J. Knorr, A. Kerber and R. Moormann

Upgrading (V)HTR fuel elements for generation IV goals by SiC encapsulation

The pebble bed reactor is one of the most promising concepts for (V)HTR. Nevertheless recent re-evaluation of AVR and THTR results emphasizes once more that claimed advantages strongly depend on the properties of the fuel elements and their behavior under operational conditions. Additionally safeguards, waste management and disposal aspects gain increasing importance today. The conventional uncoated graphite pebbles meet only inadequately the requirements of Generation IV facilities. Since long experts agree, that corrosion-resistant pebbles with high retention capability for fission products would considerably improve the chances of the pebble bed reactor concept. With laser beam joining of ceramics the key technology is now available for the silicon carbide (SiC) encapsulation of (V)HTR components. The envisaged innovative fuel element consists of a robust SiC hollow sphere filled with moderator and TRISO coated particles. The positive assessment according to Gen IV criteria should justify necessary R&D efforts to obtain qualified fuel elements and demonstrate their superiority under operational conditions.

Verbesserung der (V)HTR Brennelemente gemäß Generation IV-Zielen durch SiC-Einkapselung. Der Kugelhaufenreaktor ist eines der vielversprechendsten Konzepte für den (V)HTR. Nichtsdestotrotz zeigen neuere Einschätzungen der AVR- und THTR-Ergebnisse einmal mehr, dass die reklamierten Vorteile stark von den Eigenschaften der Brennelemente und ihrem Verhalten unter Betriebsbedingungen abhängen. Zusätzlich gewinnen heute Aspekte von Safeguards, Abfall-Management und Endlagerung eine wachsende Bedeutung. Die konventionellen, unbeschichteten Graphit-Kugeln erfüllen die Anforderungen an Generation IV-Anlagen nur ungenügend. Seit langen ist man sich in Fachkreisen einig, dass korrosionsresistente Brennelemente mit hoher Rückhaltefähigkeit für Spaltprodukte die Zukunftschancen des Kugelhaufenreaktor-Konzepts erheblich verbessern würden. Mit dem Laser-Fügeverfahren für Keramik ist nun die Schlüsseltechnologie für eine Siliziumkarbid (SiC)–Kapselung von (V)HTR-Komponenten verfügbar. Das vorgestellte innovative Brennelement soll aus einer robusten SiC-Hohlkugel bestehen, die mit dem Moderatormaterial und TRISO Coated Particles gefüllt wird. Die positive Einschätzung der erwarteten Eigenschaften gemäß den Gen IV-Kriterien sollte verstärkte F&E-Anstrengungen rechtfertigen, um qualifizierte Brennelemente herzustellen und ihre Überlegenheit unter Betriebsbedingungen nachzuweisen.

1 Introduction

The (V)HTR is a next step in the evolutionary development of high-temperature gas-cooled reactors. It becomes especially attractive when core outlet temperatures higher than about 1000 °C would enable nuclear heat application to such processes as hydrogen, steel and aluminum production. Recent re-evaluation of AVR and THTR results however emphasizes once more that the claimed advantages of pebble bed reactors (PBR) strongly depend on the properties of the fuel elements and their behavior under operational conditions [1]. Additionally, safeguards, waste management and disposal aspects gain increasing importance for Generation IV facilities [2]. From the present point of view the conventional nude graphite pebbles meet only inadequately the requirements of Generation IV facilities. Since long experts agree, that corrosion-resistant pebbles with high retention capability for fission products would considerably improve the chances of the PBR concept [3, 4].

2 Goals and design criteria

Demonstrating the viability of the (V)HTR core requires meeting a number of significant technical challenges. Novel fuel elements and materials must be developed [2] that

- permit core-outlet temperatures of about 1000 °C and preferably even higher
- permit the maximum fuel temperature following accidents to reach 1800 °C
- permit maximum fuel burn-up of 150–200 GWD/MTHM
- avoid power peaking and inadmissible temperature gradients in the core, as well as inadmissible hot streaks in the coolant gas.

Generation IV Roadmap identifies 8 goals in 4 areas and related 15 criteria and their 24 metrics assigned to the various goals. On this basis and our present results of feasibility studies the chances were assessed, how SiC encapsulation can help to fulfill Generation IV requirements.

3 SiC encapsulated pebble fuels

3.1 Standard graphite fuel element

Since the very beginning with AVR different types of spherical fuel elements have been developed [5]. Over the years one particular design evolved which can be considered today as the standard pebble fuel element. A two-zone pressed graphite body with an outer diameter of 60 mm is manufactured in several steps. The inner zone (diameter 50 mm) is a graphite matrix containing the fuel as coated particles, today preferably UO_2-low-enriched TRISO particles with SiC or ZrC coating. The outer graphite shell is free of fuel with no additional coating. Hereafter this nude graphite fuel element is referred to as standard element.

The dominating weak point of this design is the outer graphite shell. Basic properties and its consequences will be discussed later in this paper in comparison with the new SiC encapsulated pebble.

3.2 Corrosion-resistance coating methods

Many efforts have been undertaken to improve the properties of the standard element by coating the surface of the outer shell or by encapsulation in ceramics. Under discussion are coating by SiC CVD or PVD methods as well as silicon infiltration. Also encapsulation of spherical fuel elements – moulded and pressed like standard elements – in SiC half spheres has been considered.

With the laser beam joining of ceramics the key technology is now available for the SiC encapsulation of (V)HTR components and waste [6]. The irradiation behavior of the solder is under investigation. Results will be published in a separate paper.

Carefully weighing the pros and cons of the different methods the SiC hollow sphere is selected as the most promising concept taking up an early days idea when hollow spheres were machined from graphite and filled with a moderator-fuel matrix. To close the opening a graphite plug was screwed into the shell, which does not form a gas-tight seal, but the graphite shell has in any case only a limited retention capability.

3.3 SiC hollow sphere encapsulation

The envisaged new fuel element consists of a robust SiC hollow sphere filled with moderator-fuel matrix. For better comparison, the same geometrical dimensions as standard element have been chosen (Fig. 1).

The hollow sphere offers the possibility to precipitate an additional special diffusion barrier on the inside surface thus enhancing the retention capability for overall fission product release from coated particles and contaminated moderator material as well.

The moderator-fuel matrix is preferably the same as for standard element, e. g. graphite and TRISO coated particle according state-of-art and favoured fuel (U or Th). After filling the moderator-fuel matrix in the SiC hollow sphere and densification by vibrating technology the opening is closed with a SiC plug and gas-tight sealing through laser beam joining [6].

Recent R&D efforts were focussed on two issues:

- SiC material development and
- manufacturing technology for SiC hollow spheres.

There are different types of silicon carbide:

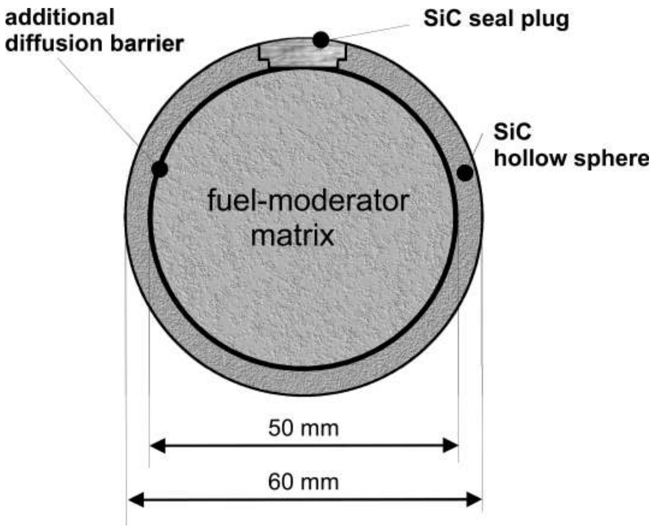

Fig. 1. Cross section of SiC hollow sphere fuel element

Liquid-phase sintered SiC (LPSiC), solid-state sintered SiC (SSiC), silicon-infiltrated SiC (SiSiC), re-crystallised SiC (RSiC) and nitride bonded SiC (NSiC). Selected properties of those are listed in table 1 [7]. Recrystallized SiC (RSiC) and nitride bonded SiC (NSiC) are not suitable due to their open porosity.

It is widely agreed that after accidents pebble bed cores can reach temperatures up to 1600 °C at least for several days. Therefore SiSiC – despite of its other excellent properties – is ruled out because of its too small maximum temperature of use. Only pressureless sintered SiC can sustain the required high temperatures. Additionally, zero porosity and great hardness make SSiC the best choice.

Modifications in the SSiC standard composition are necessary to garantuee a low neutron absorption cross-section. With SICANA@ a new boron-free material composition was developed for advanced in-core application.

After trials and errors the authors now trust to a simple and effective method for manufacturing SiC hollow spheres, which simultaneously has the promising capability for production on industrial scale (Fig. 2).

In a final step the fuel element gets a laser-engraved fabrication Safeguards Code and the Safeguards plug seal (Fig. 3).

4 Assessment of fuel upgrading by SiC encapsulation

Up to now essential manufacturing procedures for SiC encapsulation have been developed and tested on laboratory scale.

Table 1. Selected properties of the dense SiC types

parameter	symbol	unit	LPSiC	SSiC	SiSiC
open porosity		[Vol%]	< 1	0	0
4-point bending strength	σ_B	[MPa]	600	260–600	180–460
elastic modulus	E	[GPa]	420	350–450	270–400
specific heat	c_p	[J/kg/K]	600	600–1000	650–1300
thermal conductivity	λ	[W/m/K]	100	40–120	100–160
thermal shock resistance	assessed	[–]	good	good	very good
maximum temperature of use	T	[°C]	1400	1760	1380

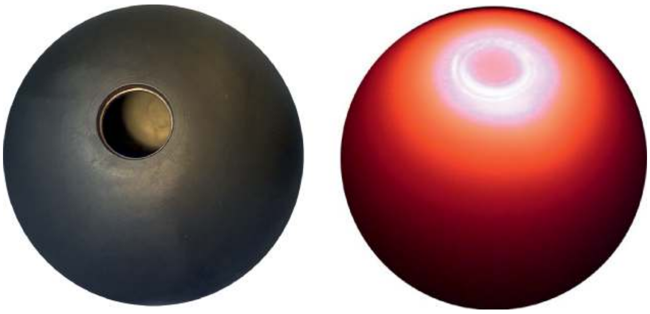

Fig. 2. SSiC hollow sphere after sintering (l) and during sealing with laser beam (r) [8]

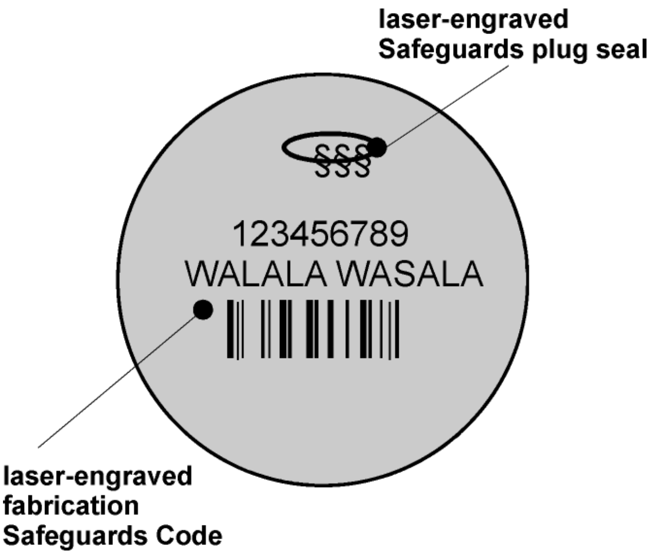

Fig. 3. Laser engraving for nuclear material control

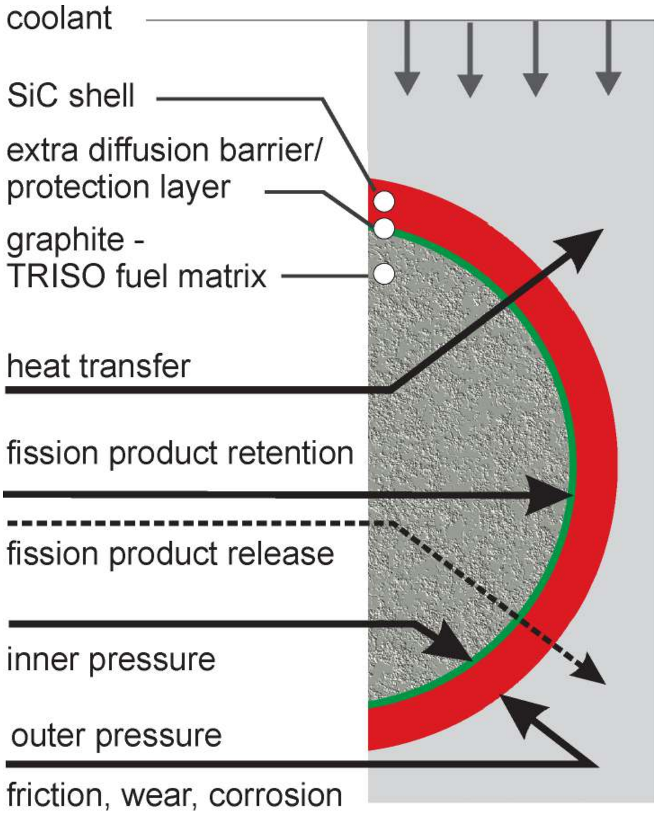

Fig. 4. SiC shell: multi-functional protection

Taking into account our present state of experience and our understanding of necessary future R&D efforts this paper does not claim for a todays overall competence in manufacturing of qualified SiC encapsulated fuel elements. But with the subsequent assessments and comparison with standard element it tries to foster further discussions and developments in the field of SiC encapsulation of components for Generation IV facilities.

The outer shell is a well-stressed part of the pebble with many functions for mechanical, thermal, chemical and nuclear stability (Fig. 4):

- guarantee heat transfer and fission product retention
- sustain inner and outer pressure
- provide low corrosion rate, little wear
- a friction, which allows a predictable pebble bed dynamic.

SiC encapsulation shall result in improvements in the areas sustainability, economics, safety and reliability and proliferation resistance and physical protection. The main objectives are listed as follows:

- higher retention capability by additional (specially tailored) diffusion barrier/protection layer (in particular for metallic fission products)
- minimized dust generation, therefore drastic reduction of source term and explosion risk
- corrosion resistance in case of air and/or water ingress
- fresh fuel already pre-conditioned for interim and final storage
- identification of each individual fuel element for safeguards and core load history.

Upgrading of (V)HTR fuel elements for Generation IV goals by SiC encapsulation are met by some features alone or in their combination. Taking into account the properties of SiC and graphite respectively as materials for the outer shell and comparing their behaviour under operational and accidental condtions the superiority of SiC encapsulated pebbles becomes apparent. The condensed opinion of the authors is summarized in a 5 point score table (Table 2).

Only few verbal comments are offered in this paper.

For example, the thick SiC shell ($s = 5000\ \mu m$) provides a superior retention capability for fission products over TRISO particles (SiC thickness only $s = 35\ \mu m$). Common criteria are the break through time t_B (prop. to s^2) and the diffusion rate (prop. to 1/s). SiC encapsulation of the pebble prolongs t_B by a factor 2×10^4, not to mention the retention of fission products of failed TRISO particles or a contaminated matrix, which are released almost totally by the standard fuel element. Even in case of intact coated particles the diffusion rate is reduced by more than two orders of magnitude. Altogether this may allow to overcome the limits of current fuel of at most 750 °C He-temperature [9].

A second example: With a laser engraved identification number each SiC pebble becomes 1 batch in the terminology of international nuclear material control. Now the complete history of each pebble can tracked down and recorded from manufacturing over operation, interim storage, transport to final storage.

As long as there is no possibility to identify each individual fuel element with recorded history and predictable movement

Table 2. Estimated superiority of SiC encapsulated pebbles over standard uncoated graphite fuels (++: much better, +: better, =: equal, – : worse/less, ?: open/undecided)

No	generation IV metrics	estimated superiority of SiC pebble over standard fuel elem.
1	use of fuel resources	=
2	waste mass/volume	= +
3	heat load	=
4	radiotoxicity	+
5	environmental impact	++
6	overnight construction cost	?
7	production costs	– ?
8	construction duration	=
9	risk to capital	+
10	forced outage rate	= +
11	routine exposures	+
12	accident exposures	++
13	reactivity control	+
14	decay heat removal	=
15	phenomena uncertainty	+
16	long fuel thermal response	++
17	integral experiments scalability	?
18	source term	++
19	mechanisms energy release	?
20	long system time constants	+
21	long/effective holdup	+
22	separated materials	+
23	spent fuel characteristics	++
24	passive safety features	=

in the core, the core load of a pebble bed is more or less a "black box" in its actual material composition and geometrical arrangement, which complicates thermohydraulic, nuclear, safety and safeguards estimations. For pebble bed immanent reasons there is no permanent in-core instrumentation of local neutron flux and local temperatures. This underlines strongly the necessity, that the components and the overall concept must be robust enough to tolerate even reduced safety margins and mitigate their consequences for operation as well as in case of an accident.

5 Upgrading fuel elements – recent knowledge gain on PBR performance

Knowledge gain of the past 3 years enhances doubts about the feasibility of the present PBR concepts for high temperature operation and force improvements, particularly those of fuel elements: At first, post examinations of the most advanced PBR TRISO fuel developed in Germany, GLE-4, which was irradiated in the EU1bis experiment in the Petten reactor at up to 1200 °C fuel temperature to a burn-up of 11.7 % fima, revealed that during irradiation unacceptable large amounts of metallic fission products were released (Cesium and Silver) [9]. It means that a maximum operational fuel temperature of 1250 °C, which was assumed in the past to be tolerable for modern TRISO fuel and which was the basis of process heat generating PBR designs, is too optimistic. Following [9], coolant temperatures of 750 °C should not be transgressed. In [9] temperature anomalies as observed in AVR and THTR-300 were not taken into account and may worsen the situation. Desirable process heat applications as CO_2-free hydrogen generation by water splitting or coal gasification, which were intended by PBRs in the past but require coolant temperatures > 900 °C, are thus not achievable with current fuel, except a conventional overheating of the nuclear preheated coolant is applied. The latter is however not an economically viable option. Further, because of the much larger yield of Silver in Plutonium based fuel, Pu-burners based on PBRs seem to be particularly problematic for present TRISO fuel. These results of the EU1bis experiment are in line with predictions performed in [1] and are due to the before mentioned weak point of the current PBR fuel concept: Due to the comparably high diffusion coefficients of Cs and Ag in SiC, the SiC layer cannot act as a long term diffusion barrier for these nuclides at fuel temperatures > 800 °C (Silver) respectively > 1100 °C (Cesium) because of its small thickness of only 35 μm. Accordingly an additional diffusion barrier, as a thick SiC layer on the outer surface of the fuel elements, is worth to be examined more detailed.

At second Chinese experiments on the pebble flow behaviour in pebble beds indicated that stagnant regions may occur near to the walls where the cylindrical and the funnel like core sections join [10]. It is however not fully clear, to which extent these experiments are representative for real pebble beds. Stagnant pebble bed regions have to be excluded for current fuel elements because the high burn-up reached may be far outside from guaranteeing the stability of their activity retention barriers.

At third Chinese examinations on the influence of the particularly large friction coefficient of graphite in Helium on the pebble flow behaviour, which was not sufficiently considered in the past, indicated that significant deviations in the flow pattern may occur [11]. Having in mind that temperature anomalies in THTR-300, in particular locally too high fuel element temperatures, were related to unexpected pebble flow behaviour [14], this result requires more detailed examinations. As for AVR, there are diverse explanations for the temperature anomalies in THTR-300, and these PBR temperature problems remain unresolved. A recent development of a more realistic simulation model for pebble flow in a PBR of some hundred MWth power indicated still impracticable large calculation times even on multiprocessor systems [12]. This underlines the complexity of the PBR pebble flow, although influence of dust or changes of pebble shape by damage are not yet taken into account. With respect to graphite dust, formed during pebble flow under high friction, the widely unknown item has to be added that some coolant channels in the bottom reflector of the THTR-300 were partly blocked by dust and by pebble debris. There were examinations on an improved PBR design, which tries to avoid such blockage [13].

6 Conclusion

There are good reasons to believe that the mechanical stability and the flow characteristics of SiC encapsulated fuel elements under operational conditions of (V)HTR are not worse – but even better – than those of standard nude graphite elements. Under this assumptions, which of course have to be proven by qualification of the fuel manufacturing process and extensive fuel testing in large-scale experiments, the potential of SiC encapsulation for upgrading fuel elements for Generation IV goals is obvious and should justify enforced R&D before premature solutions endanger the pebble bed concept as (V)HTR.

Acknowledgement

The authors very much appreciate the creative contributions of Mr. H. Aicher, Managing Director of ATP Produktentwicklung GmbH, Oberteisendof Germany, to the development of SiC hollow sphere manufacturing process.

(Received on 20 December 2011)

References

1 *Moormann*, *R.*: AVR prototype pebble bed reactor: a safety re-evaluation of its operation and consequences for future reactors. Kerntechnik 74 (2009) 8–21, http://www.nuclear-engineering-journal.com/KT110001
2 A Technology Roadmap for Generation IV Nuclear Energy Systems. U.S. DOE Nuclear Energy Research Advisory Committee and the Generation IV International Forum, December 2002
3 *Schulten*, *R.*: Alte und neue Wege der Kerntechnik. Fusion 1/1990 und 1/2010, p. 20–23
4 *Kugeler, K.*: private communication, 2010
5 *Hackstein, K; Wimmer, M.*: Chapter 2.4 Fuel Cycle, Experimental High temperature Reactor, Düsseldorf (1990)
6 *Knorr, J; Lippmann, W; Reinecke, A-M; Wolf ,R; Kerber, A; Wolter, A.*: SiC encapsulation of (V)HTR components and waste by laser beam joining of ceramics. Nuclear Engineering and Design 238 (2008) 3129–3135
7 TECHNICAL CERAMICS Breviary Association of the German Ceramics Industry Fahner Verlag, Lauf 2004
8 *Reinicke, A-M.*: TU Dresden, Department of Nuclear Engineering, 2005
9 *Freis, D.*: Störfallsimulationen und Nachbestrahlungsuntersuchungen an kugelförmigen Brennelementen für Hochtemperaturreaktoren. PhD thesis RWTH Aachen, 2010, http://darwin.bth.rwth-aachen.de/opus3/volltexte/2010/3307/. English translation: pbadupws.nrc.gov/docs/ML1127/ML11278A104.pdf
10 *Yang, X; Hu W; Jiang, S.*: Experimental Investigation on Feasibility of Two-Region-Designed Pebble-Bed High-Temperature Gas-Cooled Reactor. Journal of Nuclear Science and Technology 46 (2009) 374–381
11 *Xiaowei, L; Xiaotian, L; Suyuan, Y.*: Nuclear graphite friction properties and the influence of friction properties on the pebble bed. Nuclear Engineering and Design 240 (2010) 2674–2681
12 *Cogliati, J.J; Ougouag, A.M.:* PEBBLES Mechanics Simulation Speedup. Pres. PHYSOR2010, Report INL/CON-09-17136, May 2010
13 *Gerwin, H; Scherer, W.*: Kugelhaufenreaktor, Patent DE19547652C1, 06.03.1997
14 *Kalinowski, H.*: THTR core physics and pebble flow. presentation to an US-NRC delegation (2001), in Safety aspects of HTR technology, NRC Adams webserver document ML092250104, http://wba.nrc.gov:8080/ves/

Authors of this contribution

Prof. (em.) Dr. *Juergen Knorr*
GWT Technische Universitaet Dresden GmbH
Blasewitzer Str. 43
D-01307 Dresden
Email: juergen.knorr@gwtonline-fb.de

Dr. *Albert Kerber*
SiCeram GmbH
Am Nasstal 10
D-07751 Jena-Maua
Email: a.kerber@jsj.de

Dr. *Rainer Moormann*
Forschungszentrum Juelich GmbH
D-52425 Juelich
Email: r.moormann@gmx.de

You will find the article and additional material by entering the document number **KT110218** on our website at *www.nuclear-engineering-journal.com*