Journal of Nuclear Materials 402 (2010) 136–146

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

The influence of annealing temperature on the strength of TRISO coated particles

I.J. van Rooyen [a,b,*], J.H. Neethling [b], P.M. van Rooyen [a]

[a] Pebble Bed Modular Reactor (Pty) Ltd., 1279 Mike Crawford Avenue, Centurion, South Africa
[b] Department of Physics, Nelson Mandela Metropolitan University, Port Elizabeth, South Africa

ARTICLE INFO

Article history:
Received 17 March 2010
Accepted 10 May 2010

ABSTRACT

The integrity of the Pebble Bed Modular Reactor (PBMR) fuel, and specifically the SiC layer system of the Tristructural Isotropic (TRISO) coated particle (CP), namely inner pyrolytic carbon, silicon carbide and outer pyrolytic carbon (I-PyC–SiC–O-PyC), determines the containment of fission products. The PBMR fuel consists of TRISO coated particles (CPs) embedded in a graphite matrix. One of the characterization techniques investigated by PBMR is the determination of strength of CPs. It is a well known metallurgical fact that temperature, amongst many other parameters, may influence the strength of a material. A recently developed method for measuring the strength of the TRISO coated particles was used and is briefly described in this article. The advantages of this method are demonstrated by the comparison of strength measurements of five experimental PBMR CP batches as a function of annealing temperature. Significant modification of strength after annealing was measured with increased temperature within the range 1000–2100 °C. The interesting feature of decreasing standard deviation of the strength with increasing temperature will also be discussed with a possible explanation. A significant difference in coated particle strength is also demonstrated for two CP batches with layer thickness on the extremities of the SiC layer thickness specification. The effect of long duration annealing on these strength values will also be demonstrated by comparing results from 1 h to 100 h annealing periods of coated particles at a temperature of 1600 °C.

1. Introduction

The Tristructural Isotropic (TRISO) coated particle (CP) is the first level of containment for the fission products in a Pebble Bed Gas Reactor with the nominal 35 μm thick SiC layer (Fig. 1) as the main fission product transport barrier [1]. The TRISO coated particle needs therefore to withstand the internal pressures due to fission and transmutation products released during irradiation. The SiC strength is further also the main parameter used in performance modelling of the High Temperature Reactor (HTR) fuel and Nabielek emphasizes that no strength measurements were done on the specific SiC of the high performance fuel which formed the conceptual basis of the modern HTRs [2]. The strength determination of the SiC layer of a coated particle poses unique challenges as to ensure that the test samples are representative of the coated particle's material properties and the stress patterns.

Bongartz et al. [3,4] developed one of the first methods, namely the brittle ring test which could be used to determine the strength and Young's modulus of the specific SiC layer of a coated particle. These tests consist of crucial sample preparation techniques that can possibly induced defects during sample preparation, which may influence the strength values. Various researchers [5–7] performed and analyzed crushing tests on whole coated particles as it holds the advantage that no sample preparation needs to be done. They found that the crushing test was still dependent on the particle radius and that crushing tests can be useful for the measurement of the Young's modulus of particle outer coatings. Snead et al. [8] summarized the Weibull modulus and characteristic strength of β-SiC that show a large spread of results within the same type of test. The observation was also made that the internal pressurization tests showed approximately 4 times lower strength values. These different values and methods resulted in the establishment of a detailed experimental programme for the identification and development of a suitable and reliable strength measurement for the PBMR fuel coated particle in cooperation with our technology partners.

This article describes the recent results of the fracture strength of five experimental batches of PBMR coated particle batches after annealing at very high temperatures. The fracture strength was determined by the recently developed method by Van Rooyen et al. [9] by compressing the CPs between soft anvils. Finite Element Analysis (FEA) modelling was used to quantify the stress to which the particle was subjected [9]. The results by Van Rooyen et al. [9] showed that the removal of the outer layer of PyC by decarburization, still gave results comparable with the results obtained with the O-PyC layer intact. Therefore it was decided to do

* Corresponding author at: Pebble Bed Modular Reactor (Pty) Ltd., 1279 Mike Crawford Avenue, Centurion, South Africa. Tel.: +27 12 641158.
E-mail address: Isabel.vanrooyen@pbmr.co.za (I.J. van Rooyen).

0022-3115/$ - see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2010.05.009

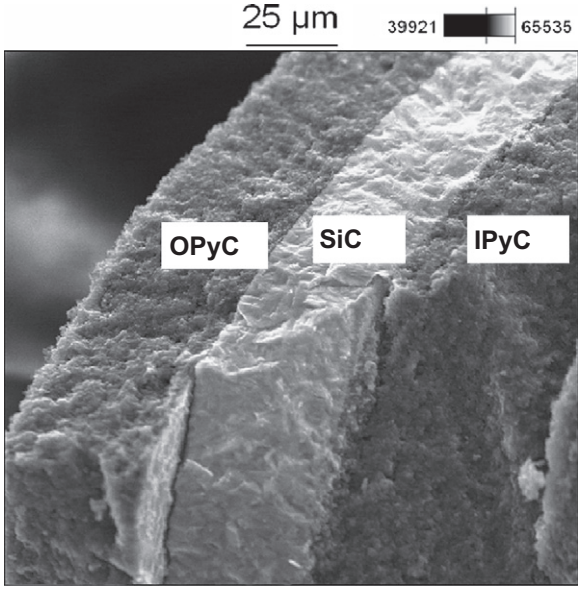

Fig. 1. SEM image from a reference PBMR coated particle of batch G118 showing the SiC layer sandwiched between the I-PyC and O-PyC.

the strength measurements for this study with the O-PyC layer intact, in other words, the CP with all three layers intact was measured, eliminating artificial contributions due to sample preparation.

The main purpose of this work is to study the effect of high temperature annealing on the CP strength as it will give an indication of the behaviour during elevated temperature conditions. This article covers mainly the annealing temperature range from 1600 °C to 2100 °C as Snead et al. [8] found that no significant strength degradation of CVD SiC occurs for temperatures up to 1500 °C. Previous studies also indicated that phase transitions from β- to α-SiC and decomposition may occur above 1600 °C which also motivates this work to start with 1600 °C as the lower temperature [11–14]. This work was undertaken to test the design parameters of the CPs for application in very High Temperature Reactors, therefore the expansion of the temperature to 2100 °C. Furthermore, it is important to note that the fuel sphere manufacturing process consists of a sinter operation at 1950 °C, and therefore it is important to know how this manufacturing process step may influence the inherent strength properties of the CPs even before use in the reactor.

2. Material and methods

The five experimental CP batches used for this study were produced by PBMR Fuel Development Laboratories at Nuclear Energy Corporation of South Africa (NECSA) during the period between 2002 and 2007. These five batches were chosen as it gave a collection of varied manufacturing conditions as well as being manufactured in two different Chemical Vapour Deposition (CVD) coaters namely the Research Coater Facility (RCF) and the Advance Coater Facility (ACF) with a capacity of 1 kg and 5 kg respectively. The RCF is a smaller coater to enable specific research and development work to be undertaken. The two coaters having a different volume will serve to identify a possible feed gas volume effect on the strength of the CPs. Due to the smaller reactor volume of the RCF coater, the heat transfer properties may be more effective which may assist layer growth and enhance layer properties. Although the main purpose of this article is to evaluate the influence of annealing temperature on the strength of the experimental batches, selected manufacturing data are given in Table 1 and preliminary observations are made. It is noted that the SiC deposition rates for the two ACF batches (0.23 and 0.24 μm/min) are 41% higher than those of the two RCF batches (0.17 μm/min).

The five experimental batches were characterized by Transmission Electron Microscopy (TEM) examination and strength measurement prior and after the high temperature annealing. The annealing was performed at two facilities namely the PBMR Fuel Development Laboratories (FDL) at NECSA and the Physics department of Nelson Mandela Metropolitan University (NMMU). The furnace types were a "PVA COV 361 TePla" vacuum furnace manufactured by ECOM Company and a Webb 89 vacuum furnace supplied by R.D. Webb Company, USA respectively. Approximately 3000 TRISO coated particles from the five batches each were annealed in ceramic crucibles (volume 3 cm^3) for the relevant holding times at temperatures ranging from 1000 °C to 2100 °C, as shown in the Research Project Plan flow diagram Fig. 2.

3. Fracture strength statistical analysis

The fracture strength values measured on the samples were analyzed using the Weibull statistical behaviour reported by various researchers [4,7,10,15]. The Weibull cumulative probability density function of the measured fracture strength values is given by:

$$F(\sigma_F) = 1 - e^{-\left[\frac{\sigma_F}{\sigma_0}\right]} \quad (1)$$

where σ_F is the fracture strength values of a sample, σ_0 is the characteristic strength value of the measured strength values of the sample and m is the Weibull modulus of the measured strength values.

Eq. (1) can be transformed to:

$$\ln\left[\ln\left[\frac{1}{1-F(\sigma_F)}\right]\right] = m * \ln[\sigma_F] - m * \ln[\sigma_0] \quad (2)$$

Eq. (2) is in the format of a straight line as given in the following equation.

$$y(x) = ax + b \quad (3)$$

Table 1
Selected manufacturing data of the five experimental PBMR coated particle batches.

Batch	SiC deposition temperature (°C)	SiC deposition rate (μm/min)	SiC characteristic strength (MPa)	Weibull modulus, m	SiC thickness (μm)	SiC density (g cm^{-3})
G118 (ACF)	1510[b]	[a]	999	4.33	32	3.19
G146 (ACF)	1450	0.23	926	2.61	39	3.2
G169 (ACF)	1510	0.24	1119	5.09	32	3.2
B14 (RCF)	1585	0.17	1507	2.15	30	3.19
B10 (RCF)	1510	0.17	1718	5.03	30	3.09

[a] Not available.
[b] Not confirmed (part of ACF commissioning tests) and not used in graphs.

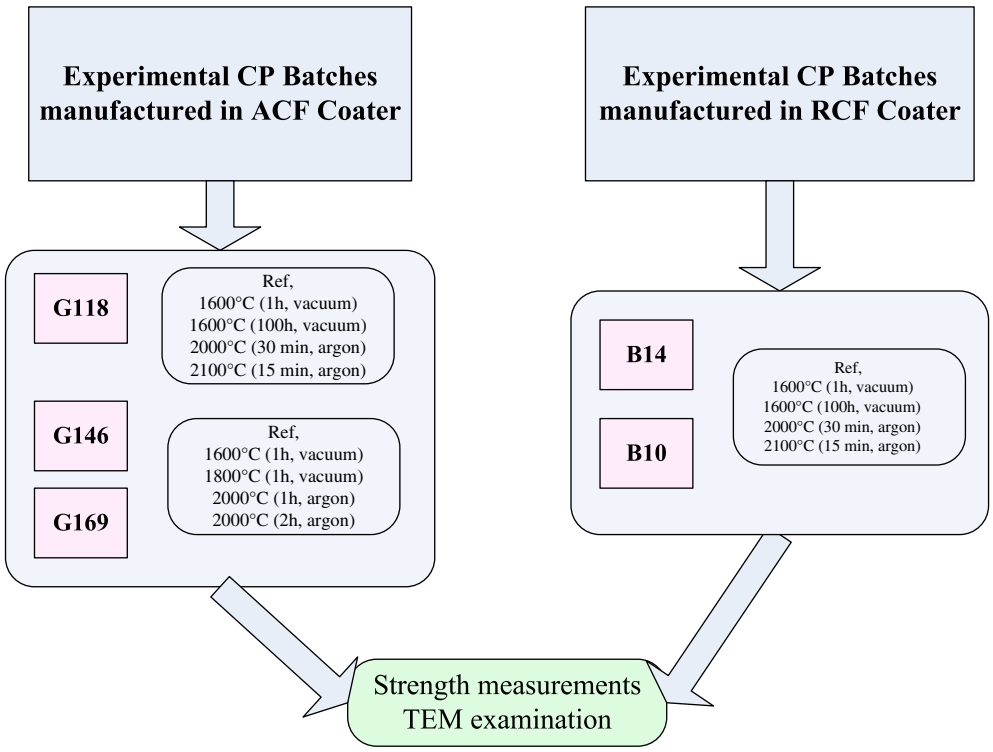

Fig. 2. Experimental research plan.

Table 2
Weibull statistics of the fracture strength determined by compression tests of the five experimental PBMR coated particle batches.

Sample description	Temperature	Number of tests	Average fracture strength (MPa)	Characteristic strength (MPa)	Weibull modulus, *m*
SiC-G118 (ACF)	Reference	30	915	999	4.33
	1600 °C (1 h)	30	1090	1191	4.35
	1600 °C (100 h)	30	1240	1336	5.62
	2000 °C (1 h)	29	1140	1214	6.85
	2100 °C	7	1180	1230	7.34
SiC-G146 (ACF)	Reference	102	820	926	2.61
	1600 °C (1 h)	25	860	987	1.86
	1800 °C (1 h)	31	1190	1318	1.35
	2000 °C (2 h)	100	1060	1245	1.31
SiC-G169 (ACF)	Reference	102	1030	1119	5.09
	1600 °C (1 h)	31	1160	1268	4.56
	1800 °C (1 h)	31	1490	1638	4.79
	2000 °C (1 h)	31	1240	1323	7.28
SiC-B14 (RCF)	Reference	31	1340	1507	2.15
	1600 °C (1 h)	31	1550	1644	7.59
	1600 °C (100 h)	31	1530	1653	5.28
	2100 °C	22	1120	1185	7.77
SiC-B10 (RCF)	Reference	31	1570	1718	5.03
	1600 °C (1 h)	31	1520	1586	10.93
	1600 °C (100 h)	31	1590	1682	8.46
	2100 °C	20	980	1079	3.34

By plotting the data using Eq. (2) and doing a least squares fit on the plotted data, the values for a and b in Eq. (3) are determined.

where a is the value of the Weibull modulus m, and the characteristic strength σ_0 is calculated using the following equation.

$$\sigma_0 = e^{-\left[\frac{b}{m}\right]} \tag{4}$$

4. Results and discussion

The Weibull statistics of the fracture strength determined from the compression tests for the experimental PBMR coated particle batches are summarized in Table 2 and are graphically shown in Figs. 3–9. By comparing the Weibull statistics plot of the various batches in Fig. 3, no specific trend is observed to differentiate between the CPs manufactured in the RCF and ACF coaters in the unannealed condition. However, the Weibull statistics plot of the fracture strength after annealing at 1600 °C (Fig. 4) shows an interesting feature where the Weibull modulus values (7.59 and 10.93) for the RCF batches have higher values if compared with those of the ACF batches (1.86, 4.35 and 4.56). The characteristic strength values also follow this same trend namely 1644 MPa and 1586 MPa for the RCF batches and have higher values than the 987 MPa, 1191 MPa and 1268 MPa for the ACF batches.

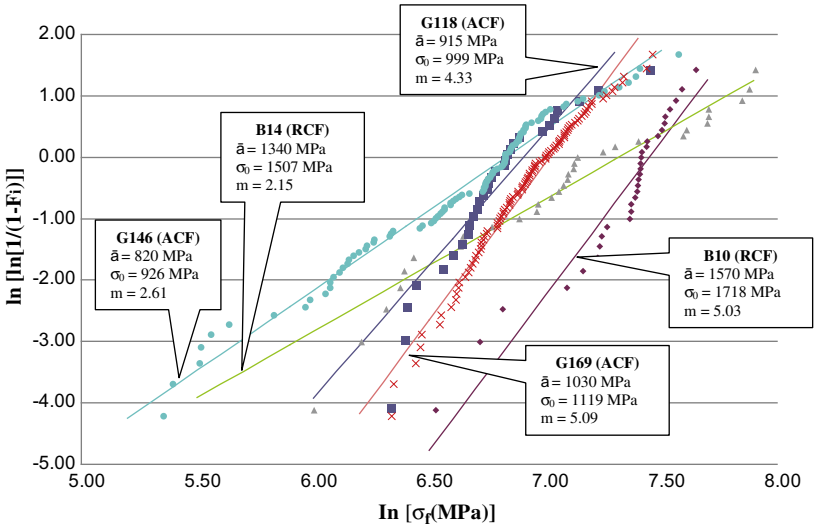

Fig. 3. Comparison of fracture stress data for experimental PBMR TRISO coated particles as manufactured ($\bar{a}$ = average strength, σ_0 = characteristic strength and m = Weibull modulus).

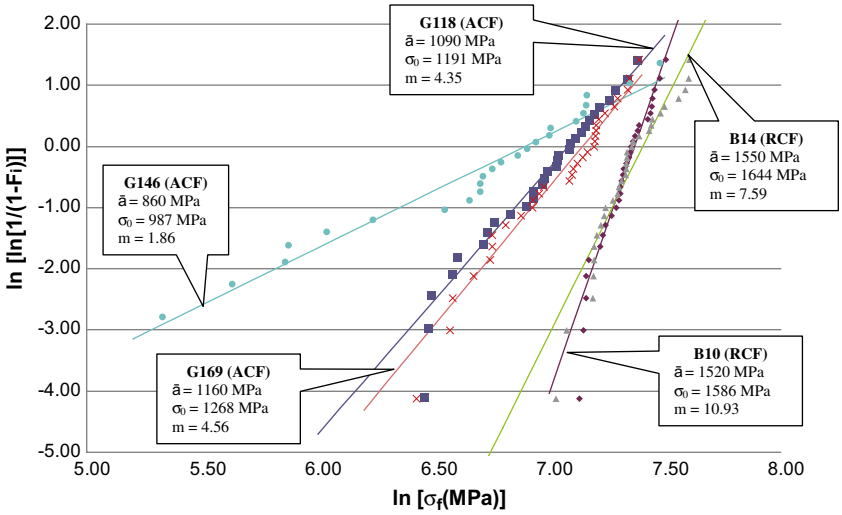

Fig. 4. Comparison of fracture stress data for experimental PBMR TRISO coated particles after a 1 h annealing at 1600 °C ($\bar{a}$ = average strength, σ_0 = characteristic strength and m = Weibull modulus).

It should further be noted that the values of the Weibull modulus for the RCF batches (7.59 and 10.93) after annealing at 1600 °C show a significant increase in the Weibull modulus values of 2.15 and 5.03 for the reference samples of batches B14 and B10. The increase in the Weibull modulus after annealing at 1600 °C is an indication of a decrease in the spread of the fracture strengths of the coated particles.

The temperature dependence of the Weibull statistics for the RCF batches, B14 and B10, is shown in Figs. 5 and 6. The graphs show the same trend in temperature behaviour namely an initial slight displacement to higher values, of the straight line after annealing at 1600 °C suggesting a slight toughening of the particles [9,15]. It also shows an increase in the modulus values as described above. However, the Weibull graph of the B10 RCF batch is displaced to lower values after annealing at 2100 °C with a significant decrease in the characteristic strength and a decrease in the Weibull modulus. The Weibull graph shows a significant decrease in the characteristic strength for values obtained for annealing at 1600 °C and 2100 °C but the modulus remained approximately similar.

The temperature dependence of the ACF batches is shown in Figs. 7 and 8. It is shown that for the G118 batch the Weibull modulus and characteristic strength increased from 4.33 to 7.34 and 999 MPa to 1230 MPa respectively with increasing annealing temperatures of 1600 °C, 2000 °C and 2100 °C. (It must be noted that the data for the 2100 °C annealing was not sufficient for statistical analysis, but that the increasing trend was suggested.) The Weibull graphs of G169 also show this same trend of increased modulus and characteristic strength with increased annealing temperature. However, the results of batch G146 show no significant increase in modulus, only an increase in characteristic strength. Another interesting trend is that batch G146 shows the lowest Weibull modulus (1.86) of the five batches evaluated after annealing at 1600 °C. It is interesting to note that this batch also shows a deviation from the characteristic Weibull straight line after annealing at 2000 °C with two distinct value populations as can be seen in Fig. 7.

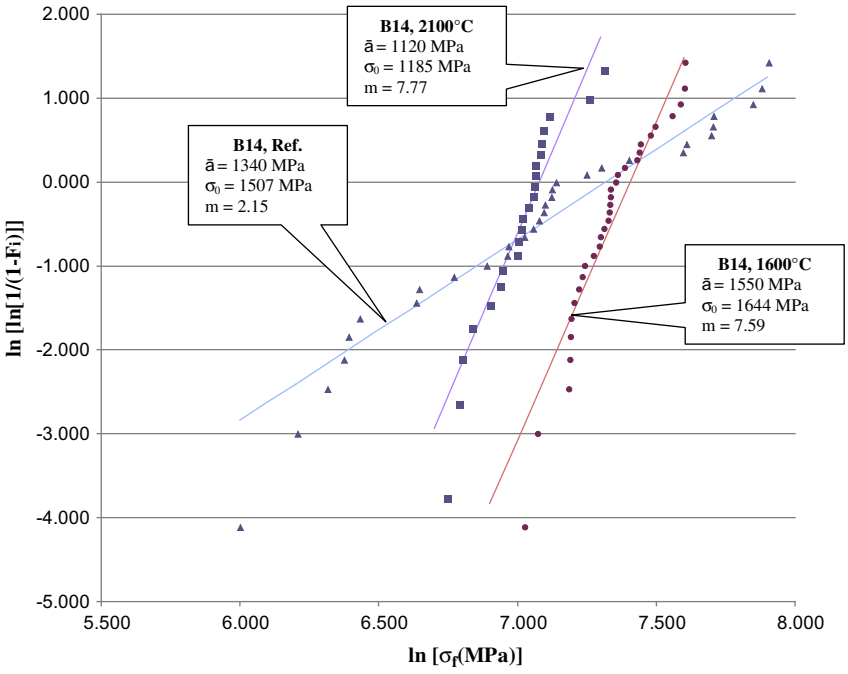

Fig. 5. Comparison of fracture stress data for the B14 experimental PBMR TRISO coated particles manufactured in the RCF coater as a function of annealing temperature ($\bar{a}$ = average strength, σ_0 = characteristic strength and m = Weibull modulus).

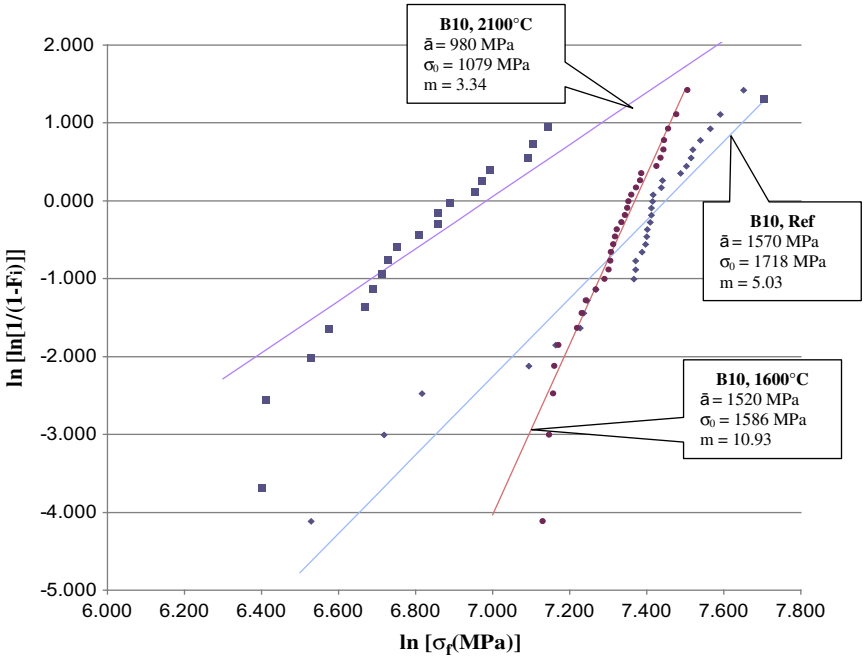

Fig. 6. Comparison of fracture stress data for the B10 experimental PBMR TRISO coated particles manufactured in the RCF coater as a function of annealing temperature ($\bar{a}$ = average strength, σ_0 = characteristic strength and m = Weibull modulus).

The strength-temperature dependence comparison between the batches manufactured in the two different coaters is also presented in Fig. 9. From this presentation, it can be seen that the RCF coater batches exhibits higher characteristic strength if compared to the ACF batches. It further also shows the tendency described previously that the characteristic strength increased for the three ACF batches and the one B14 RCF batch after the 1600 °C annealing. The reason for the reverse trend for the B10 RCF batch is not clear at this stage. It is however important to note that the characteristic strength values of the two RCF batches are not significantly different after 1600 °C annealing. This may suggest that the stresses were reduced after annealing irrespective of the initial characteristic strength.

Kim et al. [17] found in their study that the deposition rate tends to increase with increasing coating temperature. The results of this study suggest that the dependence of deposition temperature and deposition rate is not clear due to the limited data points as presented in Fig. 10. It should also be taken into account that the

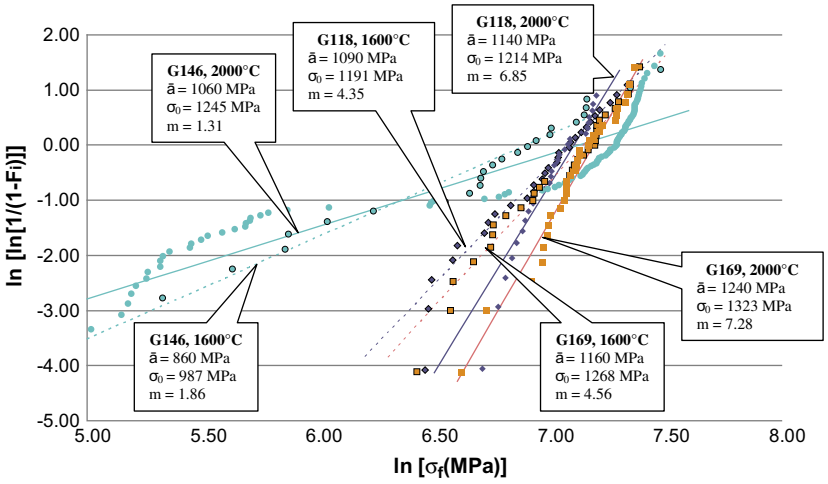

Fig. 7. Comparison of fracture stress data for the three experimental PBMR TRISO coated particles manufactured in the ACF after 1 h annealing at 1600 °C and 2000 °C ($\bar{a}$ = average strength, σ_0 = characteristic strength and m = Weibull modulus).

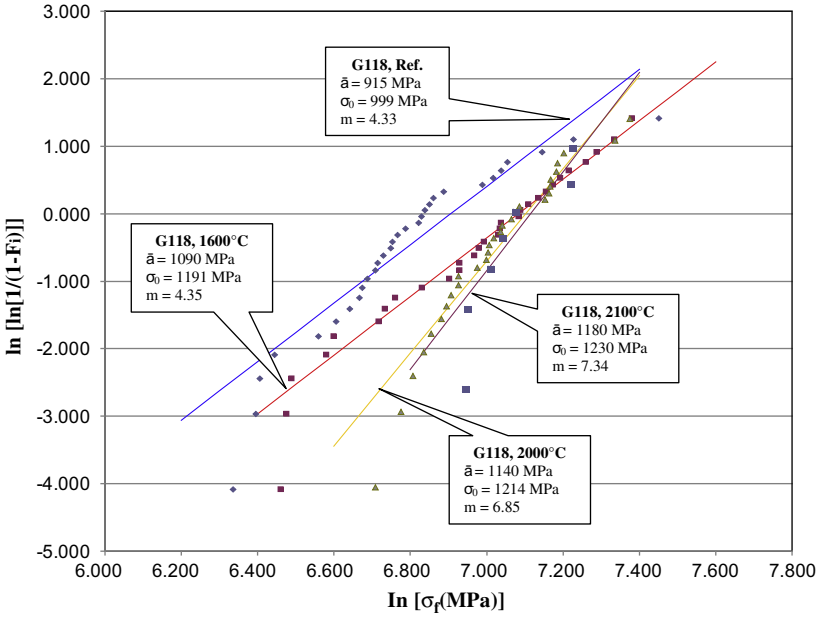

Fig. 8. Comparison of fracture stress data for the G118 experimental PBMR TRISO coated particles manufactured in the ACF coater as a function of annealing temperature ($\bar{a}$ = average strength, σ_0 = characteristic strength and m = Weibull modulus).

MTS flow rate between the RCF and AFM was different and it is therefore recommended that this work be expanded to create a representative statistical population to study. Fig. 10 however shows that the deposition rate for the RCF batches are lower than those of the ACF batches which may be the reason for the higher characteristic strength of the B10 RCF batch if compared with the G169 ACF batch as the deposition temperature for both these batches is 1510 °C.

The difference in the initial as-manufactured characteristic strength of the two RCF batches can most probably be attributed to the higher deposition temperature of B14 (1585 °C) if compared to those of B10 (1510 °C) (Fig. 11). The influence of deposition temperature on the modulus and characteristic strength was reported by Kim et al. [17] who indicated that with increased SiC deposition temperature, the modulus and characteristic strength increase. However, Xu et al. [18] found that the strength of SiC was a maximum for temperatures between 1500–1550 °C and that it decreased significantly for deposition temperatures below and above this temperature range. It is clear from Figs. 11 and 12 that the highest characteristic strength and modulus for both batches are reached at a deposition temperature of 1510 °C. This observation confirms the findings of Xu et al. [18].

The manufacturing quality measurements control data showed that the density measurements from all five batches are very similar namely from 3.19 to 3.20 g/cm^3. The only noted difference in these manufacturing quality data is the thicker SiC layer (39 μm) of batch G146 in comparison with the other four batches (30–32 μm) but all these values comply with the average specification value of 35 μm (25 μm < t < 45 μm) and is not considered as a contributor to the lowest modulus value. It is further noted that the I-PyC thickness (72 μm) of this G146 batch is the thickest compared with the other batches but the density of the I-PyC is slightly below the specification (1.8–2.0 g/cm^3) and the lowest (1.67 g/cm^3) of the other ACF batches. The low Weibull modulus and slightly lower

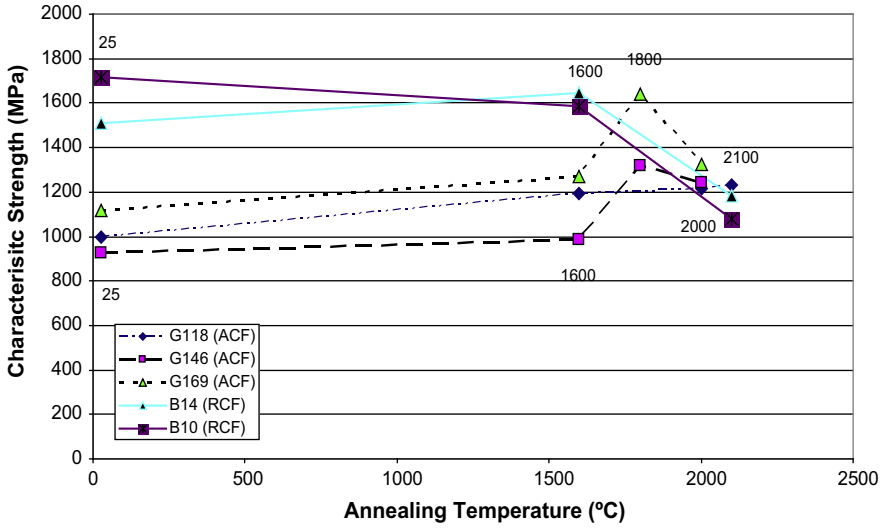

Fig. 9. Comparative trends to demonstrate the influence of annealing temperature and coater facility on the characteristic strength of the five experimental batches.

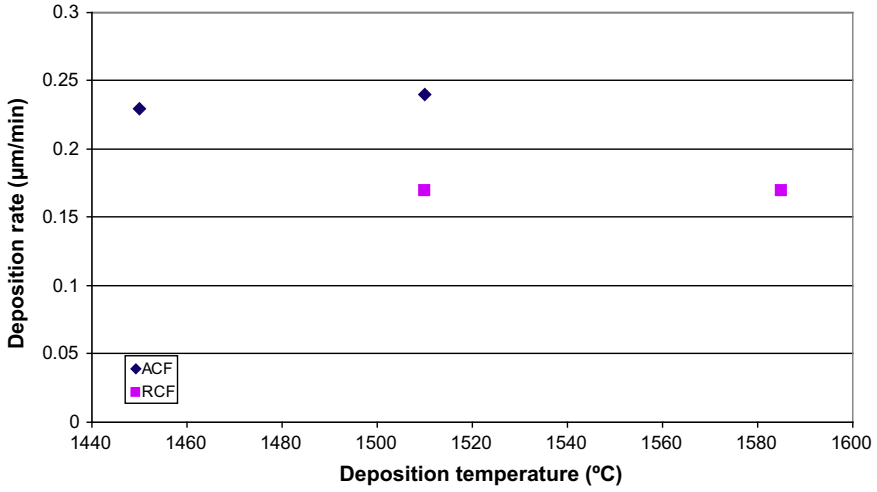

Fig. 10. Deposition rate of the SiC layer as a function of coating temperature.

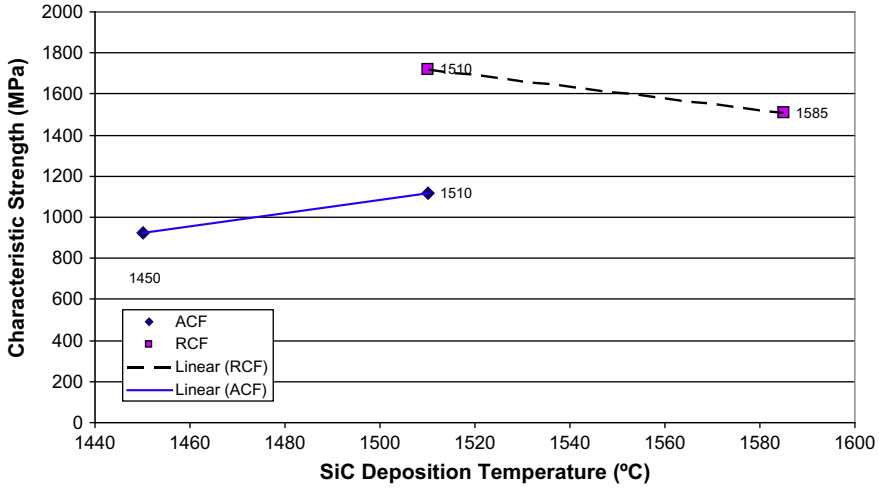

Fig. 11. Influence of SiC deposition temperature and coater facility on the characteristic strength (as manufactured) for the batches manufactured in the two different coaters (G146 (ACF), G169 (ACF); B14 (RCF) and B10 (RCF)).

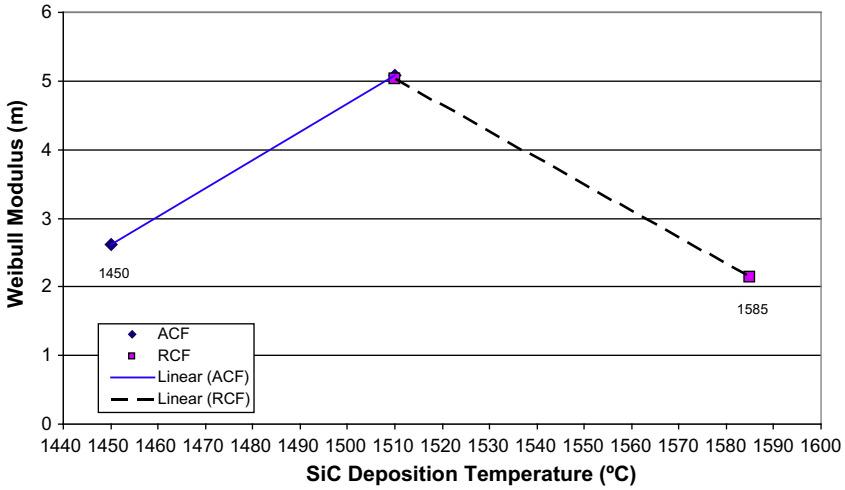

Fig. 12. Influence of SiC deposition temperature and coater facility on the Weibull modulus, *m*, (as manufactured) for the batches manufactured in the two different coater (G146 (ACF), G169 (ACF); B14 (RCF) and B10 (RCF)).

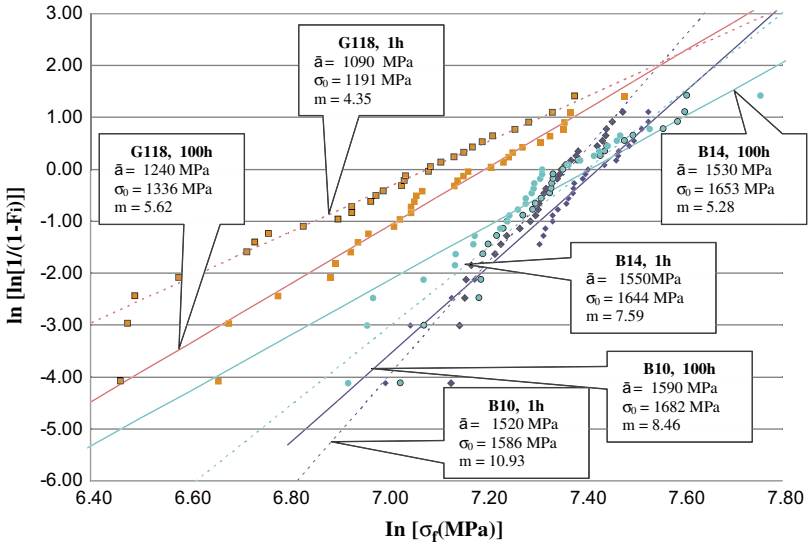

Fig. 13. Comparison of fracture stress data for the G118 (ACF); B14 (RCF) and B10 (RCF) experimental PBMR TRISO coated particles as a function of time at the annealing temperature of 1600 °C ($\bar{a}$ = average strength, σ_0 = characteristic strength and m = Weibull modulus).

I-PyC density of this G146 batch may be attributed to the significant amount of porosity in the I-PyC and SiC which was observed during the TEM analysis as shown in Fig. 14. The high amount of porosity in the I-PyC results most probably in an irregular I-PyC–SiC interface that causes stress concentrations and in its turn cause fracture initiating points.

The modulus range for this work is 2.2–5.1 and is lower than the modulus values of 4–6 proposed by Snead et al. [8]. However, if only the strength values of batches G118, G169 and B10 are considered, the modulus range are 4.3–5.1 which corresponds to the modulus values proposed by Snead et al. [8]. It is however important to note that Miller et al. [12] used a SiC modulus of 6 for CP performance modelling and the modulus of all these experimental batches are lower than 6. It is also important to note that the test methods are different and a direct comparison may not be accurate. Snead et al. [8] also reported that no significant strength degradation of CVD SiC occurs for temperatures up to 1500 °C and further that an increase is noted for temperatures above 1100 °C. Results of this study generally support this observation but in addition this work shows an increase in strength up to 2000 °C with a decrease in strength after annealing at 2100 °C.

The Weibull graph for the strength after long duration annealing of 100 h is presented in Fig. 13. The results of the two RCF batches show no significant change in the characteristic strength values although the modulus decreases from 7.59 to 5.28 and from 10.93 to 8.46 for batches B14 and B10 respectively, after long duration annealing. Opposed to this observation, the G118 ACF batch shows a slight increase in the characteristic strength from 1191 MPa to 1336 MPa and a slight increase in the modulus value from 4.35 to 5.62.

The results of this study are compared with results obtained from other types of strength measurements and are summarized in Table 3. For the purpose of Table 3, only the strength statistics of PBMR coated particles prior to annealing are compared because no indications are given that for results reported by other researchers, the samples have been annealed prior to testing. The characteristic strength of this work compares favourably with the other types of compression tests although the modulus is lower. The

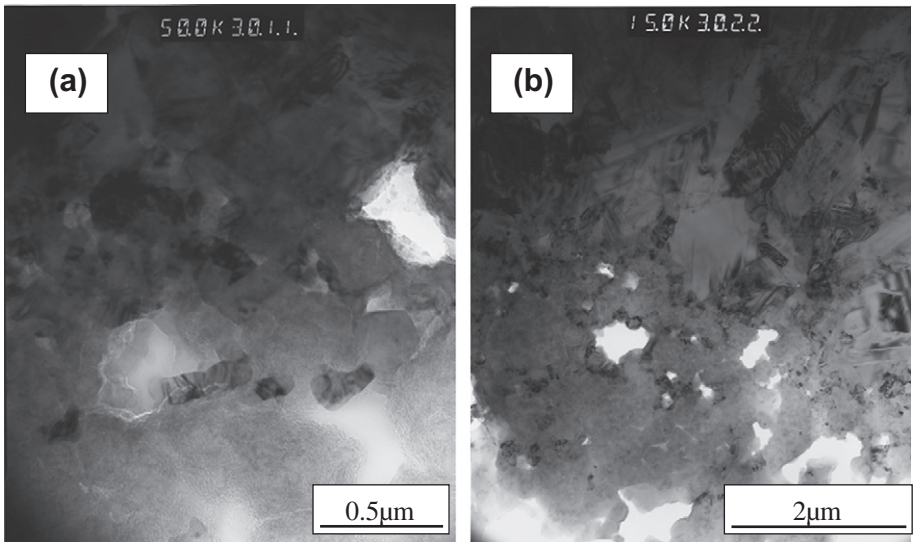

Fig. 14. Bright-field TEM images of batch G146 where (a) is the unannealed reference sample and (b) is a sample from the same batch annealed at 1600 °C for 1 h.

Table 3
Comparison of Weibull statistics for coated particles as determined by various test methods.

Strength test method	Characteristic strength (MPa)	Weibull modulus	Reference
Internal pressurization	222–320	4.3–6.2	[8]
Diametrical compression	356–427	5.8–7.5	[8]
O-ring compression	1050–1890	4.8–9.4	[4,8]
C-ring compression	980–2200	4.0–9.0	[4,8]
Modified compression	1507–1718[a]	2.2–5.0[a]	[9] Method; RCF coater
Modified compression	926–1119[a]	2.6–5.1[a]	[9] Method; ACF coater
Internal pressurization	484–654	3.69–7.66	Deposition at 1200–1400 °C; [16]
Hemispherical compression	517–491 (mean stress)	7.9–9.4	[17]

[a] Results of this work.

Table 4
Summary of the TEM investigation on the coated particles after annealing.

Batch number	SiC phase (25–2100 °C)	Interlayer integrity (25–2100 °C)	PyC graphitization
G118 (ACF)	3C–SiC 4H at 2100 °C	No debonding	1980 °C
G146 (ACF)	3C–SiC (investigated only up to 1600 °C maximum)	No debonding (results only available to 1600 °C)	Not observed at 1600 °C porosity in PyC see Fig. 10
G169 (ACF)	3C–SiC (investigated only up to 1600 °C maximum)	No debonding (results only available to 1600 °C)	Not observed at 1600 °C
B14 (RCF)	3C–SiC	Debonding at 1980 °C	1980 °C
B10 (RCF)	3C–SiC 4H at 2000 °C	Debonding at 2100 °C	1000 °C

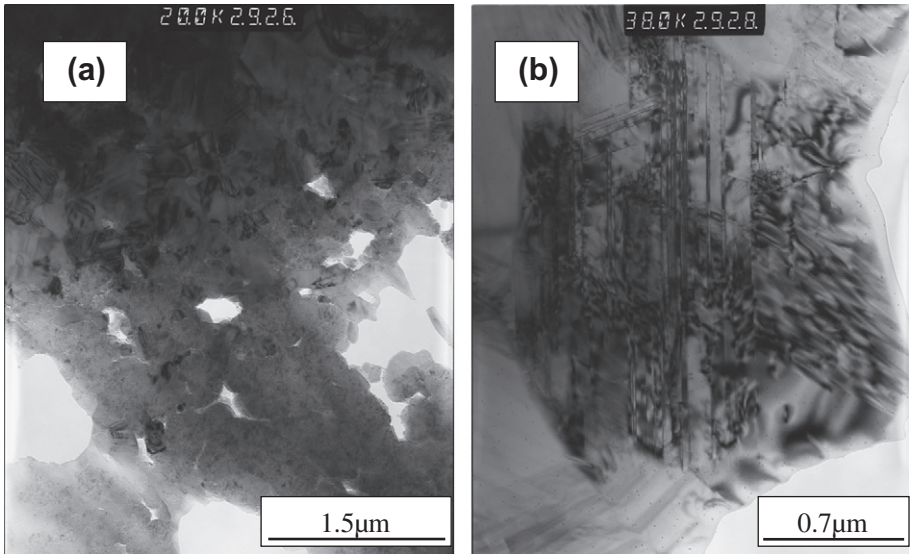

Fig. 15. (a) Bright-field TEM image of the interface between the I-PyC and SiC layers of sample batch G118 and was annealed at 2100 °C for 10 min. Bright-field TEM image (b) showing the high density of twins in a SiC grain with the cubic 3C phase.

lower modulus can be contributed to the fact that these batches are still experimental batches.

The coated particles were also investigated by TEM and these results are summarized in Table 4. The bright-field TEM images of batch G146 is shown in Fig. 14 and show the interfaces between the I-PyC and SiC layers, showing an interlayer. The I-PyC layers contain a high concentration of pores in the region close to the I-PyC–SiC interface. Fig. 14a is the unannealed reference sample and Fig. 14b is a sample from the same batch annealed at 1600 °C for 1 h. No debonding between the I-PyC and SiC was observed in this batch. The 1800 °C and 2000 °C TEM investigations for G146 and G169 are not available.

Fig. 15a shows the bright-field TEM image of the interface between the I-PyC and SiC layers of sample Batch G118 which was annealed at 2100 °C for 15 min. The I-PyC layer contains a high concentration of pores in the region close to the I-PyC–SiC interface. No debonding between the I-PyC and SiC was observed. The bright-field TEM image, Fig. 15b, shows the high density of twins in a SiC grain with the cubic 3C phase.

For batch G118 after annealing at 2100 °C, some SiC grains had the 4H–SiC structure and for batch B10 this is observed after 2000 °C annealing. Debonding of the I-PyC–SiC interlayer took place for the B14 batch as previously reported in [14] and debonding of batch B10 is observed for the 2100 °C annealing sample only.

5. Conclusions

The purpose of this work is to provide a demonstration of the application of the modified compression strength test method to reach a comparison of strength measurements after high temperature annealing of five experimental PBMR CP batches. The main conclusions drawn from this study are as follows:

- Although the actual strength values determined from the modified strength method [9] is not validated yet, these values provide a platform to compare the various experimental PBMR CP batches to establish the effect of annealing temperature on strength.
- The RCF coater batches exhibits higher characteristic strength compared to the ACF batches. The layer thickness and density differences did not provide a plausible explanation for this observation. The deposition rate for the ACF batches are 41% higher than those of the RCF batches which may be a reason for the higher characteristic strength of the B10 RCF batch if compared with the G169 ACF batch, given that deposition temperature of 1510 °C was the same for both batches. From these results it is suggested that the CP strength values are not coater volume dependent.
- The difference in the initial as-manufactured characteristic strength of the two RCF batches can most probably be attributed to the different deposition temperature of B14 (1585 °C) if compared to that of B10 (1510 °C). This result agrees with the finding by Xu et al. [18] that the strength decreases for deposition temperatures higher than 1550 °C.
- It is observed from this work that the highest characteristic strength and modulus for both coater batches are reached at a deposition temperature of 1510 °C which confirms the findings that the strength of SiC was a maximum for temperatures between 1500 °C and 1550 °C and that it decreased significantly for deposition temperatures below and above this temperature range.
- It further also shows the tendency described previously that the characteristic strength increased for the three ACF batches and the one B14 RCF batch after an annealing treatment at 1600 °C. The reason for the reverse trend for the B10 RCF batch is not clear at this stage. It is however important to note that the characteristic strength values of the two RCF batches are not significantly different after 1600 °C annealing. This may suggest that the stresses were reduced after annealing irrespective of the initial characteristic strength.
- The RCF batches exhibit an initial slight displacement to increased strength values of the Weibull plot after annealing at 1600 °C suggesting a slight toughening of the SiC layer as well as a decrease in the spread of the fracture strength measured with the Weibull modulus determined as 7.59 and 10.93 for batches B14 and B10 respectively.
- The Weibull graph of the RCF batches show a decrease in values after annealing at 2100 °C with a significant decrease in the characteristic strength and only a decrease in the Weibull modulus of batch B10. The TEM investigation revealed that debonding of the SiC–I-PyC interlayer takes place at 2100 °C and 1980 °C for the B10 and B14 batches respectively. The debonding of the SiC–I-PyC interlayer may cause rougher interface surfaces and higher stress concentrations, which can contribute towards lower strength values and larger spread in strength values.
- The annealing temperature dependence of the ACF batches, G118 and G169, show that the Weibull modulus and characteristic strength increase with increasing annealing temperature from 1600 °C to 2100 °C.
- The results of the ACF batch G146 shows very low modulus values and typically this batch would not be recommended for use in a High Temperature Reactor because the low-strength tail will cause CPs to fail early. Another interesting phenomenon is that batch G146 has the lowest Weibull modulus (1.86) of the five batches evaluated after annealing at 1600 °C. This batch also deviates from the characteristic Weibull straight line after annealing at 2000 °C with "two" distinct value populations. This aspect is the subject of further investigations. The density of the I-PyC is slightly below the specification (1.8–2.0 g/cm^3) and the lowest (1.67 g/cm^3) of the ACF batches. It is suggested that the low SiC layer deposition temperature and high deposition rate may have contributed towards the very low modulus values.
- The low Weibull modulus and slightly lower I-PyC density of the ACF G146 batch may be attributed to the significant amount of porosity in the I-PyC and SiC that was observed during the TEM analysis. The high porosity of the I-PyC results most probably into an irregular I-PyC–SiC interface that may cause fracture initiating points from stress concentrations.
- The modulus range for this work is 2.2–5.1 and is slightly lower than the modulus values of 4–6 as proposed by Snead et al. [8]. However, if only the strength values of batches G118, G169 and B10 are considered, the modulus range are 4.3–5.1 which corresponds to the modulus values as proposed by Snead et al. [8]. It is however important to note that Miller et al. [12] used a SiC modulus of 6 for CP performance modelling and the modulus of all these experimental batches are lower than 6. It is also important to note that the test methods are different and a direct comparison may not be accurate.
- Results of this study generally support the observation made by Snead et al. [8] that no significant strength degradation of CVD SiC occurs for temperatures up to 1500 °C because results of this study shows that an increase in strength is noted for temperatures above 1600 °C. In addition to the above findings, this work shows also an increase in strength up to 2000 °C with a decrease in strength only after annealing at 2100 °C.
- The Weibull graph for the strength after long duration annealing of 100 h of the two RCF batches shows no significant change in the characteristic strength values although the modulus decreases from 7.59 to 5.28 (~33%) and from 10.93 to 8.46

(~22%) for batches B14 and B10 respectively. In contrast to this observation, the G118 ACF batch showed a slight increase in the characteristic strength from 1191 MPa to 1336 MPa and a slight increase in the modulus value from 4.35 to 5.62 (~29%).

6. Recommendations

It is recommended that the actual values of the strength determination methods be validated against other method(s) and that the influence of interlayer thickness and grain size distribution on the strength of the CPs be investigated. It is also further recommended for future investigation that the strength of the PyC layers also be determined by calculation from the results obtained from the modified compression method. Although this study did not focus on the influence of manufacturing parameters on the strength profile, it revealed interesting relationships which are recommended for further study.

Acknowledgements

This research was sponsored by PBMR's Fuel Optimization Technology Programme. The use of the NMMU and PBMR Fuel Development Laboratory facilities are gratefully acknowledged. Johannes Mahlangu (PBMR), Ellen Nquma (PBMR), Jaco Olivier (NMMU) and Jacques O' Connell (NMMU) are thanked for the annealing operations. The reviewing of this article by Etienne de Villiers (PBMR) and Fanie Swanepoel (PBMR) is much appreciated. We thank Professor Gerrit T. van Rooyen (University of Pretoria) for the strength determination of the CPs.

References

[1] H. Nabielek, P.E. Brown, P. Offerman, Nucl. Technol. 35 (1977) 483.
[2] H. Nabielek, SiC Workshop, Paris, 2009.
[3] K. Bongartz, E. Gyarmath, H. Nickel, H. Schuster, W. Winter, J. Nucl. Mater. 45 (1972) 261–264.
[4] K. Bongartz, E. Gyarmath, H. Schuster, K Täuber, J. Nucl. Mater. 62 (1976) 123–137.
[5] T. Ogawa, K. Ikawa, J. Nucl. Mater. 98 (1981) 18–26.
[6] K. Minato, K. Fukuda, K. Ikawa, H. Matsushima, S. Kurobane, J. Nucl. Mater. 119 (1983) 326–332.
[7] A. Briggs, R.W. Davidge, C. Padgett, S. Quickenden, J. Nucl. Mater. 61 (1976) 233–242.
[8] L.L. Snead, T. Nozawa, Y. Katoh, T. Byun, S. Kondo, D.A. Petti, J. Nucl. Mater. 371 (2007) 329–377.
[9] G.T. van Rooyen, Rudy du Preez, Johan de Villiers, R. Cromarty, J. Nucl. Mater. JNM-09-00472, 2009.
[10] T.S. Buyn, E. Lara-Curzio, R.A. Lowden, L.L. Snead, Y. Katoh, J. Nucl. Mater. 367–370 (2007) 653–658.
[11] R. Benz, W. Jungen, K. Lesten, A. Schirbach, IRW-TN-124/82, 1982, pp. 5.
[12] G.K. Miller, D.A. Petti, J.T. Maki, D.L. Knudsen, J. Nucl. Mater. 355 (2006) 150–162.
[13] H. Nabielek, W. Schenk, A.W. Mehner, D.T. Goodin, Nucl. Technol. 84 (1989) 62–81.
[14] I.J. van Rooyen, J.H. Neethling, J. Mahlangu, In: Proceedings of the 4th International Topical Meeting on High Temperature Reactor Technology, 28 September 2008, Washington, DC, USA, HTR 2008-58189, 2008.
[15] Y. Katoh, L. Snead, J. ASTM Int. 2 (8) (2005) (paper ID JAI12377).
[16] M.W. Kim, J.H. Kim, H.K. Lee, J. Park, W. Kim, D.K. Kim, J. Ceram. Process. Res. 10 (3) (2009) 373–377.
[17] M.W. Kim, J.N. Park, M.S. Cho, J.Y. Park, J. Nucl. Mater. 392 (2) (2009) 213–218.
[18] S.J. Xu, J.G. Zhou, B. Yang, B.Z. Zhang, J. Nucl. Mater. 224 (1995) 12.