

DETERMINATION OF HYDROGEN - ADSORPTION CHARACTERISTICS OF FULLERITE C₆₀

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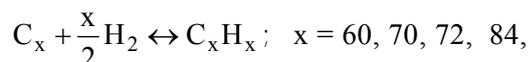
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Introduction

The perspective of hydrogen adsorption for new materials theoretically looks as optimistic enough. In the case of each carbon atom joins one hydrogen atom (it's likely enough) there is a possibility to produce these materials based sorption matrix which could accumulate to 7,7% (mas.) of hydrogen.

This value satisfies and exceeds all demands to this class materials of as USA Ministry of Energetics and as other international agencies.

Therefore, finding the way of complete and reversible carrying out of the reaction



could use these materials as the systems of hydrogen accumulation and storage in different fields of technique and technologies, if one takes into account the peculiarities of their structure and properties. It is known, that absence of such class materials suppresses wide application of hydrogen as fuel and energy carrier.

Results and Discussion

As it was denoted by authors [1], hydrofullerite of C₆₀H₂₋₁₈ composition was produced by direct solid-phase hydrogenation of fullerene with catalyst absence. Hydrogenation of fullerite was carried out in 300-350°C temperature range and under 50-85 MPa hydrogen pressure in the paper which is discussed.

Such high values of hydrogen pressure led to polymerization of fullerene molecules, destruction of their cages, and hydrogenation of fragments which were formed. It was shown by mass-spectrometric data under dehydrogenation of products, produced under these conditions.

To avoid polymerization running and destruction of fullerene molecules it was needed to lower hydrogen pressure, and therefore to rise synthesis temperature at the same time.

Hydrogen pressure of 12 MPa during catalytic hydrogenation of fullerene molecules in toluene solution did not lead to fullerene molecule destruction and allowed to produce hydrofullerenes with high hydrogen content. From this point of view, working hydrogen pressure 12 MPa was chosen in our experiments.

The next step before beginning of the experimental investigation of fullerene molecules hydrogenation peculiarities was choosing the temperature range, that is the temperature interval, where catalyst free chemical interaction between carbon atoms in fullerene cage and hydrogen atoms leading to covalent bonds formation, occurs. With this aim, slow heating of the system to the temperature above hydrofullerite hydrogenation temperature (530°C), which was given in [3], under hydrogen pressure 12 MPa was needed to carry out.

Due to this reason, the investigations were carried out at 200-600°C temperature range under 12 MPa in pureed hydrogen which was taken from metal hydride unit for hydrogen storage. Experiments show the temperature range 400-500°C to be the optimal value for hydrogen interaction with fullerite C₆₀.

As one can see (Fig.1), temperature arising to 200°C in the reactor and the following 12-hours keeping the system after vacuum making led to pressure rising on 0,4 MPa due to desolvation of the sample. After that the installation reached stable temperature-pressure working regime, adsorption-desorption processes were stabilized associated with distribution of lattice hydrogen on the sample surface and in its volume. After initial preparing of the sample in such manner temperature is gradually arised from 200 to 600°C under hydrogen pressure 12,9 MPa. Temperature rising rate was 0,125° per minute.

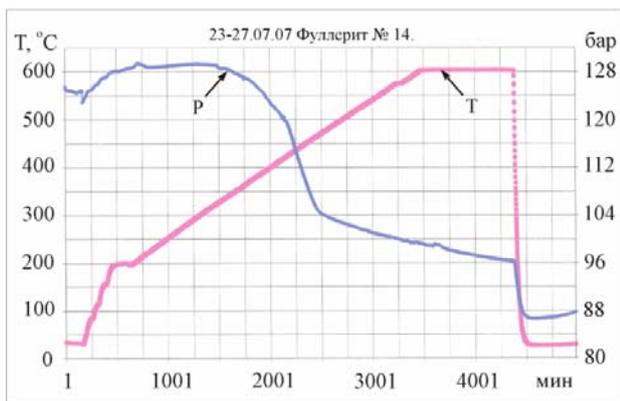


Fig.1. Sorption properties changing of fullerites under heating (dependence of hydrogen pressure changing versus time (temperature)).

During 75-hours experiment is established that the maximal rate of sample mass changing was detected in the time interval between 1500 and 2500 minutes (Fig.2). It corresponded to temperature range 400 - 450°C (Fig.3) in turn.

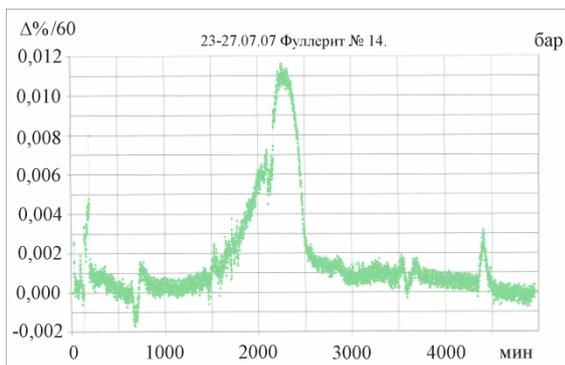


Fig.2. Sorption properties changing of fullerites under heating (rate changing of fullerite interaction with hydrogen during time at $P = 12,9$ MPa).

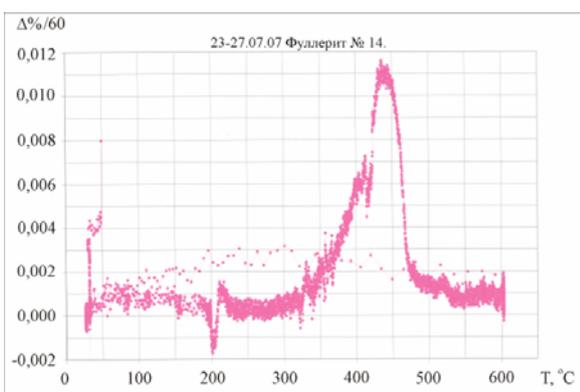


Fig.3. Rate changing of fullerite interaction with hydrogen under heating (duration of heating – 75 hours, rate of heating – 7,5° per hour).

The following experiments on hydrofullerene phases synthesis were carried out at $P = 12,0$ MPa in the found temperature range 400 – 450°C. Various content of hydrogen in hydrofullerite was achieved experimentally by changing of time keeping at these thermodynamic conditions.

Conclusions

The optimal conditions for fullerene C_{60} hydrogenation process are $P = 12$ MPa and $T = 400 - 450^{\circ}C$.

- Long time hydrogenation leads to complete hydrogenation of fullerene molecule under these conditions.

- It is shown experimentally that during molecular hydrogen interaction with fullerite induction time period, which includes surface processes such as adsorption and dissociation of molecular hydrogen, is absent.

- The following steps of hydrogenated molecule $C_{60}H_{60}$ formation are: dissolving of hydrogen molecules in octahedral pinholes of FCC fullerite lattice, dissociation of molecules under hydrogen transfer into tetrahedral pinholes, and chemical interaction of hydrogen atoms with fullerene molecule.

Acknowledgement

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References

1. Jin C, Hettich R, Compton R, Joyce D. Blencoe O.L.J., Burch T. Direct solid-phase hydrogenation of fullerenes. *J. Phys Chem* 1994; 98; 4215.
2. Shigematsu K., Abe K., Mitani M., Tannaka K. Catalytic hydrogenation of fullerene C_{60} . *Chem Express* 1992; 7(12); 37 – 40
3. Schur D.V., Dubovoj A.G., Zaginaichenko S.Yu., Adejev V.M., Kotko A.V., Bogolepov V.A., Savenko A.F., Zolotarenko A.D. Production of carbon nanostructures by arc synthesis in the liquid phase // *Int. J. Carbon.* – 2007. -Vol. 45/6. -P. 1322-1329.