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Development, Fabrication and Study of Fullerene-Containing Carbon Material (FCC) for Immobilization of Iodine: Progress reports 1-1 and 1-2

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Research-Industrial Enterprise “KIRSI”, branch of the V.G. Khlopin Radium Institute (KRI)

UNLV – KRI-KIRSI Agreement 280203-1

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

Progress Reports #1-1 and #1-2

Preparation of FCC samples for gamma-irradiation
Experiments on irradiation and examination of irradiated
FCC by precise XRD and other methods

Saint-Petersburg - 2004

1. Introduction

Immobilization of highly radioactive and long-lived isotope of ^{129}I , which is a fission product in spent nuclear fuel, requires development of new durable host-materials. Such materials should be characterized with the following principal features:

- high loading capacity to iodine incorporation;
- chemical durability and radiation resistance in order to prevent iodine release over long time (higher than 10^5 years) storage under conditions of underground repository of radioactive wastes;
- possibility to use iodine-doped material as a target for iodine transmutation.

Iodine is a very volatile chemical element, and even its chemically strong compounds such as AgI and CuI are not stable under ultraviolet irradiation or oxidizing conditions. Therefore, development of host materials for iodine immobilization is based on unusual approach – the search and testing of new compounds which were not studied before in respect of iodine sorption and strong fixation.

Laboratory of Applied Mineralogy and Radiogeochemistry of the V.G. Khlopin Radium Institute carries out investigation of prospective materials for iodine immobilization including transmutation since 1994 year. Recently, a family of new discovered carbon molecules – fullerenes – was suggested for incorporation and strong fixation of highly radioactive elements. It was found that pure crystalline fullerenes were not effective for the iodine incorporation. However, fullerene-containing carbon material (FCC) which is used as starting precursor for fullerene extraction was characterized by high capacity to iodine sorption (higher than 450 mg iodine per gram FCC). This powder consists of three carbon forms: graphite, fullerenes and amorphous carbon, which associate with each other by well not studied bonds.

Although FCC material demonstrated high loading capacity to iodine sorption it is necessary to take into account that FCC itself is not a final form of ^{129}I immobilization. Either for disposal in geological formation or iodine transmutation in nuclear reactors the FCC should be converted into more chemically and mechanically durable material. In case of transmutation it is necessary to provide, also, an acceptable «transparency» of final material to neutron fluxes. Therefore, we decided to consider two main groups of durable compounds: silicon carbides and carbon nitrides (of fullerene structure) as possible targets of FCC conversion. It was assumed that synthesis of Si_xC_y might be carried out at relatively low temperature as a result of chemical reaction between FCC and Si-organic chemicals. Successful synthesis of C_xN_y of fullerene-like structure might be carried out using N-ion bombardment of carbon material.

In the framework of first year (“KRI-KIRSI”-HRC Agreement 280203) we have studied optimal synthesis conditions and principal features of FCC. The preliminary results have been obtained from precise XRD analyses of iodine-doped and pure FCC samples before and after gamma-irradiation. It was found that iodine doping and irradiation caused principal changes in phase composition of FCC such as substituting of crystalline fullerene C_{60} (formed by benzene-like carbon rings) for crystalline chaoite phase (formed by carbyne carbon chains). It was decided to continue these experiments during second (current) year (“KRI-KIRSI”-HRC Agreement 280203-1) in comparison with study of activated carbon. In our experiments we decided to use samples of activated carbon SKT-3S that is widely used in Russian nuclear industry for different purposes including sorption of ^{129}I .

2. Synthesis of FCC samples, iodine doping and irradiation of FCC and activated carbon

The graphite of high purity was used as starting material for the synthesis of FCC. Highly disperse powder of FCC was obtained by electric-arc method under conditions of glove box in helium atmosphere (Fig. 1).

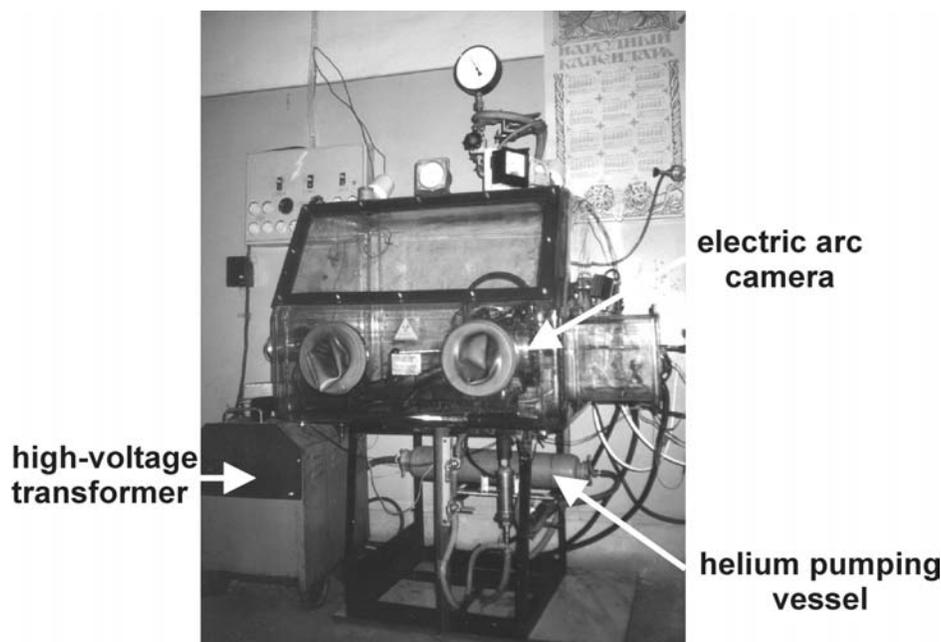


Figure 1. Electric-arc equipment installed into glove box for the synthesis of FCC material.

The samples of FCC and activated carbon SKT-3S were dried at 105-110°C up to constant weight, and then placed in glass ampoules. The crystalline iodine was put into the extension of the same ampoules. Then the atmosphere in the ampoule was replaced on high purity nitrogen and the ampoules were sealed. Then the ampoules were placed in a furnace and maintained at 300°C for 8 hours. After the exposure the unreacted iodine was re-crystallized into the extension and the ampoules were cooled to room temperature.

Parts of initial and iodine-doped samples of FCC and activated carbon were sealed in glass ampoules in nitrogen atmosphere (0.7 atm. N₂) and then irradiated by gamma-source (⁶⁰Co). Irradiation of FCC samples was completed after cumulative dose 3.63 x 10⁸ Rad. Irradiation of samples of activated carbon started later and at the moment is not finished.

The following samples have been obtained:

- 1) Initial pure FCC with fullerene (mainly C₆₀) content 6.2 wt.%;
- 2) Iodine doped FCC with iodine content 683 mg/g;
- 3) Pure FCC after gamma-irradiation for **3.63 x 10⁸ Rad** in nitrogen atmosphere;
- 4) Iodine-doped FCC (683 mg/g) after gamma-irradiation for **3.63 x 10⁸ Rad** in nitrogen atmosphere;
- 5) Initial pure activated carbon SKT-3S;
- 6) Iodine doped activated carbon SKT-3S with iodine content 571 mg/g.

3. Precise XRD analyses

Results of precise XRD analyses confirmed previous typical data that initial FCC consisted of three main carbon phases: graphite; fullerene (mainly C₆₀) and amorphous carbon (Fig. 2, spectrum 1). Iodine doping caused essential decrease of graphite content and substitution of C₆₀ for chaoite phase (Fig. 2, spectrum 2).

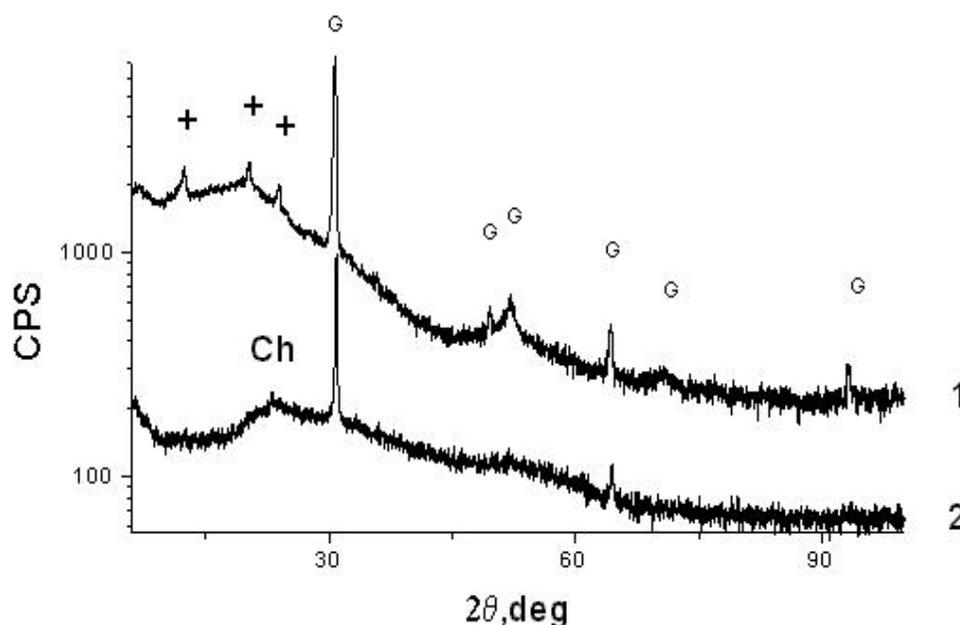


Figure 2. XRD patterns of FCC: 1) initial undoped with fullerene content 6.2 wt.%; 2) iodine-doped (683 mg/g). The following peaks are marked: “G” – graphite; “+” – C₆₀; “Ch” – chaoite.

Gamma-irradiation in nitrogen atmosphere for dose 3.63×10^8 Rad did not change phase composition of initial and iodine-doped FCC (Fig. 3). However, irradiation of undoped FCC was accompanied with partial conversion of C₆₀ into amorphous carbon and increase of graphite content approximately 3 times higher in comparison with un-irradiated sample (spectrum 1 in Fig. 3 and Fig. 2). In contrast, irradiated iodine doped FCC was characterized by essential decrease of graphite content (Fig. 3, spectrum 2). It might be explained that gamma-irradiation stimulates interaction between iodine and carbon rings and formation of carbyne carbon chains.

Activated carbon SKT-3S does not contain crystalline carbon forms such as graphite and fullerenes (Fig. 4). Admixtures of calcite (CaCO₃) and sulfur, which are particular additives in this type of industrial activated carbon, caused formation of anhydride (CaSO₄) during iodine

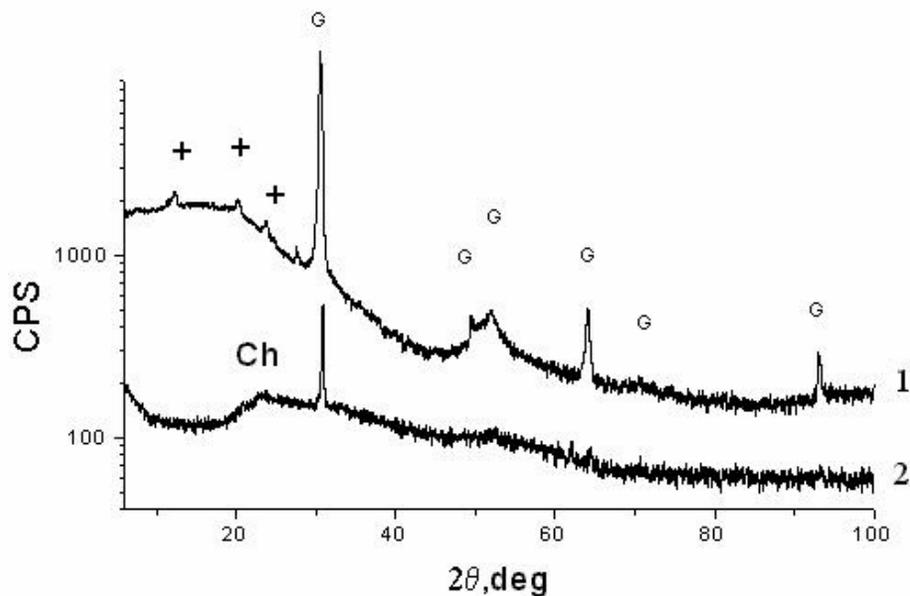


Figure 3. XRD patterns of FCC after gamma-irradiation for 3.63×10^8 Rad: 1) undoped; 2) iodine-doped (683 mg/g). The following peaks are marked: “G” – graphite; “+” – C₆₀; “Ch” – chaoite.

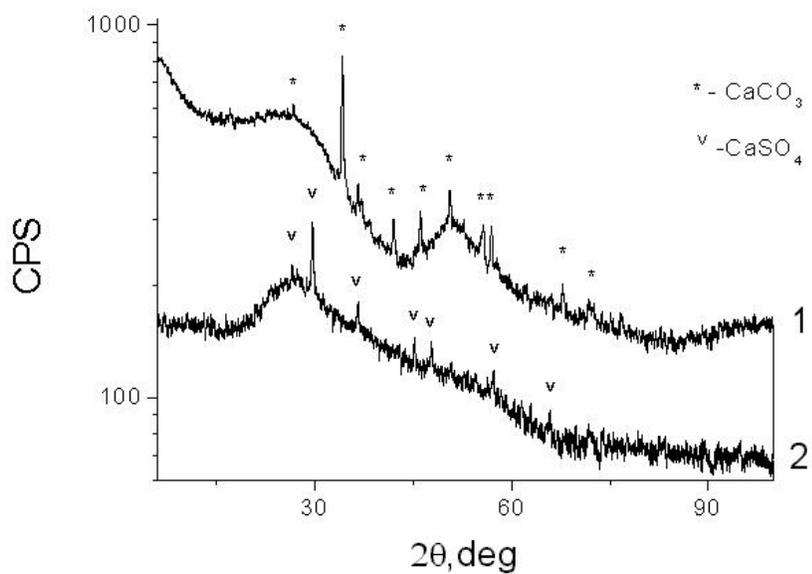


Figure 4. XRD patterns of activated carbon SKT-3S: 1) initial undoped; 2) iodine-doped (571 mg/g). There are no crystalline carbon phases in both samples. Type of activated carbon SKT-3S contains admixtures of calcite (CaCO₃) and elemental sulfur that causes formation of anhydride (CaSO₄) during process of iodine doping at 300°C.

doping at 300°C. However, no evidences of interaction between iodine and calcite were observed. It is important to note that XRD patterns of activated carbon (initial and iodine doped) in low angle area (before 20° of 2 theta) are very similar to ones of FCC (Figs. 2,4). This is additional evidence that iodine doping provides conversion of carbon rings into carbyne carbon chains.

4. Conclusions

The results obtained allow us to make the following conclusions:

- 1) New samples of pure and iodine doped FCC and activated carbon were obtained and studied.
- 2) Iodine doping of FCC is accompanied with change of FCC structure and phase composition. As a result of iodine doping the **chaoite** crystalline phase substitutes fullerene C₆₀ phase and graphite content decreases. General structure of initial FCC based on **benzene-like** carbon rings changes under iodine doping to **carbyne** chains.
- 3) Iodine doping of activated carbon, which does not contain crystalline phases, causes nevertheless similar process of carbon rings destruction and as assumed formation of carbyne chains.
- 4) Gamma-irradiation of initial undoped FCC in nitrogen atmosphere causes partial conversion of C₆₀ into amorphous carbon and essential increase of graphite content.
- 5) Gamma-irradiation of iodine doped FCC in nitrogen atmosphere is characterized by essential decrease of graphite content. It is assumed that gamma-irradiation stimulates interaction between iodine and carbon rings and formation of carbyne carbon chains.
- 6) Gamma-irradiation of samples of pure and iodine doped activated carbon is under continuation.

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