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STRUCTURAL CHANGES IN C-Pd THIN FILMS DUE TO TEMPERATURE MODIFICATION IN CVD PROCESS

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INTRODUCTION

Nanostructures based on palladium nanocrystals and nanoporous carbon matrix are promising materials for hydrogen gas sensor applications. One of the factors which decides about using such materials in hydrogen detection is presence of Pd nanograins which absorb hydrogen. Well-developed specific surface area of carbonaceous matrix could also play a positive role in hydrogen adsorption. In literature one can find many examples of the use of nano-Pd structures (nanowires, nanoparticles, thin films) as hydrogen sensing elements because of their ability to adsorb/absorb large amounts of H_2 and to form Pd hydride (PdH_x) [1-3] .

Here we present results of structural changes studies of C-Pd films obtained by PVD/CVD methods (Physical Vapor Deposition/Chemical Vapor Deposition). These changes result from used different temperatures in CVD process. SEM (Scanning Electron Microscopy) investigations prove that the temperature of CVD process affects morphology, topography and microstructure of CVD films.

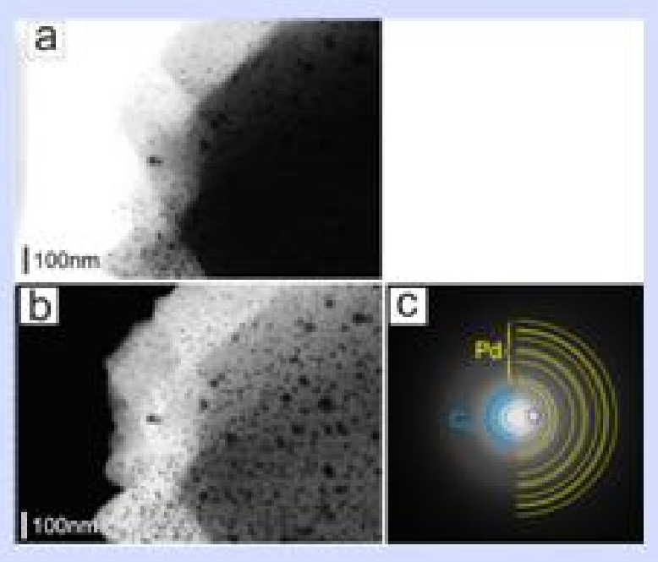
SYNTHESIS METHOD

For obtaining C-Pd films sensitive to hydrogen gas we apply two processes: 1) Physical Vapour Deposition as the first stage to form the initial nanocomposite C-Pd films and 2) Chemical Vapour Deposition as the second stage to modify PVD films and to prepare porous structure as well as to extract palladium nanograins on the films' surface. Such changes in the structure of initially obtained PVD film could cause the higher sensitivity of films to hydrogen presence in the ambient atmosphere.

Fullerene C_{60} and palladium acetate PdC_2O_4 are precursors of the initial nanocomposite films in PVD process. Both compounds are evaporated at duration time *t* from two separated sources (C_{60} and PdC_2O_4), at the distance *d* between sources and substrates.

In Table 1 parameters of PVD process are listed.

PVD film



TEM imaging P.Dłużewski IP PAS

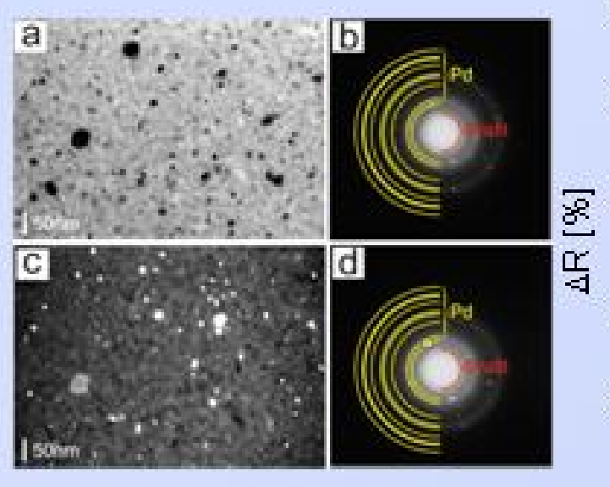
PVD process parameters			
I_{C60} [A]	I_{Pd} [A]	*d* [mm]	*t* [min]
1,9	1,3	54	10

Next PVD films were modified in quartz reactor by CVD method using xylene C_8H_{10} as an additional source of carbon in transformation process at different temperature.

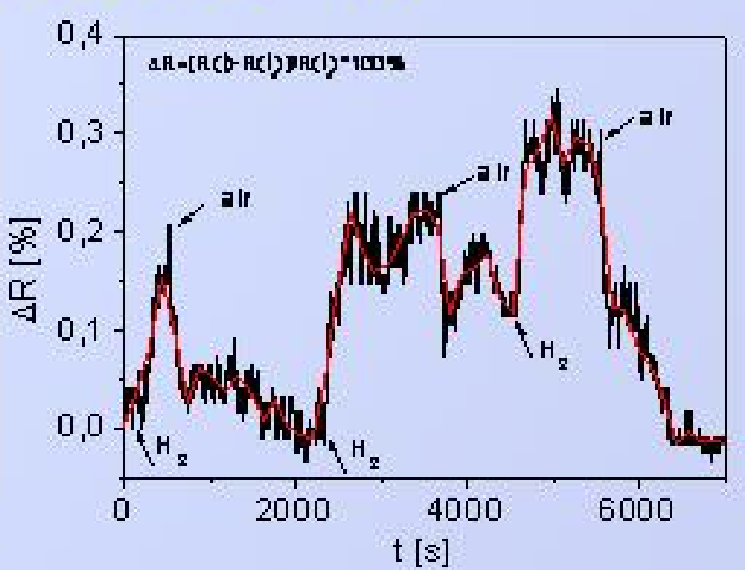
In Table 2 parameters of CVD process are shown.

No process	CVD process			
	T [°C]	C_8H_{10} feed rate [ml/min]	*t* [min]	Ar flow [l/h]
a	180/500	0,1/1	30	40
b	180/550	0,1/1	30	40
c	180/600	0,1/1	30	40
d	180/650	0,1/1	30	40
e	180/700	0,1/1	30	40
f	180/750	0,1/1	30	40

CVD film at 650 °C

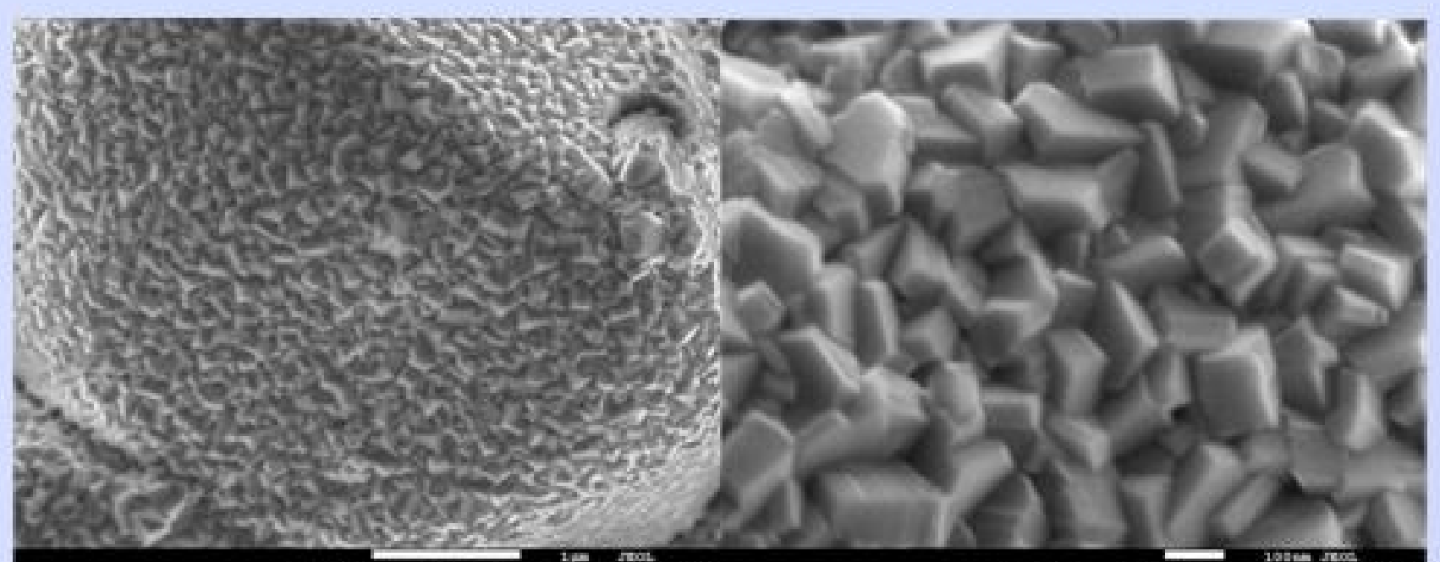


TEM imaging P.Dłużewski IP PAS



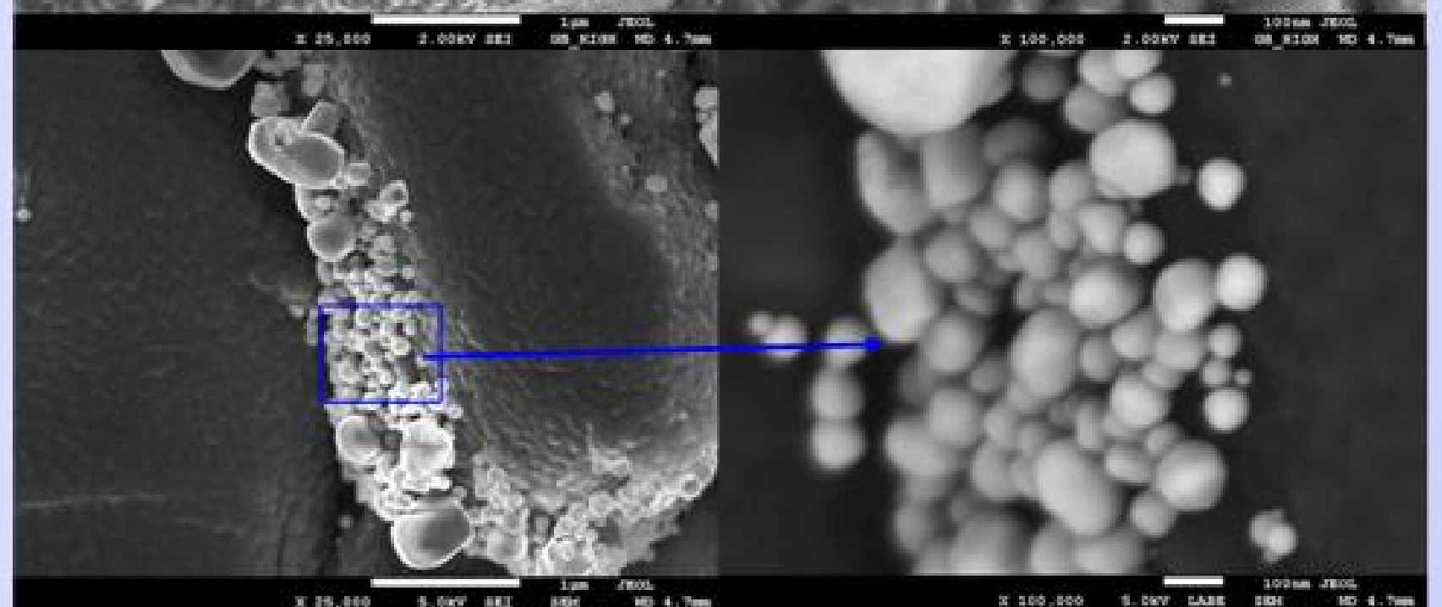
Response of CVD film (at 650 °C) to the presence of 1% H_2

SEM CHARACTERISATION



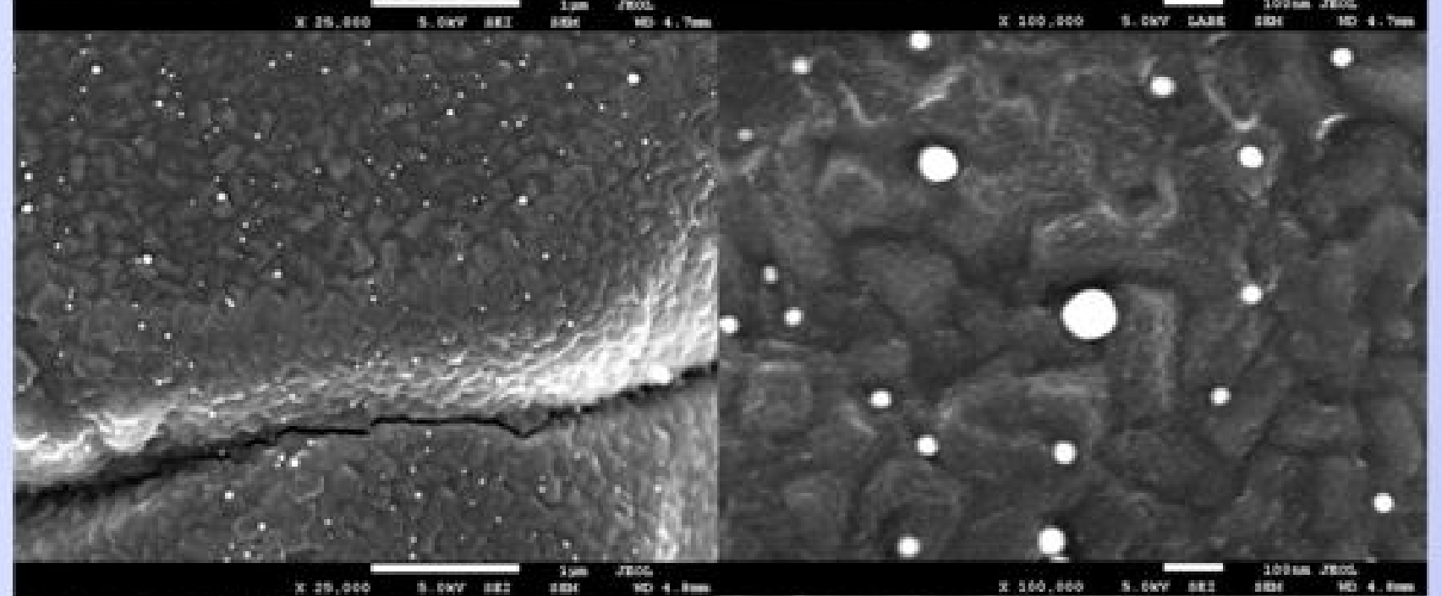
film PVD

densely packed carbonaceous grains with the size ~100-200 nm, Pd nanograins are not visible in SEM images



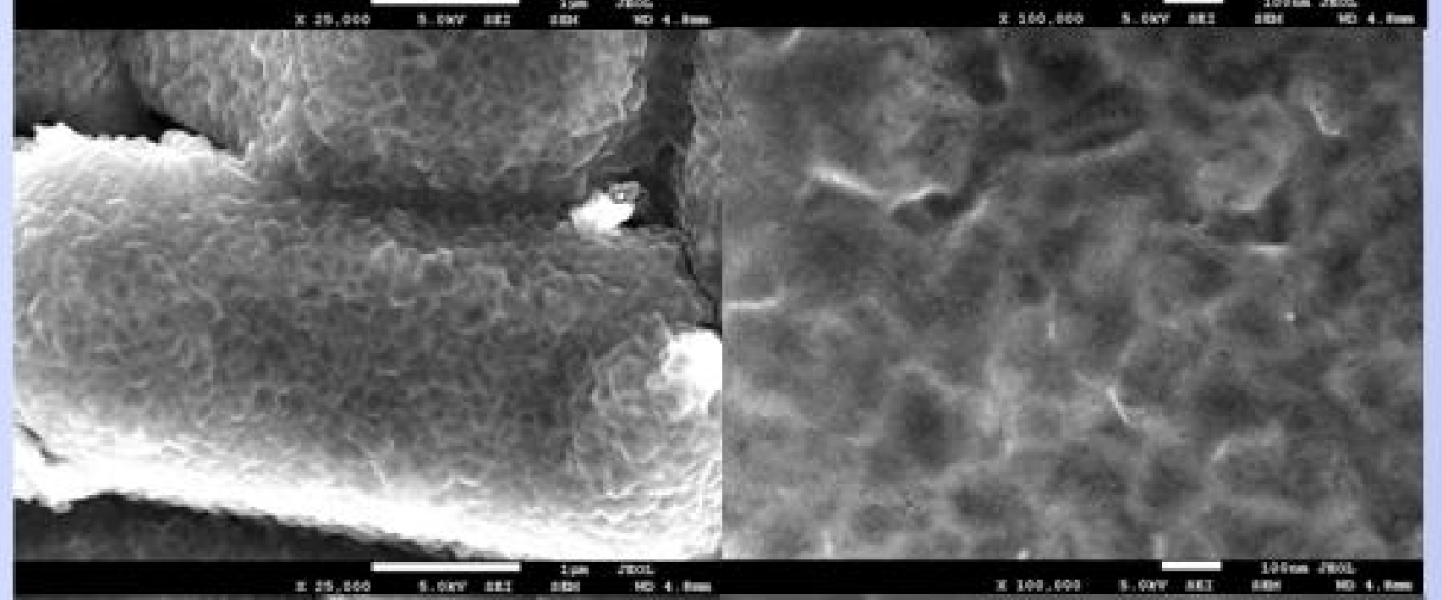
film CVD (a) – 500 °C

matrix C are little changed, the large Pd nanocrystals are collected on the substrate grain boundaries, the small Pd nanograins are dispersed below the film surface



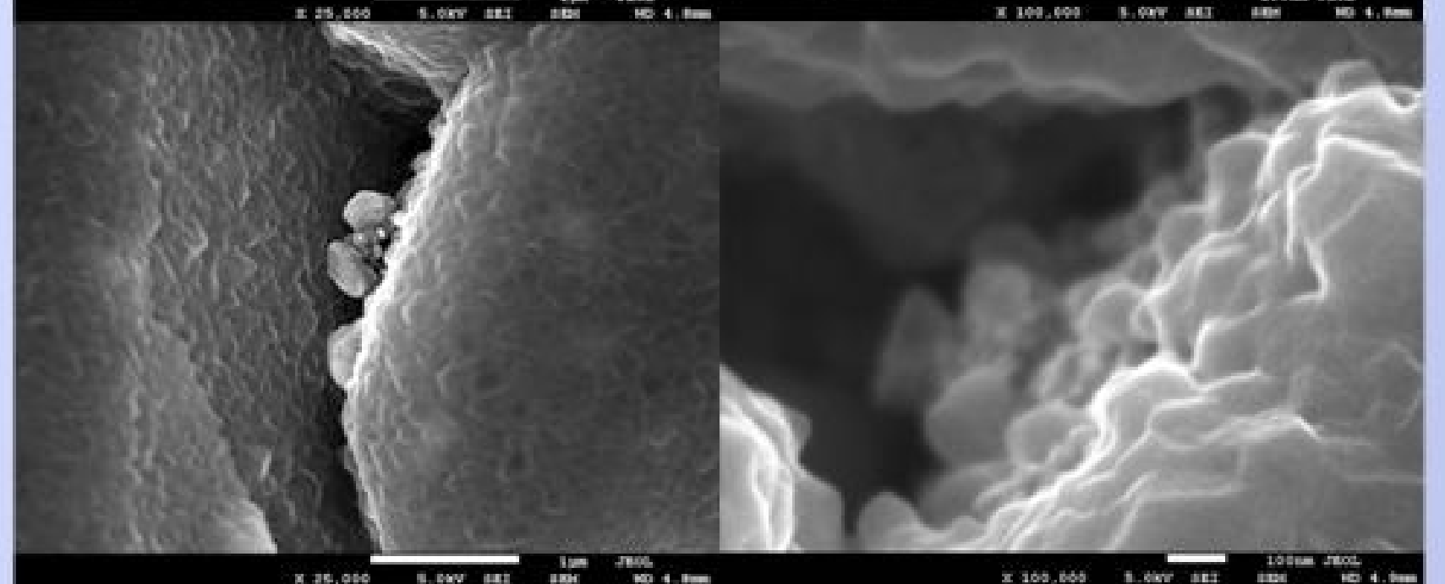
film CVD (b) – 550 °C

sharp-edges of C grains are fuzzy, a few Pd grains are visible on the film surface



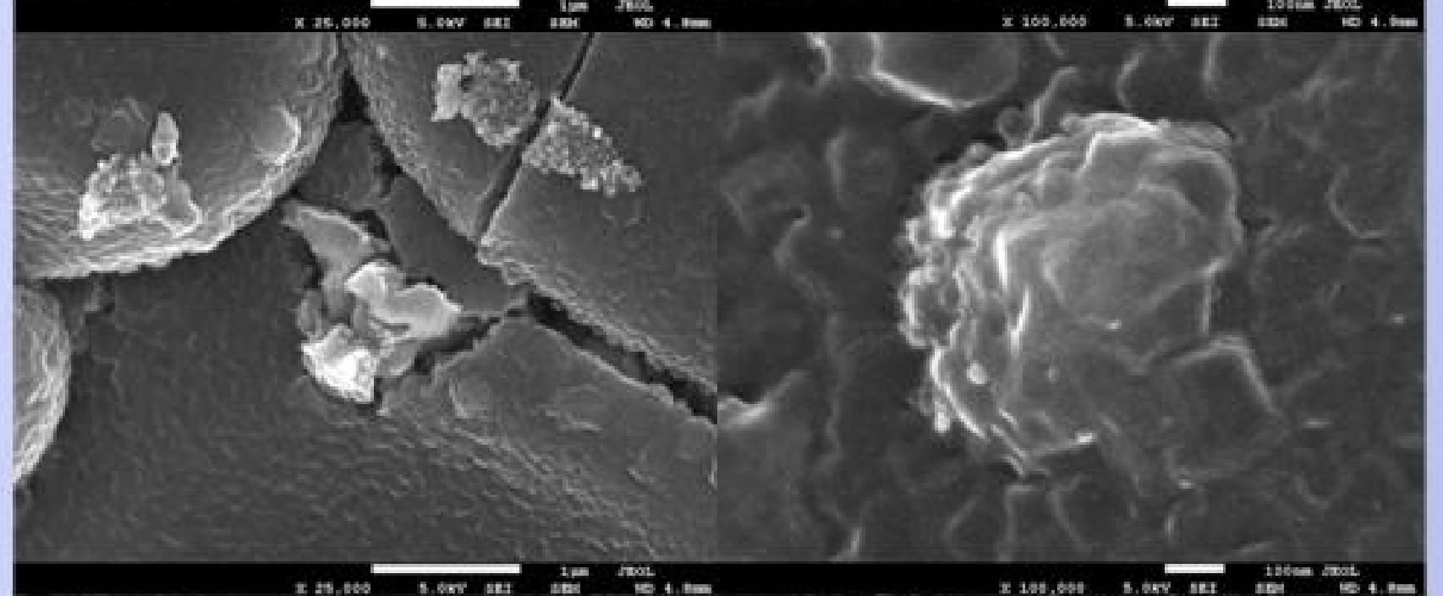
film CVD (c) – 600 °C

matrix C becomes downy, Pd nanograins are dispersed below the film surface



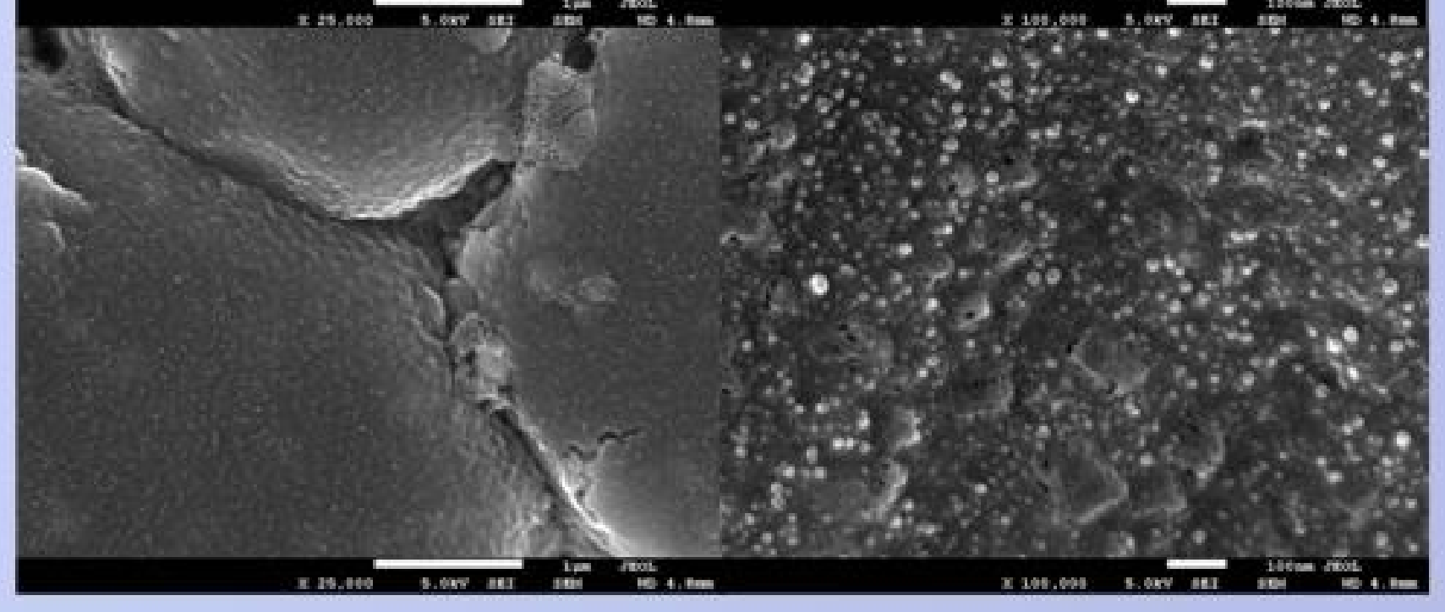
film CVD (d) – 650 °C

matrix C are more porous, Pd nanograins visible just below films surface



film CVD (e) – 700 °C

more Pd nanocrystals are presented on substrate grain boundaries, matrix C similar to film modify at 650 °C



film CVD (f) – 750 °C

edges of C grains are completely fuzzy, matrix becomes porous like foam, numerous Pd nanograins with the size of 5-20 nm are observed on film surface

magnification

25 000 100 000

RESULTS

- ➢ CVD process temperature affects structure and morphology of modified films in respect both of Pd nanograins and carbon matrix
- ➢ At low temperature (500-550 °C) the large Pd nanocrystals (100-300 nm) are collected at substrate grain boundaries, the small Pd nanograins are dispersed on the films' surface
- ➢ At intermediate temperature (600-650 °C) Pd grains are not visible on the films' surface but carbon matrix are transformed into porous structure
- ➢ At high temperature (700-750 °C) on films' surface very small Pd grains (5-20 nm) are observed and carbon matrix is still in the structure -like foam

CONCLUSION

The CVD film modified at 650 °C is sensitive to hydrogen presence, its resistance increases in hydrogen (1%) whereas in air its resistance decreases

REFERENCES

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