

The Two and Three Electrodes Systems Topology Optimization of Electrochemical Sensors

Ulugmurodov Elshod Abdusalim ugli

Student Karshi Institute of Irrigation and Agrotechnics at the National Research University “Tashkent
Institute of Irrigation and agricultural Mechanization Engineers”

ulugmurodovelshod2001@mail.ru

Annotation. Miniature electrochemical sensors can be produced using thick film technology. Fabrication technology optimization of thick-film sensors, the adjustment of optimal technological properties and especially the optimal properties of thick-film electrode materials and electrode topology are main problems of sensor design. The electrodes topology design represents one of very important parts. It is very complicated, because of many contradictory requests that must be solved. Some aspects about this problem in electrochemical sensor area are discussed in this paper.

Key words: Thick-film sensor, electrochemical sensor, electrode.

1. Introduction

Thick-film sensors are used in various human activities (food industry, agricultural industry, automobile industry, medicine, etc.). The main advantage of Thick film technology (TFT) is low price and small scale batch production [1]. At present time the TFT is used as a tool of preparation of very small electronic details by SMT [2], high reliability applications and nonconventional applications [3], especially sensors. TFT sensors are often simple, cheap, sufficiently sensitive and accurate, with good mechanical and electrical properties. In the other hand, the application of thick film technology to chemical sensors and biosensors can be complicated [4], [5].

Processes on electrodes of TFT electrochemical sensors are more complicated than in classical electrochemistry. Electrochemical methods are extremely sensitive, which means that even small impurity, which has negligible influence in classical sense, can influence the final sensitivity and other properties significantly. The materials of TFT electrodes are non-homogeneous in a microscopic point of view (composition, structure, ...), they do not have well defined area, roughness of surface, etc.

Output current signal of thick-film electrochemical sensor depends on:

- materials (working electrode, reference electrode, auxiliary electrode, substrate, dielectric layer, conductive layer, ...),
- technological parameters of production (electrode firing temperature, time of electrode firing, ...),
- packaging,
- transport,
- storage,
- influences during measurement (measuring system – stirred × no stirred, flow of sample over working electrode, temperature, light, ...),
- sensor design – size of working electrode area S_w , rate of working electrode area to reference electrode area ($S_r:S_w$), uniform current density between working and reference electrode in a two-electrode system or between working and auxiliary electrode in a three-electrode system.

2. Parameters In Electrode System Topology Design

The electrode system topology is very important problem in thick-film sensor design. In classical electrochemistry, the working electrode area A_{WE} is often defined by window in body electrode construction or by area of rod from various materials. The reference electrode area A_{RE} is defined from small window in body electrode construction, e.g. glass Ag/AgCl electrode for three-electrode system to large area of mercury reference electrodes in polarography for two-electrode system [6], [7]. The rate $S_r:S_w$ is often 50:1 in classical polarography. The large reference electrode area is better for potential stability in two-electrode system [8]. The three-electrode system increases the potential stability and allows the reference electrode size reduction. The three-electrode system is often made as 3-D system.

In thick-film technology, the electrode system is made as planar system. Therefore the thick-film electrode topology design is limited. Main problem are uniform current density, substrate dimensions, screen-printing resolution, paste parameters, etc.

The small size of sensor's substrate and fact, that all electrodes must be placed on the substrate, limit the working electrode size and the rate $S_w:S_r$. Therefore the main aims of the topology design are working electrode area ($I_{out} = f(S_w)$) and rate $S_w:S_r$ (for potential stability) increasing. This rate is often 1:10 and less. Afterwards when the three-electrode system is made on TFT electrochemical sensor, the auxiliary electrode is often connected to the reference electrode increasing the S_r area in case of two electrode system measurement. The design of such type of sensor is very big problem due to below mentioned requirements:

- high size of working electrode area,
- high size of reference electrode area and high rate $(S_r+S_a):S_w$ in the two-electrode system with auxiliary electrode connected, or high rate $S_r:S_w$ in the two-electrode system without auxiliary electrode connected,
- high size of auxiliary electrode area in the three-electrode system, the reference electrode size may be small,
- uniform current density between working and reference electrodes in the two- electrode systems or between working and auxiliary electrodes in the three- electrode systems,
- material of reference and auxiliary electrode must be same in the two-electrode system.

3. Sensor Topology Design

Various topology designs of electrode system for electrochemical sensor were designed in this work. Some designs are shown in the fig. 1. Geometrical areas of electrode surface for working (S_w), reference (S_r) and auxiliary (S_a) electrode are calculated on this figure too. The whole usable area of electrode part is $S = 5,2 \text{ mm} \times 5,2 \text{ mm} = 27,04 \text{ mm}^2$.

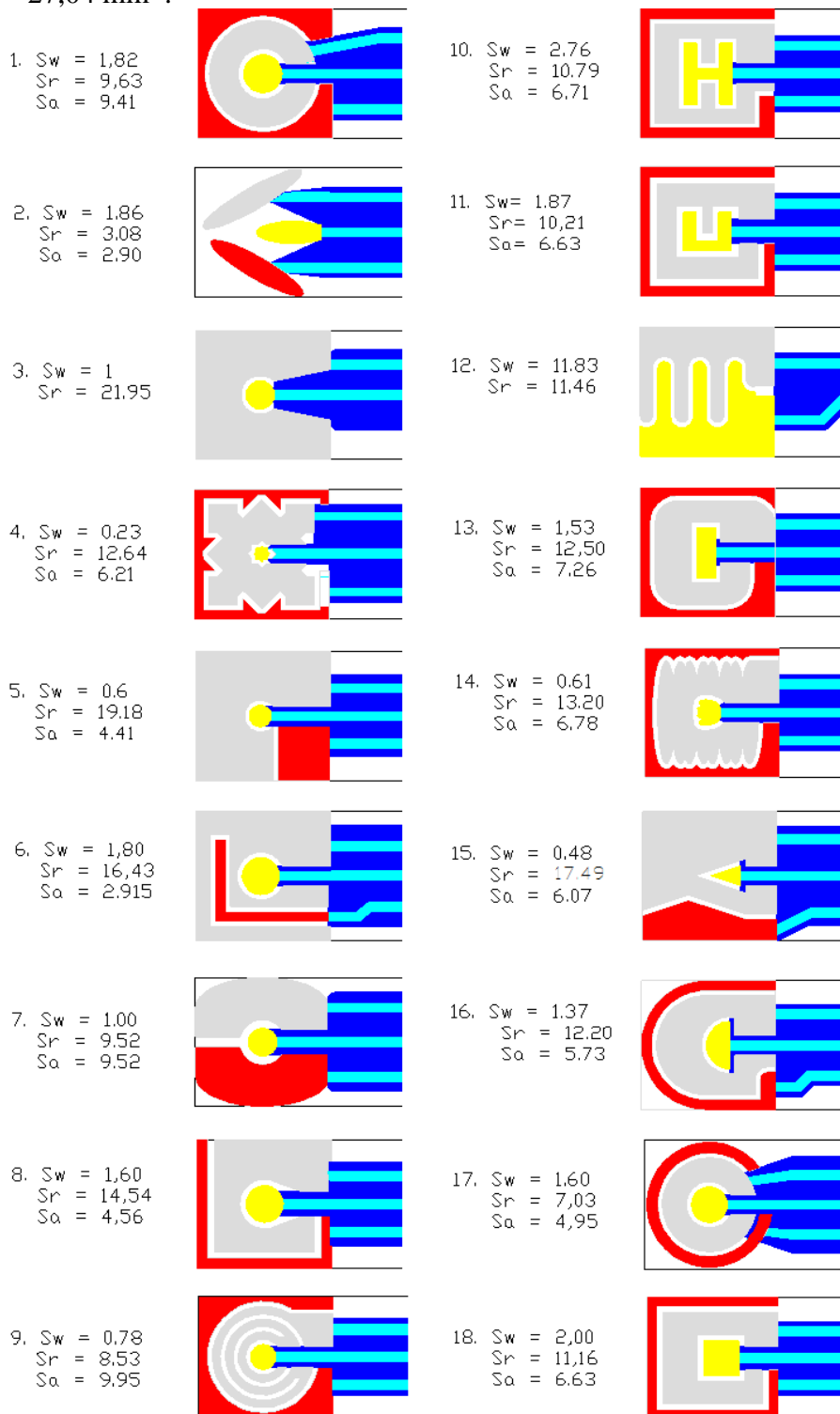


Fig. 1: The topology designs of electrode system for electrochemical sensor.

The table 1 shows the geometrical areas of electrode surface for working (S_w), reference (S_r) and auxiliary (S_a) electrode, rate S_r/S_w , $(S_r+S_a)/S_w$, S_a/S_w and information about electrode part filling of electrochemical sensor by working (S_w), reference (S_r) and auxiliary (S_a) electrode areas (rate S_w/S , S_r/S , S_a/S).

Table 1: The electrode area comparison of electrochemical sensor designs.

No.	S_w [mm ²]	S_r/S_w	$(S_r+S_a)/S_w$	S_a/S_w	S_r [mm ²]	S_a [mm ²]	S_w/S [%]	S_r/S [%]	S_a/S [%]
1	1.8	5.35	10.58	5.23	9.63	9.41	6.66	35.63	34,81
2	1,86	1,66	3,22	1,56	3,08	2,9	6,88	11,41	10,73
3	1	21,95	21,95		21,95		3,7	81,21	
4	0,24	53,57	79,91	26,34	12,64	6,22	0,87	46,77	23
5	0,6	31,97	39,33	7,36	19,18	4,42	2,22	70,97	16,33
6	1,8	9,13	10,75	1,62	16,43	2,92	6,66	60,78	10,78
7	1	9,52	19,04	9,52	9,52	9,52	3,7	35,23	35,23
8	1,6	9,09	11,94	2,85	14,54	4,56	5,92	53,79	16,87
9	0,79	10,86	23,53	12,67	8,53	9,95	2,91	31,56	36,82
10	2,76	3,91	6,34	2,43	10,79	6,71	10,21	39,92	24,82
11	1,87	5,46	9	3,54	10,21	6,63	6,92	37,77	24,53
12	11,83	0,97	0,97		11,47		43,77	42,42	
13	1,54	8,13	12,86	4,72	12,5	7,26	5,69	46,24	26,87
14	0,61	21,69	32,82	11,14	13,2	6,78	2,25	48,83	25,08
15	0,48	36,45	49,1	12,64	17,5	6,07	1,78	64,74	22,45
16	1,37	8,94	13,14	4,2	12,2	5,74	5,05	45,14	21,22
17	1,6	4,39	7,49	3,09	7,03	4,95	5,92	26,01	18,31
18	2	5,58	8,9	3,32	11,16	6,63	7,4	41,29	24,53

The illustrations of designs, which are not optimal, are shown in table 2.

Table 2: Errors in sensor design.

No.	Sensor topology	Error
2		The whole area is not used sufficiently.
4		The working electrode is small. The electrode shape is complicated (layers' register is bad).

As the main parameter from selection was chosen the size of working electrode area, second parameter was high rate S_r/S_w and last parameter was uniform current density between working and reference electrodes. First and second parameters are dependent on electrode area and third parameter is dependent on shape of electrode. The sensor No. 8 was chosen as the best design. The sensor has large working electrode area (1,6 mm²), high rate S_r/S_w (9,09) and circular shape of working electrode, which is best for uniform current density in this planar system.

4. Conclusions

The electrode system topology is important problem in the thick-film sensor design. Design of the three-electrode sensor which is possible to be connected to the two-electrode measuring system is the problem. The design of this type of sensor was made in this work. The sensor has working electrode area

size $S_w = 1,6 \text{ mm}^2$ and circular shape of working electrode. The sensor has large working electrode, which is needed to obtain good and high output current signal. The circular shape of working electrode is best for uniform current density in this planar system and good rate $S_r/S_w = 9,09$ and $(S_r+S_a)/S_w = 11,94$ is needed to obtain good potential stability.

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