EUROPEAN COMMISSION

Community Research

SEVENTH FRAMEWORK PROGRAMME

CARBOWASTE

Treatment and Disposal of Irradiated Graphite and Other Carbonaceous Waste

Grant Agreement Number: FP7-211333

Technical Report (T-5.2.2)
Interim Report on Potential New Products within the Nuclear Industry

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Reporting period: e.g. 01/03/2009 – 31/03/2010

Date of issue of this report: 16/04/2010

Start date of project: **01/04/2008** Duration: **48 Months**

Project co-funded by the European Commission under the Seventh Framework Programme (2007 to 2011) of the European Atomic Energy Community (EURATOM) for nuclear research and training activities		
Dissemination Level		
PU	Public	
RE	Restricted to the partners of the CARBOWASTE project	**X**
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CARBOWASTE		
Work package: 5 **Task: :** 5.2.2	**CARBOWASTE document no:** CARBOWASTE-1004-T 5.2.2 (e.g. May 2008 as date of issue: 0805)	**Document type:** T
Issued by: Bradtec (UK) **Internal no.:** Bradtec Report No 01/10		**Document status:** Final

Document title
Interim Report on Potential New Products within the Nuclear Industry
Executive summary

Revisions

Rev.	Date	Short description	Author	Internal Review	Task Leader	WP Leader
1	16/04/2010	Draft for WP5 review	JC Goodwin	D Bradbury	JC Goodwin	D Bradbury
	04/06/2010	Final	JC Goodwin	D Bradbury	JC Goodwin	D Bradbury

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1 Introduction

The objective of this document is to provide an interim progress report on the work undertaken by the Work Package 5 (WP5) participants under Task 5.2 – Identification and Making of New Products. The participants under this task are GrafTech, SGL, Necsa and NRG, with the management responsibility of the task held by Bradtec and Hyder. A more comprehensive report (Task 5.2.4) will be produced in May 2011 which will incorporate the information contained within this interim report with the future advance made in this area during the intervening period.

1.1 Scope of Task 5.2: Identification and Making of New Products

The following is an extract from the original scope of work, [1] "This task involves the production, on a small scale of candidate products (e.g. graphite, silicon carbide) from simulated or real recycled carbon. This will be achieved by collaboration between the members of WP5, passing intermediate materials from one facility to another. However, where this is not practical (e.g. where the recipient organisation can only, at the moment, accept non-radioactive material) care will be taken to ensure that any simulant intermediates used for product production match closely the characteristics of real intermediates, such as "synthesis gas" created from the treatment processes arising from WP4. This task may also include making candidate products directly from graphite without chemical transformation / decontamination".

This interim report provides a summary of the work to date by the WP5 participants in respects to both Sub Tasks 5.2.2. and 5.2.3, which are described overleaf.

1.1.1 Sub Task 5.2.2: Other Graphite and Carbon based Products

The following is an extract from the original scope of work [1], "In addition to the potential reuse/recycle of graphite for use in new reactors or fuel components (see Sub Task 5.2.1), other potential opportunities exist. These include its use in graphite electrodes for the immobilisation by high temperature processing of certain nuclear wastes, or in activated charcoal filters. Opportunities may be more limited, but the technical quality of the required materials may be less demanding than the reactor application and hence easier to achieve."

The work undertaken to date by GrafTech, SGL and Necsa as part of this sub task are reported here.

1.1.2 Sub Task 5.2.3: Carbonaceous Ceramics for Waste Management

The following is an extract from the original scope of work [1]," Carbonaceous ceramics for waste management addresses graphite canisters for waste storage as well as non-graphite uses of other chemical forms of carbon. This category includes materials such as silicon carbide and calcium carbonate. The definition for the purposes of this project of "recycled products" specifically includes products specifically manufactured to stabilise carbon in a radioactive waste disposal site, or to act as confinement of packing material for other wastes and thermal management, in the repository site. The task will include the production of materials which are suitable for use in disposal sites as a backfill, encapsulants etc. without oxidation of the carbon and providing safety during processing."

The work undertaken to date by NRG as part of this sub task are reported here.

2 Opportunities for Recycled Graphite Products

Under Task 5.2 of Work Package 5 (WP5) various opportunities are being investigated for the recycle / reuse of irradiated graphite (i-graphite) and its use in the production of new products for use in the nuclear industry. The contributing organisations to this work are GrafTech, SGL, Necsa and NRG; with management responsibility of the Work Package 5 held by Bradtec with 3rd part contribution from Hyder.

The opportunities for graphite reuse / recycle within the nuclear industry being considered within this interim report are its use as:

- Electrodes for use in nuclear waste vitrification (GrafTech),
- Nuclear graphite moderator for gas cooled reactors (GrafTech / SGL) ,
- Production of Silicon Carbide (NRG).
- Absorbent material to remove nuclides from liquid waste (NECSA),
- Absorbent material to remove nuclides from gaseous waste streams (NECSA),
- Electrodes to dissolve spent fuel (NECSA),
- Crucible to dissolve spent fuel (NECSA),
- In the manufacture of nanotubes for use in waste disposal technology (NECSA),

Each of options listed above is considered over the following sections with the progress to date reported by the participating organisation. It should be noted that as some of the organisations participating under WP5 are commercial entities, therefore detailed information pertaining to elements of the processes employed may be omitted due to its proprietary nature.

3 Contribution by GrafTech

GrafTech International are investigating the possibility of using a so-called "fast" manufacturing process to fabricate a graphite electrode to be used for the vitrification of nuclear waste. This graphite electrode should be made from a maximum fraction (currently unknown) of carbon black that will be derived from the purification of irradiated graphite waste.

Specifically, this work aims to define:

- the maximum fraction of carbon black or other recycled material that can be utilized in a "new" graphite artifact without compromising the strength and other physical and mechanical characteristics of the said graphite artifact.
- the ideal processing conditions to produce such a graphite artifact that will include a fraction of carbon black and/or other recycled material. This includes the investigation of a "fast" process to manufacture graphite artifacts as a possible means to produce samples for evaluation [2].

3.1 Scope of Work

GrafTech will fabricate 5 different varieties of graphite material that are made with 10%, 20%, 30%, 40% and 50% carbon black (% carbon black will be based on the dry fraction of the mix design) in order to evaluate the change in properties that occur as the fraction of carbon black within the graphite increases. They will aim to make several small billets at each of the described fractions of carbon black using there fast processing technology that allows them to drastically reduce the standard production time required to fabricate synthetic graphite artifacts [2].

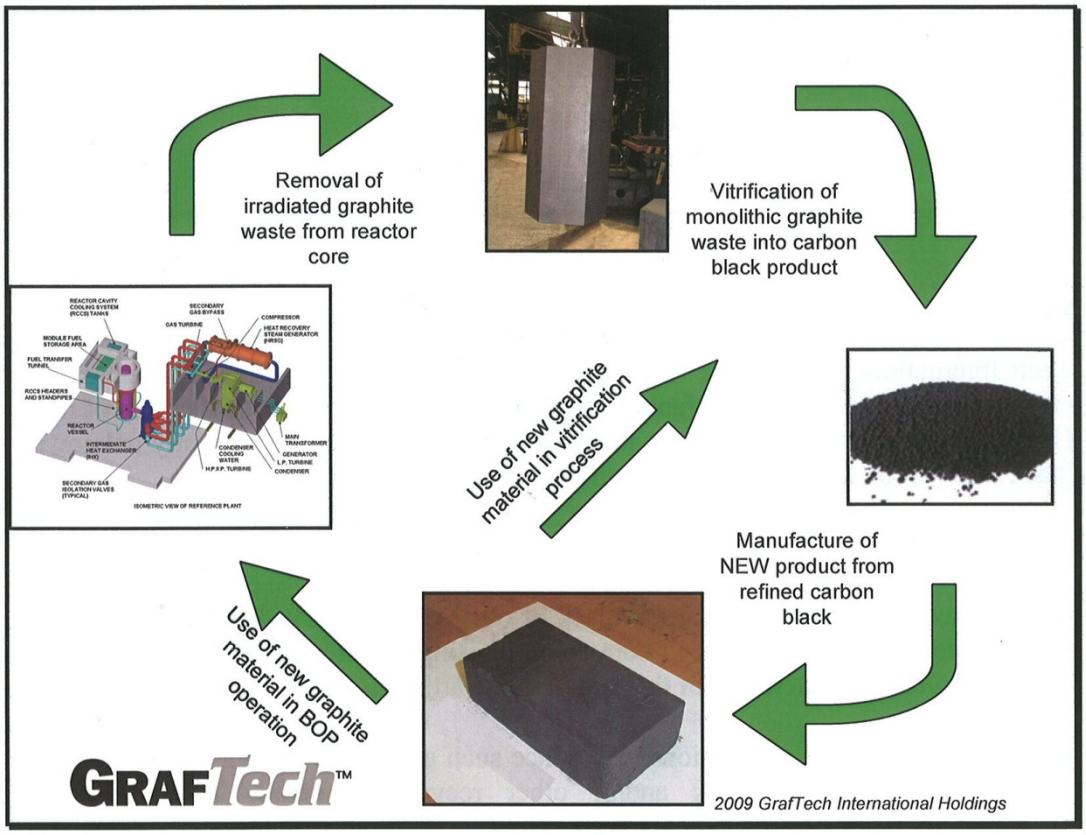

Figure 3.1: - Background Concept of the work being undertaken by GrafTech [2]

Once fabricated, the said material will be characterized for coefficient of thermal expansion, sonic modulus, specific resistance, density and flexural strength. Results will be provided to the Work Package leader [2].

The goal of this work is to prove the concept of utilizing carbon black as an additive in a new graphite artifact to be used for nuclear waste vitrification [2].

3.2 Progress to Date and Further Work

- Background survey of operational parameters for "fast-processing" technology has been investigated to optimize parameters necessary for this work.
- Raw materials for project have been ordered.
- Design and engineering of dry mixture to be used for project is complete.
- GrafTech has formed roughly 50% of the planned amount of artifacts for Task 5.2
- Artifacts have been made using proprietary 'fast' processing technology and utilize carbon black as the waste simulant.
- Artifacts made with the carbon black fraction require significantly higher amounts of pitch due to the sizing of the carbon black.
- Artifacts will continue to be formed in April. Further processing will follow forming.
- Machining and final testing is to be completed by the end of July 2010 [3].

3.3 Work outside of Carbowaste

In addition to GrafTech's work under Carbowaste, they are also involved in Deep Burn project, working closely with Oak Ridge National Laboratory on behalf of the US Department of Energy. The results of the work to date under this project were reported in two presentations given at 10th International Nuclear Graphite Specialists Meeting [4, 5]. The key conclusions arising from this work are:

- It has been shown that recycle of irradiated graphite is possible.
- A graphite specimen made from 100% recycle material was also possible, and actually outperformed a sample with synthetic graphite filler.
- Industrial scale production showed that 100% recycle graphite could be made to the same density as "virgin material".
- High fractions of recycled graphite do not significantly change the processing or manufacture of the material.
- The properties of graphite when made with a large fraction of recycled material do not change significantly versus the control material.
- Once graphitized, the structural characteristics of material made with a large recycled content is very similar to that of the original material.

4 Contribution by SGL

4.1 Scope

The scope of work which SGL are responsible for under Task 5.2, includes the following tasks:

- Identification of suitable decontaminated carbon material as raw material for new products
- Definition of options to recycle:
  - in nuclear graphite
  - additive to other carbon and graphite products
- Lab tests with corresponding carbon surrogate, including products characterization

4.2 Preconditions/Constraints:

- SGL is not able to handle decontaminated i-graphite on its premises, unless deemed completely non-hazardous. In particular, radiating elements such as Co60 cause significant obstacles for any recycling in carbon and graphite-producing industries. This has to be considered for any recycling route outside of nuclear area, especially in commercial volumes
- Option exists to produce samples at partners which are equipped to handle i-graphite after cleaning. However, for meaningful tests 0.5 to 1 kg of material is required.
- Therefore, the starting material is recommended to be taken from the gas-phase decontamination route (e.g. steam reforming).
- WP5 is strongly depending on results from WP4 (Treatment & Purification). Only, when most promising decontamination route(s) are identified, tests with corresponding material can be initiated. Therefore, involvement in WP4 progress and communications is vital [6, 7].

4.3 Potential New Products

The products which SGL is investigating include:

- Nuclear Applications:
  - Nuclear Graphite (Moderator in gas-cooled reactors)
  - Graphite powder for pebble fuel
- Other Applications:
  - Additive to conventional carbon and graphite (graphite electrodes, cathodes for Al smelting, blast furnace linings, etc., but: see constraints)
  - Additive to fuel in blast furnaces, cement kilns, etc.

4.4 Planned Activities

SGL's proposed activities under Task 5.2.:"Identification & Making of New Products" are as follows:

- For fuel application (pebble graphite) and other use in graphite manufacture, checking of graphitizability by thermal treatment of carbon black at 2400 , 2600, 2800, 3000 ºC, to evaluate whether we can (re-)generate graphite characteristics via thermal treatment. Analysis via X-Ray Powder Diffraction.
- Addition – by coke flour replacement – of 10 % and 20 % carbon black (i.e. surrogate for decontaminated i-graphite) to nuclear graphite. Identification of behaviour and properties vs. reference samples
  - Evaluation of formulation coke, carbon black, and pitch
  - Forming of pilot plant samples (kg scale), from coke, carbon black, and pitch
  - Baking and graphitization (optional: impregnation/re-baking)
  - Determination of critical product properties (e.g. apparent density, mechanical strength, thermal conductivity, purity, …)

- Based on outcome of WP4, alternative recycling routes within nuclear or other carbon and graphite products may be considered, theoretically or in practice, depending on scope, timeline, and material available [7].

5 Contribution by NRG

NRG are responsible for elements of work under Sub-Task 5.2.3: Carbonaceous ceramics for waste management. The focus of this sub task is in respects to carbonaceous ceramics for waste management. It addresses graphite canisters for waste storage as well as non-graphite uses of other chemical forms of carbon. This category includes materials such as silicon carbide and calcium carbonate. The definition for the purposes of this project of "recycled products" specifically includes products specially manufactured to stabilise carbon in a radioactive waste disposal site, or to act as confinement or packing material for other wastes and thermal management, in the repository site. The task will include the production of materials which are suitable for use in disposal sites as backfill, encapsulants etc. without oxidation of the carbon and providing safety during processing [1].

NRG are focusing on the synthesis of silicon carbide, through a two stage approach, which is:

1. Synthesis of ceramic power from virgin graphite and characterization of the made powder, and if successful, this will then be followed by,
2. Synthesis of ceramic power from irradiated graphite and characterization of the made powder.

Characterization methods employed by NRG, include X-Ray powder diffraction (XRD), Microscopy (Licht microscopy and SEM), Particle size measurements (LALLS, Malvern), DTA/TG (up to 1350ºC) and Density measurments (geometric and He-pycnometry) [8].

5.1 Properties of Silicon Carbide

When considering the fabrication and synthesis of silicon carbide, it is important to understand the properties of the end product in order to consider the potential applications for which the end product is well suited for. A summary of the properties of silicon carbide is as follows:

- **Structure**: Due to the small electro-negativity difference between C and Si → predominantly covalent bond (88 %) → strong, directional bonds, responsible for high decomposition temperature and hardness of SiC. SiC occurs in two general crystalline forms: **cubic β-SiC** (low-temperature) and **α-SiC** (high-temperature). Usually, α-SiC is obtained by carbothermal reduction of quartz (Acheson process), while β-SiC forms by direct reaction of Si and C (self-sustaining reaction), vapor phase routes and using plasma or laser heating.
- **Microstructure, properties and behavior**: Fine-grained microstructures with isometric grains are desired in SiC → best mechanical properties.
- **Typical properties of SiC**: Density 3.2 g/cm^3, hardness 21-25 GPa, fracture

  toughness 3-6 MPa·m1/2, Young's modulus 420 GPa, bend strength 450-650

  MPa, thermal expansion coefficient 4-5·10-6 K-1, thermal conductivity of order

  50 W/mK [8].

5.2 Literature Review

A literature search was undertaken in 2009 to investigate methods suitable for synthesis of Silicon Carbide (SiC) from irradiated graphite. Various methods were identified for fabrication of SiC, these include:

- Acheson Process
- Chemical Vapor Deposition
- Reaction Milling
- Microwave Synthesis
- Combustion Synthesis
- Field Assisted Combustion Synthesis (FACS)

These are discussed in more detail over the following sections (6.2.1 to 6.2.6). Of the methods investigated, Field Assisted Combustion Synthesis was identified as the most appropriate means for fabrication of SiC under the requirements of NRG's commitments to WP5 [9].

5.2.1 Acheson Process

In this process, quartz sand (SiO_2) and coke (C) are heated with sawdust and salt in an electric furnace to form a mass of small hard SiC crystals called "carborundum". The SiC forms by the carbothermic reduction of sand according to the reaction:

$$\mathbf{SiO_2+3C \rightarrow SiC+2CO \qquad (T: 2000 – 2500\ ^{o}C)}$$

The resultant material is generally used as an abrasive.

As a result of the high synthesis temperatures required, and therefore the significant costs associated with it, this process was deemed unsuitable for the production of the bench scale trials proposed [8].

5.2.2 Chemical Vapour Deposition (CVD)

In CVD, silicon and carbon-containing chemical compounds (precursors) are heated to form a gas phase rich in silicon and carbon-based molecular species. The silicon and a carbon containing species, generally at temperatures of 1200-1400°C., react to form SiC according to the reaction:

Si-R1 (g) + C-R2 (g) → SiC + gaseous by-products.

Si-R1 and C-R2 represent Si and C-bearing compounds, such as silane and propane, respectively, (U.S. Pat. No. 5,704,985).

The SiC is usually deposited on a suitable substrate, typically graphite, to form a solid layer, although it is possible to form and collect SiC powder by such reaction schemes. In another variant of CVD, the precursor is a compound containing both Si and C atoms [8].

5.2.3 Reaction Milling in a Dual-Drive Planetary Mill

In this method silicon carbide is synthesized from elemental silicon and graphite powder by high-energy reaction milling in a specially built dual drive planetary mill. Reaction milling has become a very effective means of synthesizing compounds from the corresponding elemental constituents. Silicon carbide is synthesized from elemental silicon and graphite powder by high-energy reaction milling in a specially built dual drive planetary mill. Reaction milling in this type of mill is most affected by the size of the grinding media. The phase evolutions, particle size distribution and morphology of particles during milling are studied during a 40 hours grinding period. Particle size distribution shows considerable amount of material is below 10 micron. X-ray diffraction study indicates complete conversion of silicon and graphite to silicon carbide. The crystallite size varies from 10 micron from the start to 100 nm after 40 hours of milling. The SEM picture clearly reveals the progress of size reduction and the extent of agglomeration of particles.

5.2.4 Microwave Synthesis of ß-SiC Powder

A mixture of silica gel, graphite as carbon black is preheated and SiC is formed through the application of microwaves. The steps of this process are shown below in Figure 5.1, and the graph showing the X-ray diffraction patterns of samples heated in the furnace.

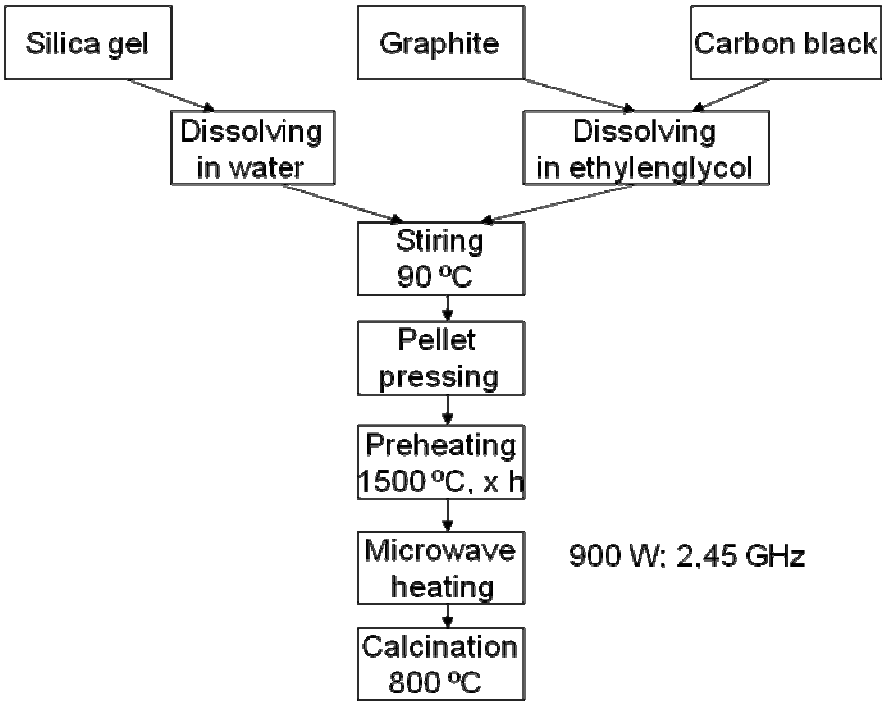

Figure 5.1: - Flow diagram highlighting the process steps for Microwave Synthesis of ß-SiC Powder [8]

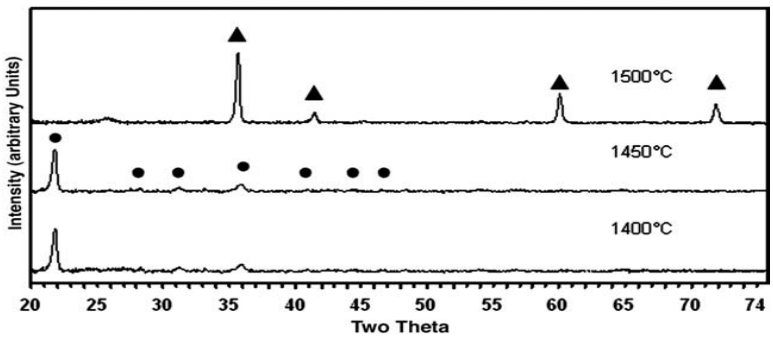

Figure 5.2: - X-Ray Diffraction Patterns of Samples Heated in Furnace at 1400° and 1450 °C for 120 min and at 1500 °C for 105 min (●; SiO_2, ▲ ; SiC) [8].

5.2.5 Combustion Synthesis of Silicon Carbide

In this study, the use of SiC combustion synthesis for immobilization of ^{14}C was considered. Due to the low exothermicity of the reaction between silicon and graphite, a highly exothermic mixture (magnesium and polytetrafluoroethylene) was used both as a chemical oven and activate additive in the mixture. With this configuration the reaction between graphite and silicon was initiated and propagated on the whole sample. The self-propagating high temperature synthesis samples were characterized by using scanning electron microscopy and X-ray diffraction [8].

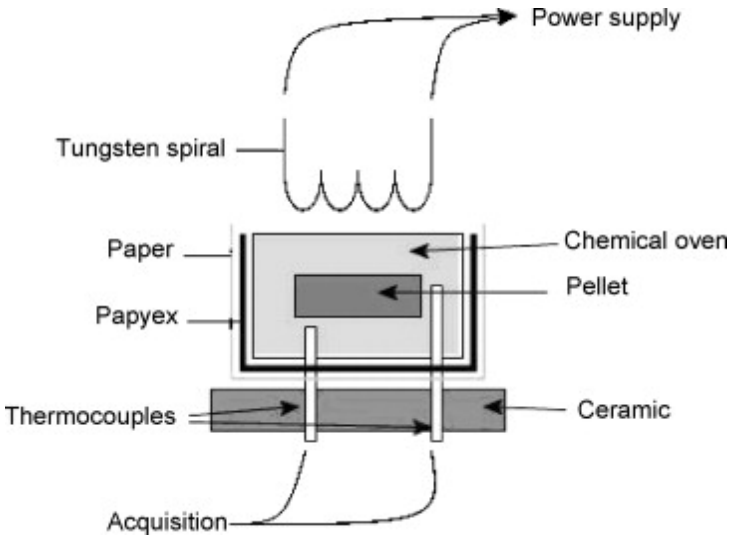

Figure 5.3: - Device used for the combustion synthesis of silicon carbide assisted by a magnesium plus polytetrafluoroethylene mixture [8]

5.2.6 Field Assisted Combustion Synthesis (FACS)

This process consists of applying, at room temperature, a significant voltage across the reactive sample and to initiate locally the reaction.

The FACS method is being tried to synthesize SiC from graphite. First tests are running with virgin graphite and silicium [9]. The synthesis is being undertaken as follows:

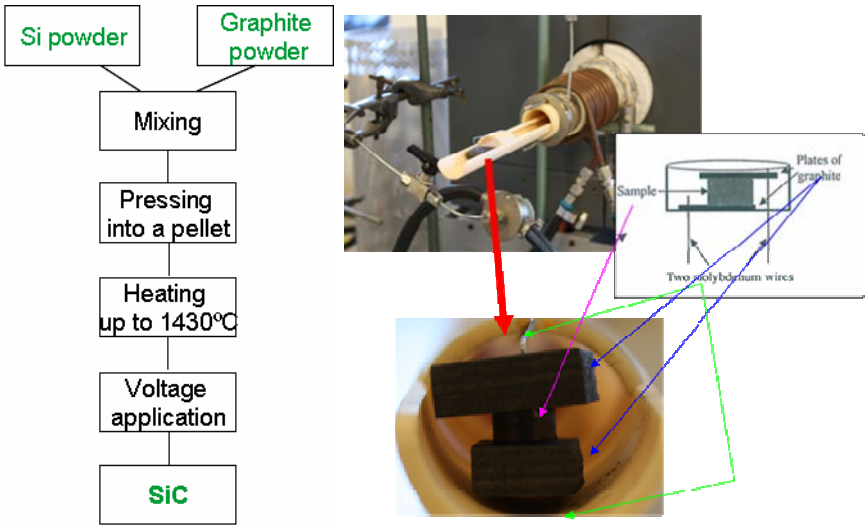

Figure 5.4: - Setup for FACS [9]

A special setup has been designed to perform this synthesis. The pressed pellets are put between two graphite plates where the molybdenum wires are placed in. This setup is put into the tubular furnace (under argon, water cooling), heated up to 1430ºC and the voltage is applied. That leads to a thermal explosion, characteristic for an SHS reaction inside the sample, i.e. SiC is formed [9].

5.3 Further Work

If synthesis of SiC is successful finished (including characterization), the same technique will be applied for irradiated graphite to synthesize $Si^{14}C$. All obtained powders will be characterized by suitable techniques (XRD, LM/SEM) [9].

6 Contribution by NECSA

6.1 Absorbent for Removal of Soap

At nuclear installations clothing decontamination is achieved with different detergents and the resulting effluent contains organic residues. In order to release this effluent to the environment, the organic contaminants must be removed. The result indicated in Figure 6.1 clearly demonstrates that irradiated graphite can be used to remove detergents used in South African nuclear institutions [10].

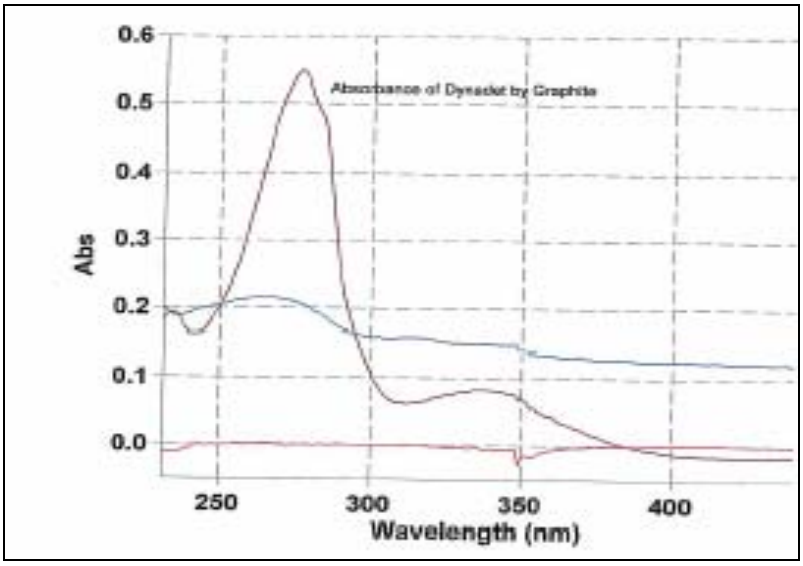

Figure 6.1: UV-spectra of the removal of organic detergents used in South African nuclear industries from effluent waste streams [10]

Key
Brown: original waste stream with detergent
Blue: waste stream after 1st absorption
Red: waste stream after 2nd absorption

6.2 Removal of Radionuclides from Waste Streams

Research in the past has been focused on improving processes for the removal and recovery of radionuclides from nuclear waste streams using ion exchange resins. Generally speaking, the expensive resin material is contaminated and has to be disposed of. By replacing the resin with irradiated graphite, contaminated material from the PBMR could be used to recover radionuclides from a PBMR waste stream. The absorption coefficient values for ^{89}Sr (50 000 ml/g), ^{110m}Ag (40 000 ml/g) and ^{131}I (8 000 ml/g) in Figures 6.2 and 6.3, illustrate that irradiated graphite can be used as an ion exchange medium [10].

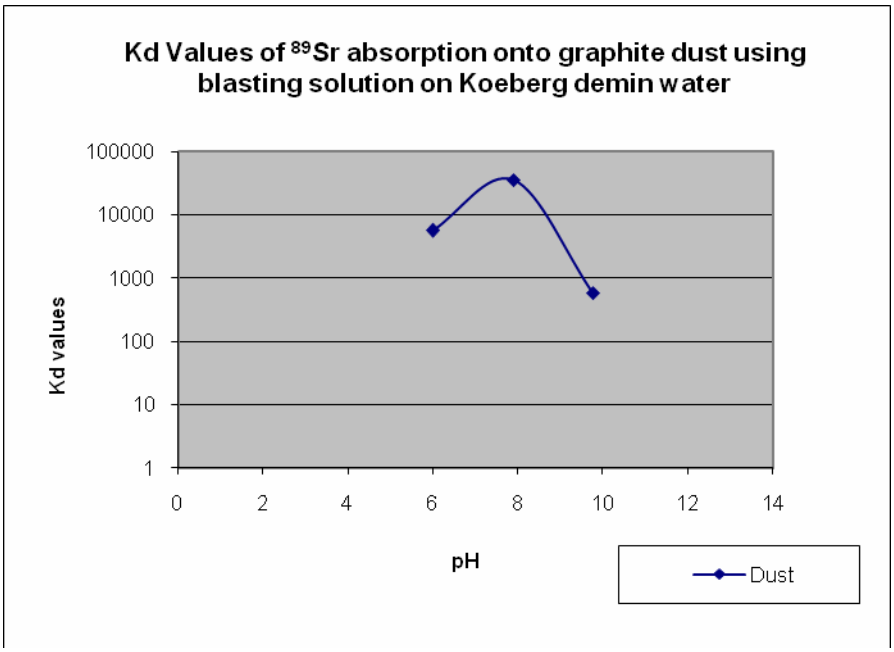

Figure 6.2: Removal of ^{89}Sr from a PBMR waste stream using irradiated graphite as ion exchanger material [10]

The absorbed ^{110m}Ag was recovered from the graphite by using an eluting solution of 1M HNO_3 and 2% H_2O_2. The results (Figure 6.4) indicate that more than 95% of the ^{110m}Ag was recovered after one bed volume.

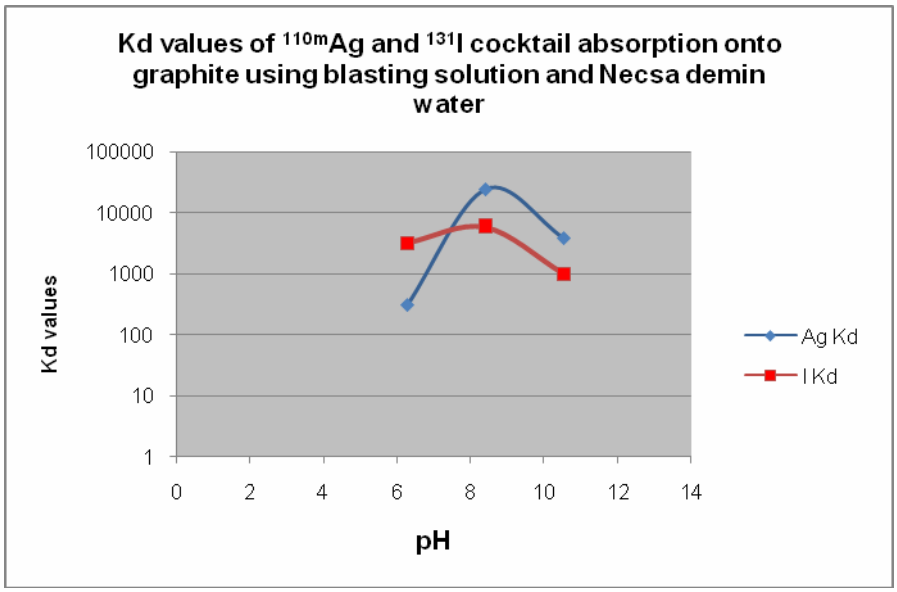

Figure 6.3: - Removal of ^{110m}Ag and ^{131}I from a PBMR waste stream using irradiated graphite as ion exchanger material [10]

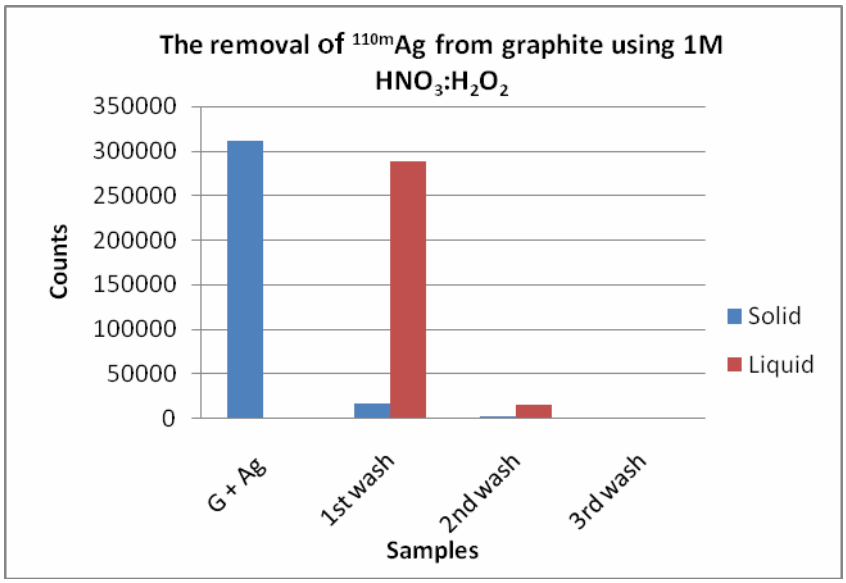

Figure 6.4: - The removal of ^{110m}Ag from graphite using 1M HNO_3 and 2% H_2O_2 [10]

In addition, the treatment of graphite with silver nitrate was undertaken to enhance the removal of radioactive iodine from waste streams. Preliminary results of experiments to determine the absorption of I-131 on the carbon nanotubes were most promising. The Kd value for I-131 on carbon nanotubes was about 50 000 units. As the nanotubes were conditioned in an acidic AgNO3 solution for 24 hours, the Kd value for Ag-110m was very low, but it had a substantial effect on the Kd value for I-131.

Preliminary results of experiments to determine the absorption of Cs-137 and Co-60 on graphite are promising [10]. Results of experiments are summarised in Table 6.1:

Table 6.1: Kd values for Cs-137 and Co-60 on graphite [10]

Sample	K_d
Cs-graphite (pH<5) treated with $AgNO_3$	5.46
Cs-graphite (pH8-9) treated with $AgNO_3$	3.99
Cs-graphite (pH>12) treated with $AgNO_3$	1.17
Cs-graphite (pH<5)	0.24
Cs-graphite (pH8-9)	0.54
Cs-graphite (pH>12)	10.19
Co-graphite (pH<5) treated with $AgNO_3$	4.2
Co-graphite (pH8-9) treated with $AgNO_3$	153.0
Co-graphite (pH>12) treated with $AgNO_3$	631.2

The results showed that Cs could not be absorbed onto graphite, but Co is absorbed at higher pH.

6.3 Use as a Gas Absorbent

In order to determine the possibility of irradiated graphite to remove radionuclides from gaseous waste streams, an experimental setup was designed. The requirements of the planned research equipment are as follows:

- High vacuum generation + measurement.
- Mobile.
- Sub-system for desorbing (gaseous) adsorbed species on PTFE filter pieces; max temperature 500ºC; desorbed species detected by FTIR.
- Sub-system for determining the absorptive capacity of graphite's towards a number of gases (I_2, CO_2, CO, NO_x, etc) at ambient temperature.

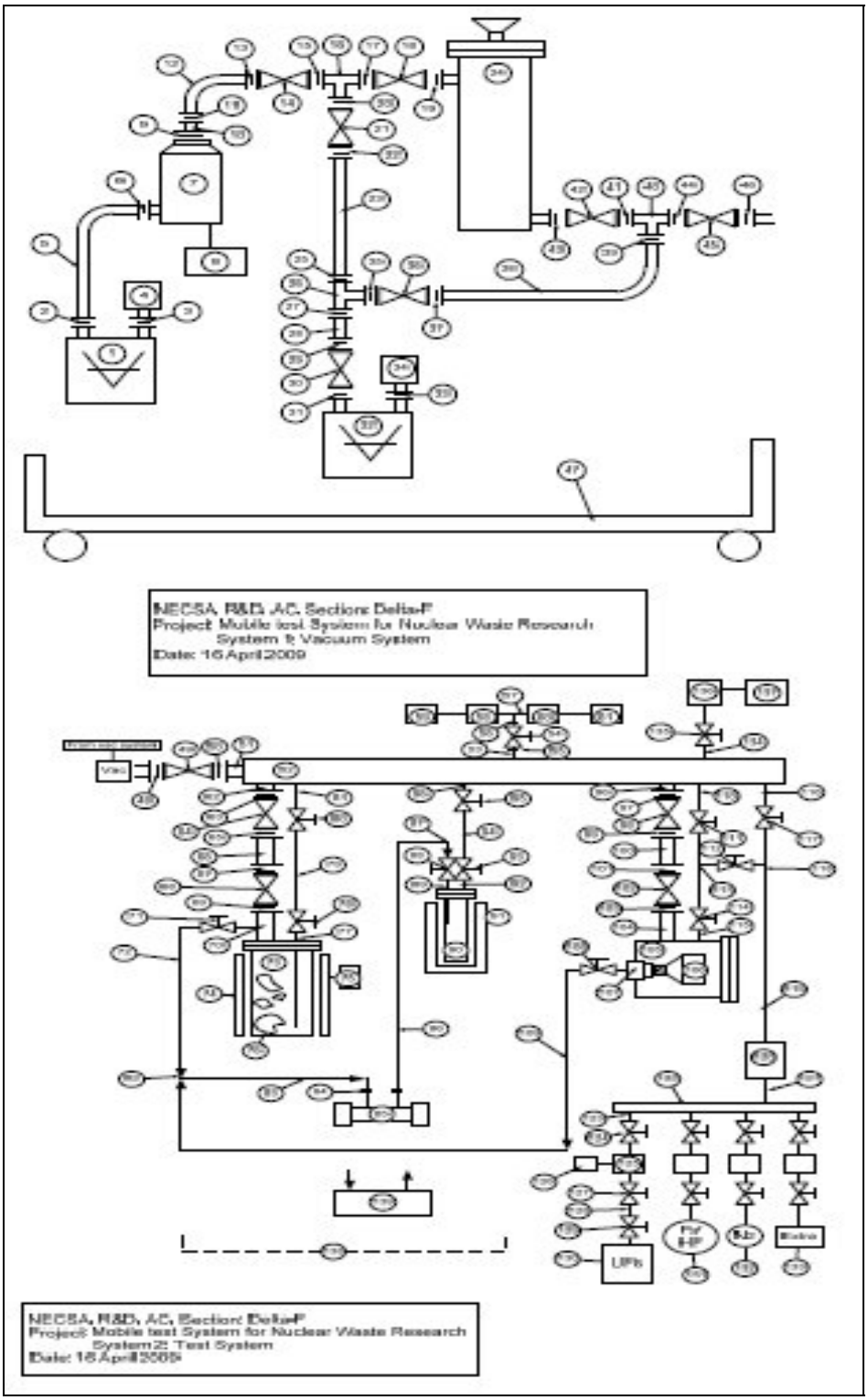

Figure 6.5: - Proposed vacuum system for gas absorption studies [11]

The proposed system is shown in Figure 6.5 and consists of the following [11]:

- Vacuum system: fore-vac pump, turbo molecular pump + backing pump, liquid nitrogen trap, trolley.
- Vacuum manifold + vacuum and pressure gauges.
- Liquid nitrogen freeze-out container + liquid nitrogen dewar.
- PTFE filter desorption vessel + heating mantle and temp control. An inert gas flush probe is included to facilitate transfer of the desorbed species to the FTIR gas cell. The vessel is connected to a standard 10 cm FTIR gas cell. Only IR active species can be detected. The gas cell is connected to the freeze-out vessel.
- Gas supply, flow control and mixing manifold. At least four supply lines, each with mass flow controller, power supply and readout unit, and isolation valves. This system allows accurate mixing and flow control of a test gas with a carrier gas.
- Test vessel for determining the absorptive capacity of graphite towards specified gases. Graphite is contained in a removable canister with screw thread for fixing it vacuum tight to the test vessel. The outlet of the vessel is connected to the FTIR gas cell. The test gas mixture, with known concentration and flow rate, flows through the graphite canister. The test gas is absorbed by the graphite until it reaches saturation. The test gas is then detected by the FTIR. The test gas concentration, flow rate, and flow time to saturation allows the calculation of the test gas absorptivity per mass unit graphite.
- The equipment to build this system is currently being sourced.

6.4 Use in Electrodes to Dissolve Spent Fuel

To date, a literature study has been completed with reference to the treatment of graphite surfaces for electrochemical technology, although no experimentation has been performed due to the unavailability of irradiated graphite as a research material (DME requested for permission to import). However, small graphite tubes were manufactured from available natural high density graphite and these were coated in order to test the electrochemistry system with these materials as electrodes. An electrochemistry setup is currently being built [11].

6.5 Use as a Crucible Material to Dissolve Spent Fuel

No progress due to the unavailability of irradiated graphite as research material (DME requested for permission to import) [11].

6.6 Use in Nanotube Technology to Immobilise Radioactive Waste

The production of CNTs from PBMR nuclear graphite using the laser ablation technique was recently demonstrated, as indicated in Figure 6.6. These structures look similar to the CNT containing spider web-like structures found in the reactor when producing CNTs with the electric arc technique. The nanotube's versatile structure allows it to be used for a variety of tasks. Research is currently underway at Necsa to functionalise these nanotubes for [11]:

1. tumour-targeting and nuclear medicine transportation inside human body;
2. removing pollutants such as organic pollutants, metals, fluorides and radionuclides;
3. absorption of expensive extractants onto nanotubes to minimise volume of liquid waste;
4. decreasing leaching of radionuclides from waste matrix;
5. encapsulation of uranium to reduce disposal risk; and

6. other applications for re-use at nuclear industries (excluding the fuel cycle),

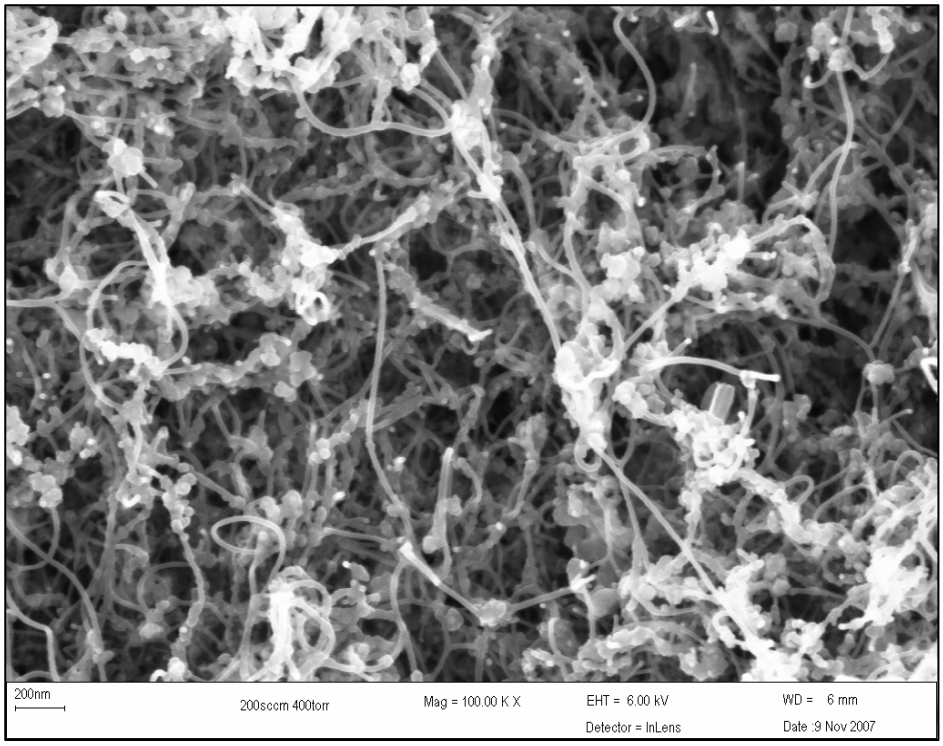

Figure 6.6: The production of CNTs from PBMR nuclear graphite, using the laser ablation technique [10]

Due to the unavailability of irradiated graphite as research material (DME requested for permission to import) to manufacture nanotubes, these were manufactured at the University of Johannesburg (UJ) instead.

After the success of iodine absorption onto Ag functionalised nanotubes (See previous progress report RC-VLG-0208), it was decided to functionalise nano tubes with phosphate groups. The aim of this functionalisation was to determine if uranium would be irreversibly absorbed on the phosphate groups, thereby decreasing the leaching probabilities when encapsulated in waste matrices. Experiments were conducted to determine K_d values of uranium on functionalised carbon nanotubes.

The results in Table 6.2 below, indicate that the uranium absorption was minimal, hence a very negative result. The UJ was contacted and a decision made to functionalise nanotubes with tri-butyl groups and to repeat the experiment. The manufacturing of nano tubes with tri-butyl groups is currently under way [11].

Table 6.2: K_d values of U on functionalized carbon nanotubes [11]

Sample	Concentration (mg/L)	Kd
Standard in 1M HNO_3	382.5	-
Standard in 1M Na_2CO_3	384.5	-
Oxidised CNTs in Na_2CO_3	Sample not clear	-
Oxidised CNTs in HNO_3	378.4	2.7
Phosphorylated CNTs in Na_2CO_3	381.2	1.5
Phosphorylated CNTs in HNO_3	364.6	8.5
Pristine MWCNTs in Na_2CO_3	378.4	3.8
Pristine MWCNTs in HNO_3	378.3	2.7

It was decided to determine the possibility of absorption of tritium and technetium. Natural nanotubes were used to determine the adsorption properties of specified nuclides. Single-walled (>75%) and double-walled (50 - 80 %) carbon nanotubes, produced by catalytic chemical vapour deposition (CVD), were sourced from a commercial supplier. Multi-walled carbon nanotubes were obtained from the University of Johannesburg where they were manufactured by means of nebulised spray pyrolysis (NSP).

The distribution co-efficients were determined for tritium, technetium and iodine. The results indicated that no tritium absorbed onto any of the CNTs at any pH. This was to be expected because of the chemical form of the tritium (as water).

For the technetium experiments Tc-99m was used as a tracer. The chemical form was assumed to be the pertechnetate ion (TcO4- (TcO4-). The distribution co-efficients were determined as per standard methods (using only about 0.02 g of the material). The results (in units of ml/g) are presented in Table 6.3 overleaf [11].

Table 6.3: Kd values of Tc-99m on carbon nanotubes [11]

	SW CNT	DW CNT	MW CNT
pH 3	1.25E+04	7.27E+03	7.98E+03
pH 7	6.55E+03	2.02E+03	5.40E+02
pH 10	7.72E+03	1.71E+03	4.94E+02

The results indicate that Tc-99m does absorb onto the nanotubes with the highest absorption being from an acidic solution onto the single-walled nanotubes [11].

7 SUMMARY

Significant work has been undertaken to date in respects to the identification and making of new products for use within the nuclear industry, and this work has been documented within this interim report. The key highlights of the work to date, includes:

- GrafTech has formed roughly 50% of the planned amount of artifacts for Task 5.2
- Artifacts produced by GrafTech have been made using proprietary 'fast' processing technology and utilize carbon black as the waste simulant.
- GrafTech have identified that artifacts made with the carbon black fraction require significantly higher amounts of pitch due to the sizing of the carbon black.
- It is critical that the links need to be maintained with WP4 to inform the work being undertaken under Task 5.2 of WP5.
- Field Assisted Combustion Synthesis (FACS) has been identified as the most appropriate method for fabricating Silicon Carbide as means of undertaking there commitments to WP5.
- Necsa have demonstrated that irradiated graphite exhibits significant potential for use as an ion exchange medium.
- Preliminary results from Necsa of experiments to determine the absorption of Cs-137 and Co-60 on graphite are promising.

A significantly more detailed report will be produced in year 4 of the Carbowaste programme which will fully report on the requirements and the results gained from the work undertaken under Task 5.2.

GLOSSARY

CNT	Carbon Nanotubes
CVD	Chemical Vapour Deposition
DME	Department of Minerals and Energy (SA)
DTA/TG	Differential Thermal Analysis and Thermal Gravimetry Analysis
FACS	Field Assisted Combustion Synthesis
FTIR	Fourier Transform Infrared Spectroscopy
i-graphite	irradiated graphite
LALLS	Low Angle Laser Light Scattering
LM/SEM	Light Microscopy / Scanning Electron Microscopy
NECSA	South African Nuclear Energy Corporation
NRG	Nuclear Research and consultancy Group
NSP	Nebulised Spray Pyrolysis
PBMR	Pebble Bed Modular Reactor
PTFE	Polytetrafluoroethylene
SEM	Scanning Electron Microscope
SHS	Self-Propagating High Temperature Synthesis
SiC	Silicon Carbide
UJ	University of Johannesburg
UV	Ultraviolet
WP4	Work Package 4
WP5	Work Package 5
XRD	X-Ray Powder Diffraction

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