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Guidelines for HTR Fuel Manufacture

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Summary

In the Work Package 4 (Fuel Fabrication) in HTR-F (Fuel Technology) funded by the European Community, the participants intend to fabricate HTR Fuel kernels and coatings.

This document is the contribution of FRAMATOME-ANP to the Work Package 4 numbered as deliverable number 18 from task 5.

It deals with UO_2 fuel and is the result of comments from WP4 participants (Commissariat à l'Energie Atomique, Joint Research Centre Institute for Transuranium Elements and Institute for Energy, Forschungszentrum Jülich and FRAMATOME ANP).

This document describes and lists :

- Overview of manufacturing processes to be used.
- Fuel Kernel Acceptance Criteria.
- Layers Acceptance Criteria.
- Fuel Coated Particles Acceptance Criteria.

This specification of the HTR Fuel has to be considered as guidelines for laboratory scale fabrication at CEA (Commissariat à l'Energie Atomique) and ITU (Institute for Transuranium Elements).

At this stage of development, this fuel manufacture specification do not deal with the characterisation methods.

Contents

List of references

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1. Introduction

This document is a preliminary fuel specification for TRISO (TRI-material ISO-tropic) coated fuel particles that will be manufactured at laboratory scale for kernels fabrication and fuel kernels coatings at CEA (Commissariat à l'Energie Atomique) and for kernels fabrication at ITU (Institute for Transuranium Elements).

In order to specify HTR fuel, this document will use previous European experience in HTR reactors: Dragon reactor (1956-1975) and the German experience with AVR (1960-1989) and THTR (1964-1989) as well as the International experience (USA : Peach Bottom (1960-1974) and Fort Saint Vrain (1964-1989), Japan : HTTR(1998-present) China : HTR-10 (2000-present).

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This specification of the HTR Fuel has to be considered as guidelines for laboratory scale fabrication at CEA (Commissariat à l'Energie Atomique) and ITU (Institute for Transuranium Elements).

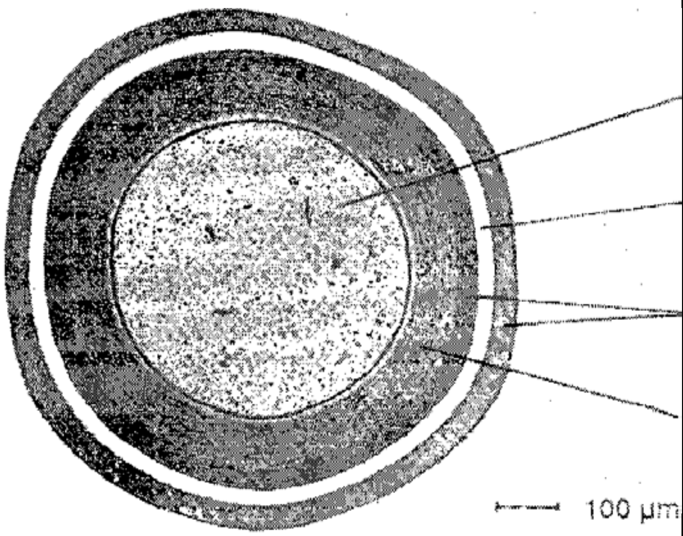
At that stage of development, this fuel manufacture specification do not deal with the characterisation methods.

2. Overview of HTR Fuel Fabrication

2.1 Fuel coated particles description

Generally, the TRISO HTR Fuel particles have a 500 µm diameter UO_2 kernel and a 200 µm thick coating, consisting of a porous buffer layer and two dense layers of pyrocarbon with a silicon carbide layer in between, named IPyC and OPyC for inner and outer pyrocarbon. While all these layers have important mechanical and chemical functions, the SiC layer guarantees the fission product retention.

The figure below gives a cross-section through an LEU TRISO coated particle :



UO_2 fuel kernel

Silicon carbide layer

Inner and outer pyrocarbon layers

Buffer layer

Cross-section through an LEU TRISO coated particle

2.2 Kernel fabrication process description

Various processes were studied to prepare fuel micro-spheres. A review of the processes is done in the task 1 of the HTR-F Work Package 4 [1].

From this work it appears the more interesting processes for oxides kernels fabrication are based on sol-gel or powder metallurgy.

The powder metallurgy route (powder agglomeration) is more adapted for big (500-2000 µm) and porous particles (80-90% theoretical density) and gives less round kernels with a higher size distribution than sol-gel processes. The sol-gel methods are also of particular interest because of the possibilities of automation and remotely controlled operations [2].

The sol-gel processes (chosen for the GT-MHR fuel) are divided in different procedures developed in the 60-70s [1,2,3] such as :

- The water extraction method (ORNL, USA).
- The nitrate extraction method (CNEN, Rome).
- The Gel Supported Precipitation (GSP) method (SNAM).
- The internal gelation with HMTA method (KEMA, Arnhem).

The more interesting sol-gel processes are the GSP and the HMTA methods.

The GSP process was used by General Atomics for ThO_2 kernels and by NUKEM for the German Fuel. Today the GSP process is used by NFI for Japanese Fuel, INET for Chinese Fuel, and is foreseen for PBMR Fuel in South-Africa.

The HMTA process was developed in the 70s for Vibrocompacted Fuel by KEMA in Netherlands and a little latter by ORNL in the USA [3]. Today it is used for the GT-MHR kernels preparation in Russia.

A description of the GSP and HMTA processes is presented on figures 1 and 2.

The different steps of the processes are:

1. Preparation of the sol (in fact the “broth” because it is not real sol-gel processes).
2. Dispersion of the sol into calibrated spherical drops.
3. Gelation of the drops maintaining the initial shape.
4. Ageing, washing and drying the spheres.
5. Heat treatment to obtain kernels.

The more difficult steps for each process are underlined in green colour on figure 2.

For the GSP process, it is mainly :

- Sphere forming step.
- Ageing and washing steps.

For the HMTA process it is mainly :

- Sol (“broth”) preparation.
- Use of an hot organic solvent for the gelation step.

The choice between the two sol-gel methods depends also on other aspects than kernel manufacture such as, for instance : global fuel cycle, means of waste treatment, cost and policy,…

At that development stage, the choice between kernel manufacture processes is not done. Evaluation is in progress.

2.3 Coating process description

Deposition is made by CVD (chemical vapor deposition).

The TRISO particles have 4 layers : buffer pyrocarbon (PyC), inner PyC, silicon carbide SiC, and outer PyC.

The coating process is made by CVD (Chemical Vapor Deposition) in a coater, in which a bed of kernels is fluidised by flowing a gas at a given flow rate. The fluidised bed is established by flowing argon, and when the process temperature is reached, a mixed gas is introduced during the needed time to form a given layer. A view of the 3 inches coater used at CEA Grenoble is given in figure 3.

The main part of a coating installation is the cylindrical device in which the fluidised bed is established. From the literature, it appears that the results are quite sensitive, for instance, to the coater shape, and mainly on :

The coater diameter which could vary from laboratory to production sizes :

- 24 mm in reference [6],
- 50.8 mm in reference [7],
- 24 to 76.2 mm first and then 241 mm in the General Atomics developments,
- 240 mm in Germany (Nukem/Hobeg) with batches of 5 kg of fuel kernels, and a project for a coater of 400 mm with batches of 10 kg [8].

The coater distributor has conic shape with gas entry. This part, associated with the gas flow rate, governs the turbulence in the fluidised bed and then the overall quality and homogeneity (figure 4). General Atomics had problems with the small coater cone, and developed a six holes distributor for the 240 mm coater.

The fluidised bed height must be relatively small, in the range of the coater diameter, for having the best homogeneity and avoid faceting of the particles.

Many other aspects of the coating process are depending on the apparatus and know-how, so that specific parameters changes slightly from one coater to another.

The figure 5 from reference [4] gives the coating steps for buffer layer, Inner PyC layer, SiC layer Outer PyC layer and the gases used at these different stages. Coating temperature are given as example.

3. Quality Requirements

3.1.1 Scope

This chapter gives a description of Quality requirements.

3.1.2 Product Quality Policy

All activities carried out to produce fuel under this specification shall be conducted in accordance with a documented Quality Assurance Program.

3.1.3 Product Quality Classification

All of the fuel products covered by this specification have been classified as being safety-related, except that the raw materials used to produce the fuel are not safety-related.

3.1.4 Quality Assurance Program

To assure that the fuel products will meet the quality requirements of these specifications and to achieve compliance with QA requirements, each organisation unit performing operations on the product shall utilise qualified personnel and controlled processes.

3.1.5 Specification Confidence Level Statements

The specifications require that the inspection procedure or sampling plan for any property apply the test for confidence statements separately to the population mean (based on the mean of the sample and its distribution) and/or the actual distribution. A confidence level statement for the value of the mean for a given property in a production unit is equivalent to a statement of the probability that any mean value measured on samples from the same parent population will be bounded by the specific interval. A confidence level applied to the population distribution for a given property defines the probability that less than the specified fraction of the population has a value outside the critical limits.

Acceptance criteria defined jointly for a mean and for a percent of the population allowable outside of the critical limits have the same stated confidence level.

A critical limit value is a value that defines an upper or lower boundary of the acceptance region and separates the rejection region from the acceptance region.

4. Manufacturing Process

The manufacturer is responsible for preparing specification for the processes.

Theses specifications shall include necessary raw material specifications.

The manufacturer shall document the design, operation, testing and maintenance of equipment use to manufacture fuel products.

The manufacturer shall prepare the quality control procedures used to verify that the fuel products satisfy the fuel product specification.

5. Kernel Fuel Particles

5.1 Scope

This chapter gives specifications for manufactured UO_2 kernels. For the laboratory scale fabrication, these specifications are to be taken as guidelines.

5.2 Substrate

This specification deals with UO_2 based kernels.

5.3 Process

The process to be used in HTR fuel manufacture is based on Sol-Gel processes described in chapter 2.2.

5.4 Kernel Acceptance Criteria

Fuel kernels shall be tested to determine conformance to specification requirements detailed in this chapter.

The requirements are listed in Table 1.

6. Fuel Particles Layers

6.1 Scope

This specification deals with TRISO (TRI-material ISO-tropic) type coated UO_2 based kernels. For the laboratory scale fabrication, these specifications regarding layers are to be taken as guidelines.

6.2 Substrate

The kernels are coated with a multi-layers coatings : buffer pyrocarbon (PyC), inner PyC, silicon carbide SiC, and outer PyC.

6.3 Process

The process to be used in HTR Fuel coating is based on CVD process described in chapter 2.3.

6.3.1 Gases and Coating Rates

Gas and coating rates for TRISO coated fuel particles shall be determined in order to have a deposition rate between 5 and 10 $\mu m.min^{-1}$.

Recent comparison at INEEL (Idaho National Engineering and Environmental Laboratory) between past German and US production have shown the importance of this parameter on the structure of PyC.

The figure 5 shows structure of PyC and BAF, True Strain versus Coating Rate.

6.3.2 Coating Continuity

Interruption during coating is not allowed.

6.3.3 Temperature

Coating temperatures are linked with density and have to be adjusted to obtained the required density.

6.4 Layers Acceptance Criteria

Layers shall be tested to determine conformance to the requirements given in Table 2.

7. Loose Coated Particles

7.1 Scope

For the laboratory scale fabrication, these specifications regarding loose coated particles are to be taken as guidelines. After coating, the loose coated particles have to be evaluate in regard of heavy metal contamination and defects.

7.2 Loose Coated Particles Acceptance Criteria

Loose coated particles shall be tested to determine conformance to the requirements given in Table 3.

Figure 1

Main difference between the GSP and the HMTA process

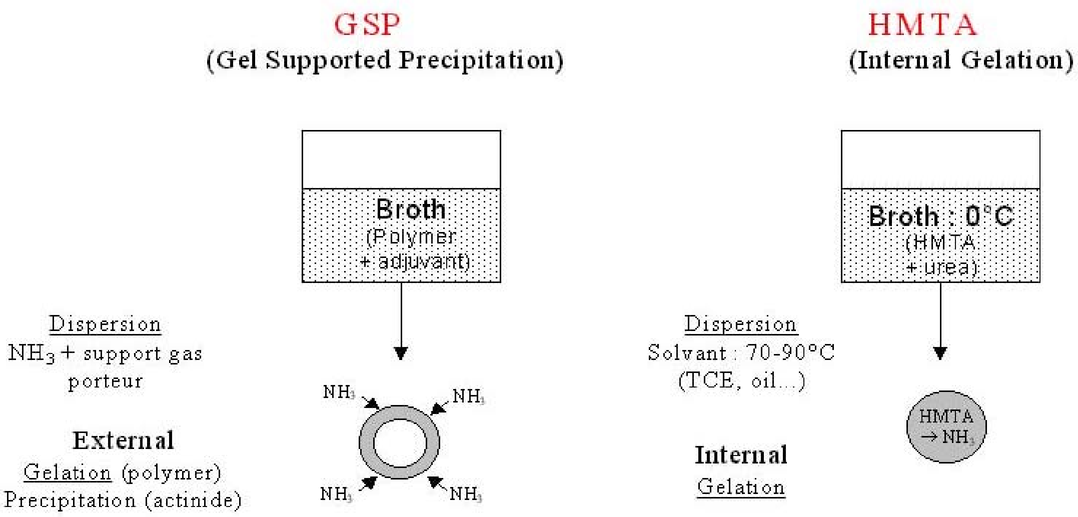


Figure 2

Main steps of the HMTA and GSP processes

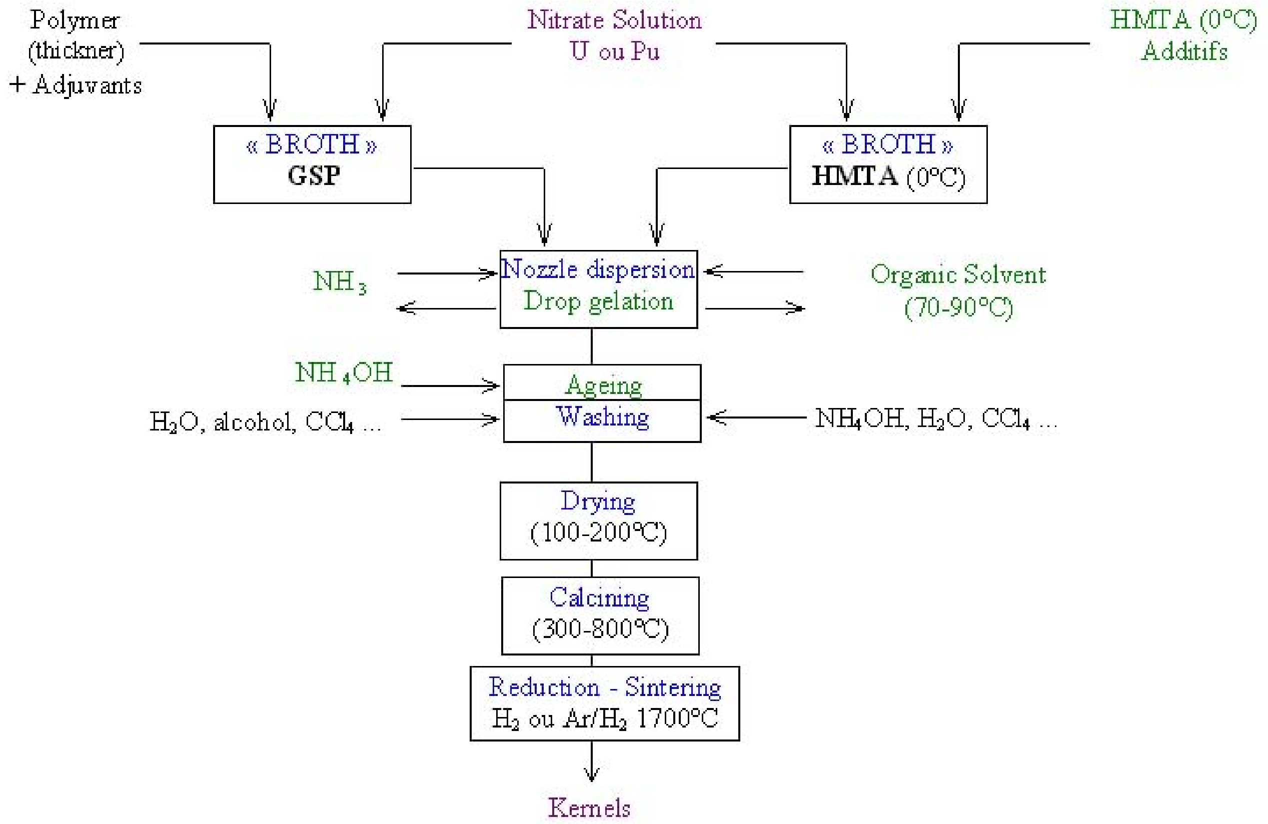
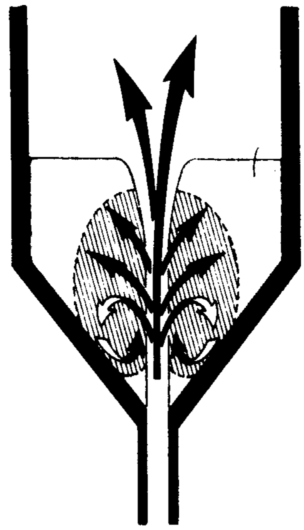


Figure 3

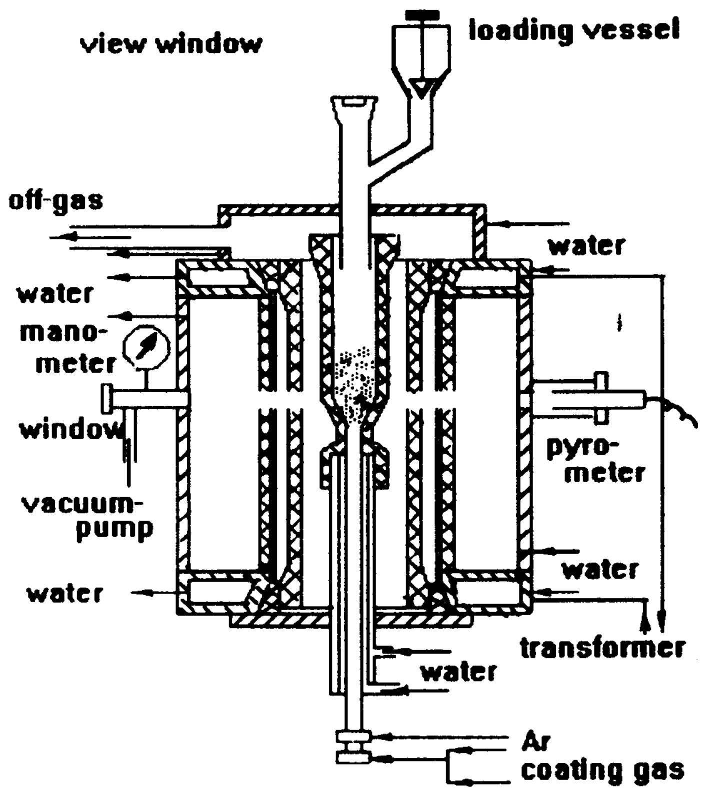
Coater 3 inches

Figure 4

Coater schematic drawing



Schematic graph of the flow pattern in a fluidized bed.



Schematic diagram of the coating.

Figure 5

Coatings Process

Coating the kernels

C_2H_2 - Ar	→	Buffer layer (1100 - 1400 °C)
C_3H_6 - Ar	→	Inner PyC layer (1350 - 1450 °C)
MTS	↘	
H_2	→	SiC layer (1500 - 1570 °C)
Ar	↗	
C_3H_6 - Ar	→	Outer PyC layer (1350 - 1450 °C)
		Coated fuel particles
		Classification
		Inspection

Figure 6

Coating rates

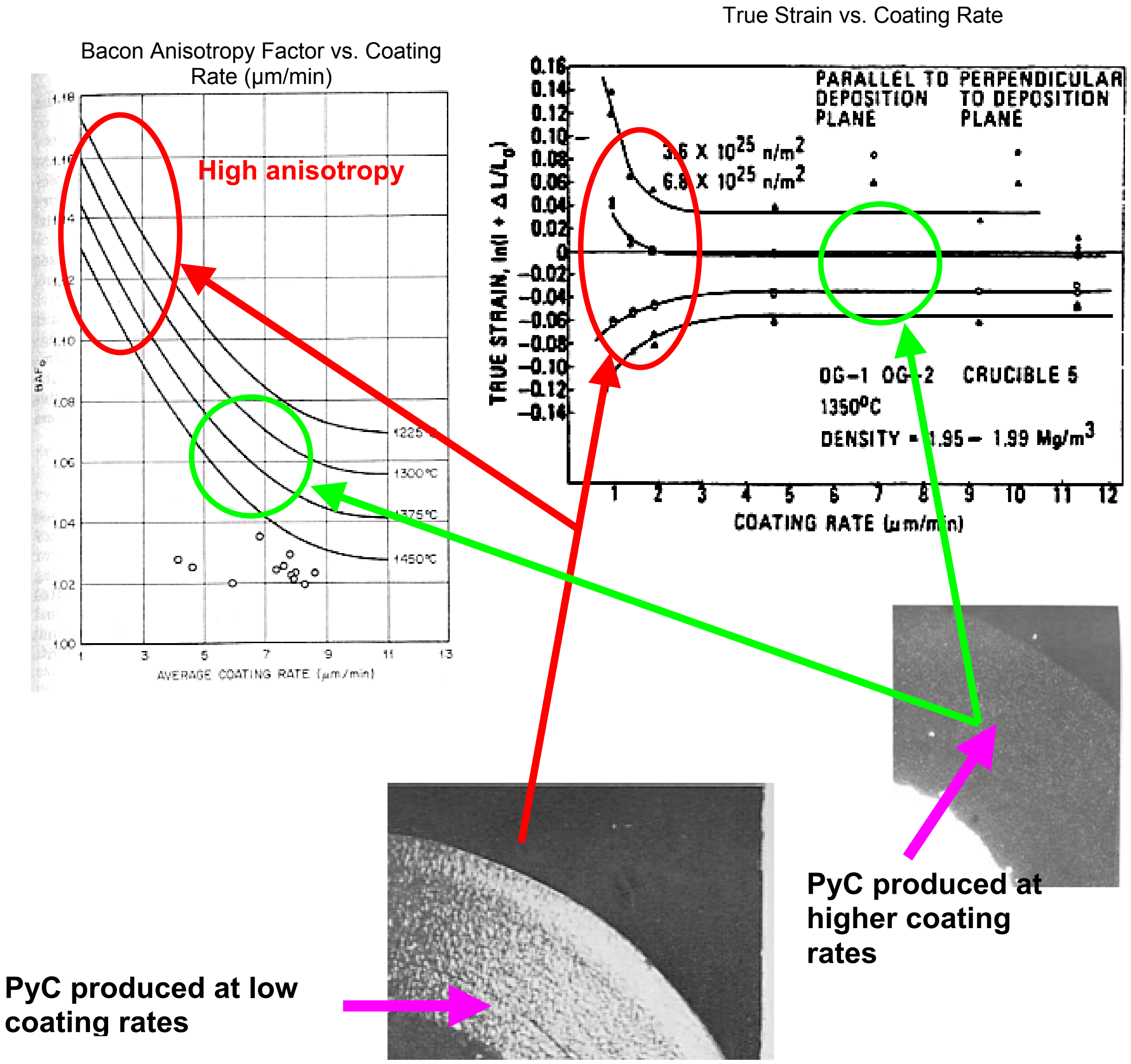


Table 1

Kernel Fuel Particles Acceptance Criteria

Parameter	**Specification**
Composition	UO_2 UO_x, 1.99 ≤ x ≤ 2.02
U235 Enrichment and isotopic composition	$\leq 19.75\%\,^{+0}_{-0.1}$
Mean Diameter	500 µm ± 40 µm
Density	≥ 10.4 (≥ 95% d_{th})
Sphericity (D_{max}/D_{min})	<1.1
Impurity content	In conformity with ASTM C 776
Moisture content	In conformity with ASTM C 776
Equivalent Boron content	In conformity with ASTM C 776 and C 1233

Table 2

Carbon Layers Acceptance Criteria

Carbon Buffer

Parameter	**Specification**
Thickness	95 μm ± 20 μm
Density	≤ 1.05

Inner and Outer Pyrolitic Layers

Parameter	**Specification**
Thickness	40 μm ± 10 μm
Density	$1.9\,^{+0.1}_{-0.05}$
Anisotropy (BAF)	BAF ≤ 1.06

SiC Layer

Parameter	**Specification**
Thickness	35 μm ± 7 μm
Density	≥ 3.18
SiC crystal structure	100% beta (cubic)

Table 3

Loose Coated Particles Acceptance Criteria

Parameter	Specification
Heavy metal contamination	$\leq 10^{-7}$
Defective coating	$\leq 5.10^{-6}$