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Development, Fabrication and Study of Fullerene-Containing Carbon Material (FCC) for Immobilization of Iodine: Progress Report 1-3

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Development, Fabrication and Study of Fullerene-Containing Carbon Material (FCC) for Immobilization of Iodine

Progress Reports #1-3

Synthesis of ceramic-like material
by conversion of iodine-doped FCC to Si_xC_y

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1. Introduction

During previous reporting periods we studied properties of iodine doped FCC in comparison with activated carbon. An additional goal was to investigate the possibility to convert iodine doped FCC and activated carbon into ceramic-like material avoiding iodine loss. In order to obtain ceramic based on C_xN_y with fullerene structure we decided to carry out gamma-irradiation of iodine doped samples in nitrogen atmosphere (0.7 atm. N_2). It was found that iodine doping and irradiation caused principal changes in composition of FCC such as substituting of fullerene C_{60} and graphite (both formed by benzene-like carbon rings) for crystalline chaoite phase (formed by carbyne carbon chains). It was assumed that iodine doping of activated carbon, which does not contain significant amount of crystalline phases, also causes formation of carbon chains.
During current reporting period the experiments on irradiation were completed and the results obtained were summarized. The experiments on synthesis of another ceramic-like material by conversion of iodine-doped FCC and activated carbon to Si_xC_y have been continued.

2. Gamma-irradiation of FCC and activated carbon under nitrogen atmosphere

The results of precise XRD analyses of different FCC samples are summarized in Figure 1.

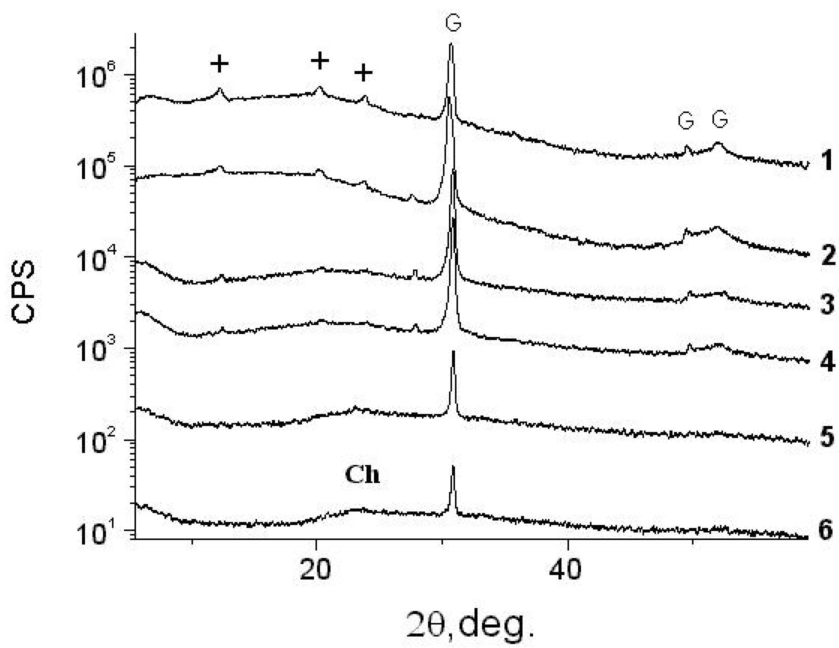

Figure 1. XRD patterns of:
1) initial undoped FCC containing 6.2 wt.% fullerenes (mainly C_{60});
2) undoped FCC after gamma-irradiation for $3.63x10^8$ Rad;
3) iodine doped (145 mg/g) FCC after gamma-irradiation for $2.1x10^7$ Rad;
4) iodine doped (145 mg/g) FCC after gamma-irradiation for $3.3x10^7$ Rad;
5) iodine doped (683 mg/g) FCC;
6) iodine doped (683 mg/g) FCC after gamma-irradiation for $3.63x10^8$ Rad.

Reflections of identified phases are marked as: graphite (**G**); C60 (+) and chaoite (**Ch**).

The results obtained finally confirmed active interaction between iodine and FCC. In our previous work there was a doubt that iodine sorption into FCC is supported by just surface effects, which are typical for highly dispersed powders. To the moment we proved that iodine doping causes destruction of phases based on benzene-like carbon rings such as graphite and fullerenes. The FCC structure based on benzene-like carbon rings is transformed under iodine doping to the chaoite structure based on carbyne chains (Fig. 1, spectra #1 and #5). High iodine doping (683 mg/g) causes complete fullerene destruction and decrease of graphite content up to two times.
Gamma-irradiation of undoped FCC (in nitrogen atmosphere) causes partial destruction of fullerenes but increase of contents of amorphous carbon and graphite (Fig. 1, spectrum #2). Similar effect of gamma-irradiation was observed in FCC with low iodine doping level (145 mg/g): increase of graphite content (depending on cumulative dose), accompanied with an additional partial destruction of fullerenes (Fig. 1, spectra #3 and #4). However, at high iodine doping level (683 mg/g) irradiation causes an additional decrease of graphite content (Fig. 1, spectra #5 and #6).

Previous results on iodine doping of activated carbon allowed us to assume that formation of carbine chains might take place in activated carbon too. However, it requires confirmation by TEM study. For new experiments on irradiation of iodine doped activated carbon we decided to use sample of other trademark without admixtures of sulfur and calcite. No essential effect of gamma-irradiation at $2.1x10^7$ and $3.3x10^7$ Rad has so far been observed (Fig. 2).

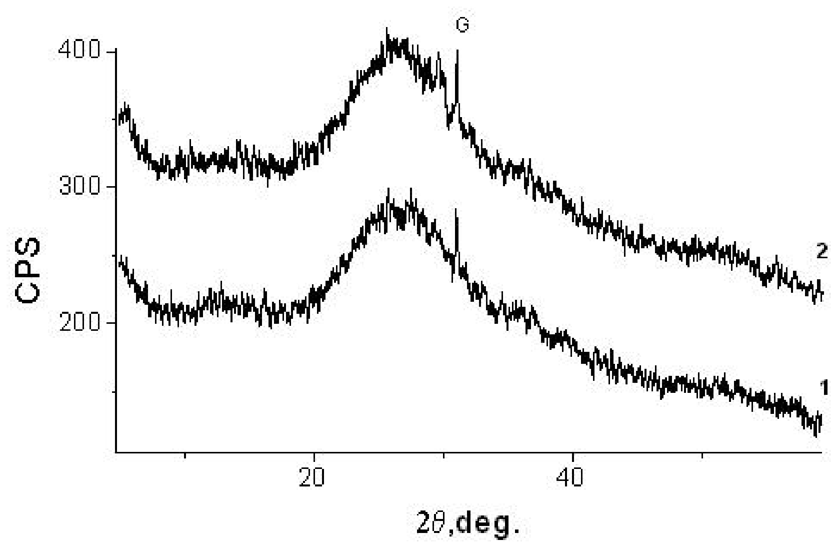

Figure 2. XRD patterns of iodine doped (432 mg/g) activated carbon after gamma-irradiation in nitrogen atmosphere at: 1) $3.3x10^7$ Rad and 2) $2.1x10^7$ Rad. Reflection of graphite phase is marked as (**G**).

It is important to note that the use even precise XRD analysis does not allow identifying formation of C_xN_y phase of fullerene structure in the matrix of FCC or activated carbon.

Further analyses of obtained samples are needed applying method of high resolution transmission electron microscopy (HRTEM).

3. Conversion of FCC and activated carbon into ceramic-like Si_xC_y

Similar method from previous study was used to convert FCC and activated carbon into ceramic-like Si_xC_y material. Cold pressed granules of iodine-doped FCC (iodine content 145 mg/g) and activated carbon (iodine content 432 mg/g) were rinsed in water-ethanol solution of tetraethoxysilane (TEOS), $Si(OC_2H_5)_4$, with the following proportions: 130 ml TEOS, 60 ml ethanol and 290 ml distilled water. Then granules were dried at 60°C for 6-8 hours and calcined in air at temperature: 300; 400 and 420°C for 10 min in order to provide formation of Si_xC_y phase. For each temperature of FCC calcination the loss of iodine during leach test has been studied (Table 1). Activated carbon completely lost iodine during calcination under the same conditions.

Table 1. Correlation between FCC calcination temperature and iodine release after static leach test at 90°C for 7 days in distilled water.

Calcination temperature, °C	Iodine loss	
	in mg/g	in wt.% of initial content
300	less 0.04	less 0.02
400	0.20	0.06
420	0.37	0.11

It was demonstrated that higher temperature of calcination provides increase of mechanical durability of the final ceramic-like material, however, it causes increase of iodine release during leach test. This means that tetraethoxysilane is not optimal chemical compound for low temperature interaction with FCC as well as with activated carbon. As in previous research of the first year, the results of XRD analysis did confirm formation of crystalline Si-carbide phase. We assumed that future TEM study of ceramic-like samples would be useful to check the presence and making identification possible impurities of crystalline S-C phases. Optimal conditions for more effective conversion of iodine doped FCC into Si_xC_y material avoiding iodine loss are still the subject for further research.

4. Conclusions

The results obtained allow us to make the following conclusions:

1) Iodine actively interacts with FCC. Doping of FCC with iodine causes destruction of phases based on benzene-like carbon rings such as graphite and fullerenes. Therefore, initial structure of FCC based on benzene-like carbon rings is transformed under iodine doping to the chaoite structure based on carbyne chains.
2) Decrease of fullerenes and graphite amounts in FCC correlates directly with increase of iodine doping level. At highest iodine doping (683 mg/g) fullerenes disappear completely and graphite amount decreases up to two times;
3) Gamma-irradiation of undoped FCC (in nitrogen atmosphere) causes partial destruction of fullerenes but increase of contents of amorphous carbon and graphite;
4) Results of gamma-irradiation of iodine doped FCC (in nitrogen atmosphere) depend on iodine content. At high iodine doping level (683 mg/g) irradiation causes an additional decrease of graphite content. At low iodine doping (145 mg/g) irradiation causes increase

of graphite content (depending on cumulative dose), accompanied with an additional partial destruction of fullerenes.

5) Iodine doping of activated carbon, which contains insignificant amount of graphite, causes as assumed similar to FCC formation of carbine chains. However, it requires confirmation by TEM study.
6) There was no essential effect of gamma-irradiation (in nitrogen atmosphere at $2.1\text{x}10^7$ and $3.3\text{x}10^7$ Rad) observed so far in samples of iodine doped activated carbon.
7) FCC samples partially converted into Si_xC_y compound are characterized with increased mechanical durability and low iodine release during leach tests. However, the search for more optimal Si-organic chemicals is needed to provide effective low temperature interaction with FCC avoiding iodine loss.
8) In comparison with FCC the conversion of iodine doped activated carbon to ceramic like material was unsuccessful due to complete iodine loss.
9) Method of X-ray powder diffraction does not allow identifying possible formation of C_xN_y phase with fullerene structure as a result of FCC gamma-irradiation in nitrogen atmosphere. The same problem is typical for investigation of ceramic-like samples based on Si_xC_y . The detailed TEM study of irradiated samples is required.

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