of "old" Adams's dream. And, in vast archives of the original reports on CPEs and related configurations, one could trace up other similar challenges…

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