Design Optimization of Multi-Layer Silicon Carbide Cladding for Light Water Reactors

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1. Introduction

Silicon Carbide (SiC) is considered as a potential alternative for the current zirconium-based alloy (zircaloy) cladding. SiC exhibits a number of attractive features for a light water reactors (LWR) cladding material. It is a well-known high temperature refractory material that exhibits excellent high temperature and irradiation tolerance [1-7]. Its high temperature steam reaction rate is a few orders of magnitudes slower than that of zircaloy, essentially eliminating safety issues associated with structural integrity degradation by oxidation [8-10]. These are advantageous features from the safety point of view. Yet, even in operation, SiC cladding is as transparent as zircaloy to neutrons [11,12], which implies minimal, if any, departure from the current LWR core designs.

Despite of the proven advantageous features of SiC, its deployment in LWRs requires to assure reliable operation against its inherently brittle fracture mode. Today, a consensus is being made on the critical importance of understanding its fracture as a cladding material [13,14], and improving its structural integrity. A recent work conducted at Massachusetts Institute of Technology (MIT) [13] proposed a duplex cladding of the inner fiber-reinforced composite (SiC_f/SiC) layer and the outer CVD-SiC layer. This cladding design is found to markedly reduce steady-state cladding failure probability than the conventional triple-layered cladding design. Even at today's highest allowable burnup level, the duplex cladding is anticipated to give steady-state fuel failure probability (P_f) less than ~10^{-4} (P_i=20MPa) whereas the triple-layered cladding design is predicted to almost certainly fail (P_f ~1). Such a marked difference in steady-state cladding failure probability with different layer compositions illuminates:

(1) Triple-layered SiC is an imprudent design based on qualitative reasoning for the presence of each layer, which was conducted in the early phase of the concept development.

(2) The cladding layer design holds a key to improving structural integrity of SiC cladding.

It is noteworthy that zircaoly cladding performance has been gradually improved by its composition and coolant chemistry optimization since its first deployment in 1960s. The presented cladding design optimization directions lie in its key failure causes. That, in case of zircaloy, its chemical affinity with water, hydrogen and irradiation induced embrittlement are the key degradation mechanisms for which chemical remedies should be used. In case of SiC cladding, load-induced brittle fracture is the dominant failure mechanism, for which management of tensile stresses holds the key.

Tensile stress managements in SiC cladding can be done in a few different ways. Advancing composite architecture, reducing residual stresses in the manufacturing stage are the two common methods that could be done on the material level. Yet, considering relatively high maturity of today's SiC composite architecture and manufacturing technologies, potential for their advancement is considered marginal. A greater improvement can be found from the cladding layer optimization. Indeed, today in the SiC cladding development community, a consensus that the management of tensile stress level in cladding is in the realm of design has been made. That, thanks to well understood material properties of both monolithic [4] and fiber-reinforced composite SiC [15-20], today we can ever accurately model stress distributions of the SiC cladding.

This study is dedicated to find an optimal cladding layer composition of multi-layered SiC cladding. It is important to note that an optimal cladding design for steady-state operation is likely to be different from that of accident conditions. An ideal cladding design optimization should, however, assure its ever most reliable performance in steady-state operation while demonstrating accident coping capability in combination of emergency core cooling systems (ECCS). Hence, in this pioneering work of SiC cladding layer optimization, the scope of our exploration is limited to steady-state operation. Once the steady-state optimization is finalized, a marginal departure from the proposed design may be allowable in the direction of improving its accident performance, which we find worthy of exploring in the future.

2. Design goals and lessons from previous studies

Design optimization of SiC cladding should be conducted to achieve its key functional roles. They are summarized as follows:

- Hermeticity of fission gases

Gas tightness of cladding is a basis for the need of monolithic CVD-SiC layer. Dispersed matrix cracking SiC_f/SiC in the course of fiber-stretching prevents the use of the sole composite layer as the cladding structure.

- Load-bearing capability

SiC cladding is subject to stresses arising from (1) mechanical loading by pressure difference, (2) differential thermal expansion across the cladding thickness, and (3) differential irradiation-swelling induced expansion across the cladding thickness. Radiation swelling in SiC depends on temperature and quickly saturates with 0.1 – 1 dpa for temperature ranges of 200ºC-800ºC. Unlike thermal expansion, SiC swelling is greater for the lower temperature. Hence, swelling induces tensile stresses in the inner (hotter) region of the cladding and compressive stress in the outer (colder) region [13]. SiC cladding layer compositions should be designed to minimize the overall tensile stresses.

- Heat transferring capability

Increasing thermal conductivity of multi-layered SiC cladding is desirable. A lower thermal conductivity of a cladding material increases fuel temperature. A higher fuel temperature increases fission gas releases and fuel pellet expansion, which eventually undesirably raises cladding stress level. Thermal conductivity of CVD-SiC (~9.5 W/m-K at ~1dpa [4,13,15]) is far lower than that of SiC_f/SiC (~1.5 W/m-K at ~1dpa [13,15,17]).Hence, a larger relative fraction of CVD-SiC is more desirable from the heat transferring point of view.

- Water reaction protection

Previous studies [21-23] found that high-temperature steam oxidation tolerance of SiC_f/SiC is found to be markedly impaired when steam ingresses through matrix cracks. The pyrolytic carbon coating of SiC fiber is submissive to high temperature steam reaction, which causes loss of its nominal mechanical behavior. This fact mandates to place a protective coating on the cladding outer surface that blocks steam ingression. In a multi-layered SiC cladding structure, CVD-could function as both steam ingression barrier and load-bearing structure.

Previous analyses showed that swelling strain of SiC cladding is significant [13,14]. In the triple-layered cladding design, irradiation swelling causes a strong tensile stresses in the inner monolithic CVD-SiC, which seriously challenges the hermeticity of fission gases as well as load-bearing capability of the cladding as a whole. This analysis is found to be in agreement with triple-layered SiC irradiation experiments conducted at HFIR in Oakridge National Laboratory (ORNL), which showed radial cracks in the inner CVD-SiC after irradiation exposure equivalent to the discharge burnup of 20MWd/kgU [24]. Hence a conclusion is drawn; a triple-layered cladding inappropriately puts tensile stresses in the inner CVD-SiC layer, which is particularly susceptible to tension. Tensile stresses in the inner CVD-SiC layer further increases with burnup with increasing internal pressurization. Calculated cladding failure probability close to unity towards the end of burnup is a clear sign that the triple-layered cladding design should be off the table. Hence, in this study, we chose the duplex cladding as an optimization target. Fig.1 shows a schematic illustration of duplex SiC cladding. The duplex cladding is engineered to load peak mechanical and swelling induced tensile stresses in the inner SiC_f/SiC and the outer CVD-monolith layer provides heremeticity and protection against steam reaction. From the load-bearing capability point of view, the duplex cladding is coherent with the norm of the material usage; SiC_f/SiC should carry the peak tensile stresses as it is significantly more tolerable of tensile stress than CVD-SiC. With the CVD-SiC layer placed at the outer most surface, the duplex cladding also is consistent with the hermeticity and water reaction protection points of view. Regarding heat transferring capability, the reduced number of layers could work in an advantageous way because it decreases undesirable contact resistances at the interfaces.

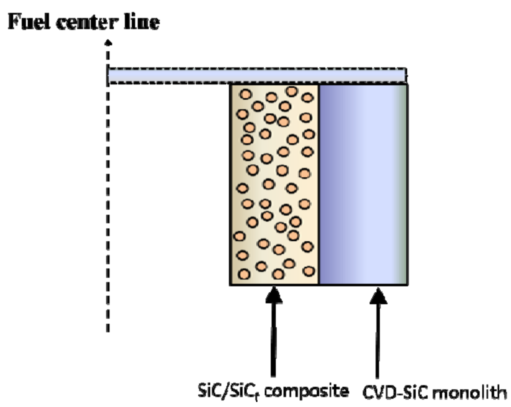

Fig.1 Schematic illustration of duplex SiC cladding [13]

Consequently, a series of previous researches clearly illuminate that the duplex cladding is a better engineered structure for which the composition optimization process is worthy of conducting. In this study, we used the multi-layer ceramic structural analysis code developed by the first author at MIT [13] for the design tool. The code was validated with independent comparisons with finite element analysis (FEA) solutions obtained from ANSYS for both the duplex and the triple-layered cladding [13]

3. Cladding optimization

3.1. Characteristic stress distribution of duplex SiC cladding

Structural integrity of SiC cladding is the most severely challenged at end-of-life (EOL) where the maximum fuel internal pressurization with the accumulation of released fission gases is found. Irradiation swelling quickly saturates shortly after the beginning-of-life (BOL). Thermal stresses levels slightly decrease towards EOL because of decreasing linear fuel pin power. Yet, the fuel internal pressurization outweighs the decrease in thermal stress, resulting in the highest total stress level in EOL. Hence, stress distribution at EOL is of particular interest in evaluating structural integrity of SiC cladding. Table 1 summarizes reference SiC properties for stress calculation at EOL [4,13,15,17].

Table 1 Reference SiC cladding thermos-mechanical properties for stress modeling at EOL

	CVD-SiC	Fiber-Reinforced SiC_f/SiC [Hi-Nicalon™ Type-S CVI-SiC]
Young's modulus [GPa]	460	230
Poisson's ratio	0.21	0.13
Thermal conductivity (W/m-K)	9.5	1.5
Thermal expansion coefficient (1/K)	4.66×10^{-6}	4.66×10^{-6}
Weibull modulus	7.5	Proportional limit: 5
		Ultimate fracture: 17.5
Weibull Characteristic Stress (MPa)	369	Proportional limit: 105
		Ultimate fracture: 290
Weibull Effective volume	6.36×10^{-10} m^3	Proportional limit: 15 x 3 x 2.33mm
		Ultimate fracture: 15 x 3 x 2.33mm

Dose-saturated temperature dependent volumetric swelling S(T) was found by fitting experimental data [15] with a linear function [13] of temperature T.

$$S(T)=-2.4 \times 10^{-5}\, T + 0.031487 \qquad (1)$$

Where T is in Kelvin (K).

A recent work of Katoh et al.,[20] resolved a controversial issue on the difference of irradiation swelling between CVD-SiC and SiC_f/SiC. The study concluded that there is no convincing reason to believe any appreciable difference between their irradiation swelling magnitudes. Hence, in this study, their swelling magnitudes were treated identical with Eq.(1).

Reference fuel operating conditions for EOL are summarized in Table 2 [13].

Table 2 Reference SiC cladding thermo-mechanical properties for stress modeling at EOL

Internal pressure (MPa)	20
Operating pressure (MPa)	15.5
Linear pin power (kW/m)	18.0
Cladding outer surface temperature (°C)	328.8

The fuel rod internal pressure and the cladding outer surface temperature are referred from results of high-burnup steady-state fuel rod simulation (67MWd/kgU) of FRAPCON-3.4 SiC, which is a modified FRAPCON-3.4 with SiC cladding properties [25]. The linear pin power is extracted from work of Bloore and Kazimi [12] on reactor physics simulation for SiC clad fueled core with Studsvik core management system.

Fig.2 shows calculated radial stress distributions in principle directions (hoop, axial, and radial) for the reference duplex SiC cladding of thickness 0.57mm with different layer compositions in EOL. For hoop and axial stresses, peak tensile stresses occur at the inner surface of the composite, which is an intended engineering outcome of the duplex cladding structure. Radial stresses (Fig.2c) are continuous throughout the entire cladding thickness. Note that the magnitudes of the radial stresses are substantially smaller than those of the composite and the monolith, indicating that they do not appreciably contribute to the material failure. Discontinuous hoop and axial stresses are found at the interface. As the composite fraction increases, stress level of the composite region rises. The opposite is true for the CVD-SiC monolith. This indicates varying importance of composite and monolith failures on the cladding-load failure probability $P_{f,clad}$, as shown in Eq.(2) [13]

$$P_{f,clad}=1-(1-P_{f,monolith})(1-P_{f,composite}) \quad (2)$$

where $P_{f,monolith}$ is the monolith failure probability, and $P_{f,composite}$ is the composite failure probability. According to the norm of the material usage, a design that leaves the monolith region under compression is preferable. Yet as shown in Fig.2, such designs inevitably increase the composite stress level, which might result in a cladding load-failure caused by composite fracture. An optimal balance between the monolith and the composite fraction should be sought in terms of statistical failure probabilities. The sole monolithic SiC cladding gives the lowest peak stresses among the other cladding composition options because of the high thermal conductivity and the absence of discontinuous stresses. Yet, the lower stress level does not mean that its structural integrity is more robust than the other options. That, the obtained stress distributions should be translated into statistical likelihood of failure.

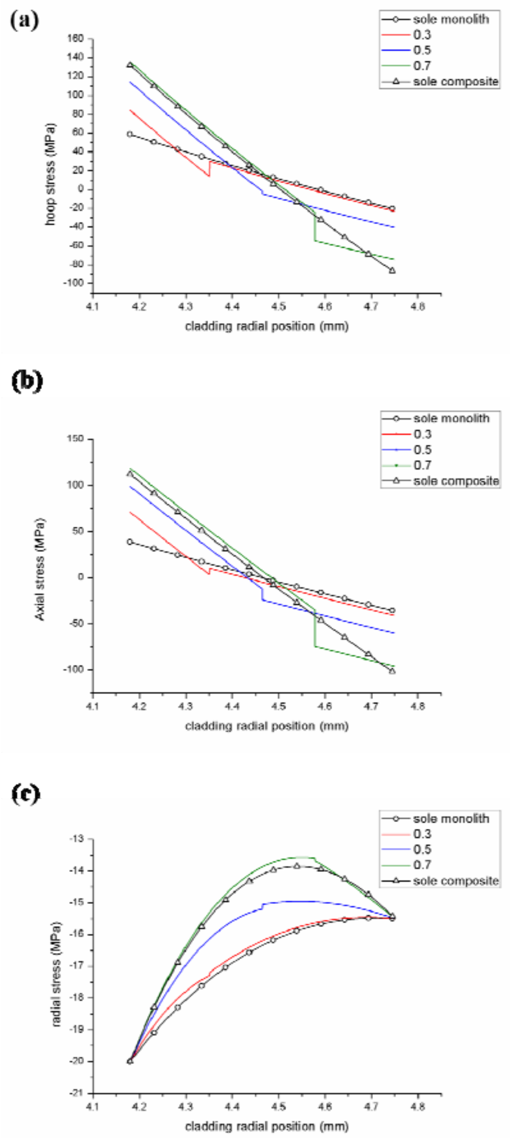

Fig.2 Radial stress distribution of SiC cladding in EOL: sole monolith, sole composite, and duplex cladding with composite layer fraction of 0.3, 0.5, and 0.7.
(a) hoop, (b) axial, (c) radial stresses

The multi-layered cladding structural analysis code [13] uses Weibull statistical fracture model to calculate material fracture probabilities. Fig.3 shows cladding failure probabilities with respect to different composite layer fractions. The cladding load failure probability ($P_{f,clad}$) is calculated with the presented composite ultimate fracture probability ($P_{f,composite}$) and the monolith fracture probability ($P_{f,monolith}$) according to Eq.(2). For the inner composite layer fraction below 0.4, the cladding load failure probability occurs because of the fracture of the monolith ($P_{f,clad} \sim P_{f,monolith}$). For those cases, the composite ultimate fracture probability is below ~10^{-7}, from which no appreciable contribution to cladding load fracture can be found. For the composite fraction greater than 0.4, the cladding load failure probability is dictated the composite ultimate fracture ($P_{f,clad}$ ~ $P_{f,composite}$). For those designs, the fracture initiated by the monolith is nominally zero as it is under compressive stresses.

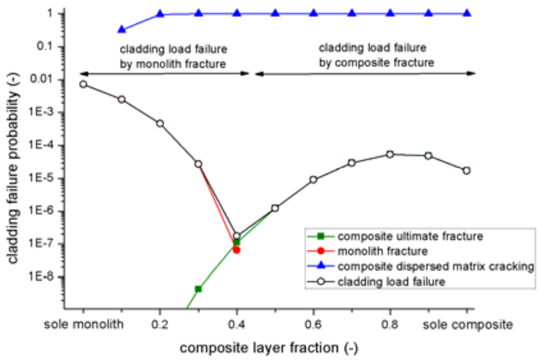

Fig.3 SiC cladding failure probabilities with different SiC_f/SiC composite fractions. Cladding load failure probability is obtained using Eq.(2)

It is noteworthy that in Fig.3 the composite dispersed matrix cracking probability is close to unity for the entire range of the composite layer fractions, which significantly challenges heremeticity of fission gases. This clearly indicates the need of the monolith (CVD-SiC) outer layer for the SiC cladding design even though the load fracture probability of the sole composite cladding is considered to be acceptable (~10^{-5}). The sole monolithic design shows the cladding failure probability close to ~0.01, which would result in an unacceptable number of failed fuel rods in steady state. Given that the dispersed matrix cracking is anticipated with high assurance, the optimal duplex cladding design should be determined to minimize the cladding load failure probability. The optimal composite layer fraction is found at ~0.4 where both the monolith and the composite relatively equally contribute to the cladding load-failure probability.

3.2. Burnup-informed cladding optimization

SiC cladding is subject to burnup-dependent stress levels in steady-state primarily with the internal fuel rod pressurization, for which the cladding design should take into account. Fig.4a shows the cladding failure probability with the different level of fuel rod internal pressurization (thickness 0.57mm). One can note overtaking of cladding failure probabilities as the fuel rod internal pressure increases. That, a cladding with a thicker SiC_f/SiC fraction is more advantageous for the state of a higher fuel rod internal pressurization, hence high burnup operation. Yet, an unaccounted fact in Fig.4 is that a higher SiC_f/SiC fraction decreases the cladding thermal conductivity, which would accelerate fuel rod internal pressurization by an increase of fission gas release at a higher fuel pellet temperature. The reference internal pressurization of SiC clad fuel rod at EOL is 20MPa for 67MWd/kgU. At 20MPa, cladding load-failure probability is minimized with SiC_f/SiC fraction ~0.4. A few designs can be altered from the reference fuel rod design of LWRs to reduce fuel rod internal pressurization of SiC clad fuel rod. A previous study [25] by Lee et al., found that UO_2 pellet with 10% central void, and increasing plenum length up to 3 times the current fuel design could be potentially allowable. With those design alteration, fuel internal pressure could decrease to the range of 10~15MPa. For such cases, the composite fraction ~0.4 is considered an acceptable design as it gives cladding load failure probability below 10^{-9}. Note that for such low fuel failure probability ranges, an optimization effort of SiC cladding would not lead to an appreciable gain in reducing failed fuel rods.

Because of manufacturing difficulties for thin multi-layered SiC cladding, 0.8 mm of the SiC cladding thickness is considered to be a near-term viable option [12,25]. Fig.4b shows the cladding load failure probability for 0.8mm thickness. Compared to 0.57mm thick cladding (Fig.4a), failure probability of 0.8mm cladding increases more slowly with respect to increasing fuel rod pressure. This is because a thicker structure is far more tolerable to pressure loading because it effectively lowers stresses due to a pressure difference. Hence, as can be seen in Fig.4, the overtaking of fuel rod failure probabilities for 0.8mm thick cladding is not as rapid as those of 0.57mm, with increasing fuel rod pressure. From the design point of view, this implies that uncertainties in fuel rod internal pressure on cladding design diminish as the cladding thickness increases. For 0.8mm thick cladding, the composite fraction of 0.3 is found to be an optimized value (Fig.4b).

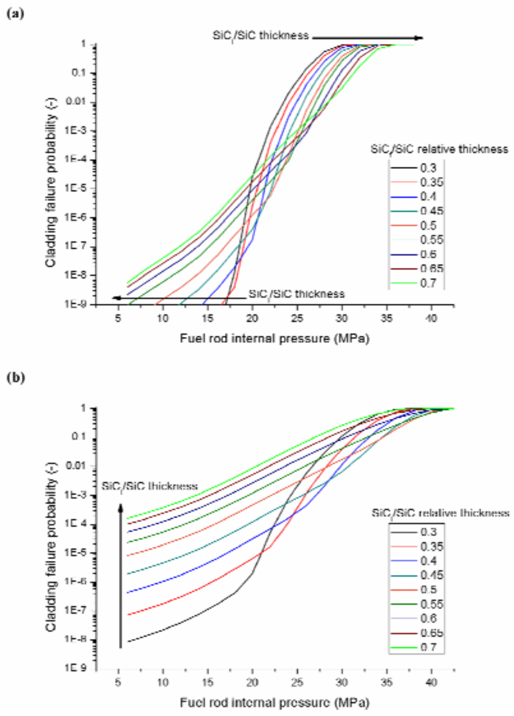

Fig.4 Cladding load fracture probability with different levels of fuel rod internal pressurization: reference case, cladding thickness (a) 0.57mm, (b) 0.8mm

3.3. Cladding designs with possible departures from the reference cladding properties

A robust design should take into account uncertainties of nominal cladding properties. Properties of nuclear grade CVD-SiC are known with high certainties [4], from which no considerable departures from the reference values are expected. SiC_f/SiC, however, is submissive to property variations depending on the fiber-architecture. Ultimate fracture Weibull modulus and thermal conductivity of SiC_f/SiC are considered to be the material properties of the greatest uncertainties [13,15,17,25]. The reference Weibull modulus of the nuclear grade SiC_f/SiC is 17.5. Yet, an experimental investigation shows its value as low as 12.5 [17]. A lower Weibull modulus increases statistical uncertainties for fracture, which is undesirable from the design perspective. With the extensive number of fuel rods ~50,000 in a PWR core, a small increase in statistical uncertainty would lead to an appreciable increase in the number of failed fuel rods. The composite fraction ~0.4 gives the minimum cladding failure probability at 20MPa for the reduced Weibull modulus (12.5) as shown in Fig.5a. Yet, its failure probability (~1.1 x 10^{-4}) is far greater than that of the reference Weibull modulus case (~1.8 x 10^{-7}). For the thicker cladding (0.8mm), the reduced Weibull modulus value results in a marked increase in the fuel rod failure probability. For the 0.8mm cladding thickness, the composite fraction ~0.3 is found to be the optimal design for the reference case (P_i =20MPa).

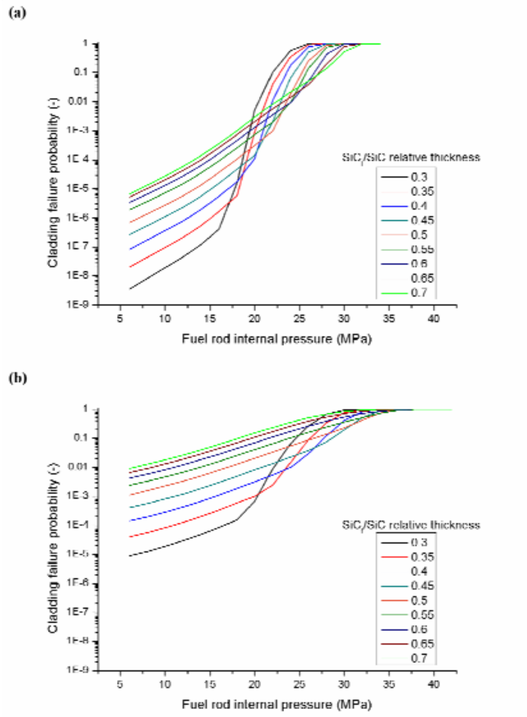

Fig.5 Cladding load fracture probability with different levels of fuel rod internal pressurization: composite ultimate fracture Weibull modulus (m) change from 17.5 to 12, cladding thickness (a) 0.57mm, (b) 0.8mm

We used a conservative thermal conductivity value for SiC_f/SiC (~1.5W/m-K). Yet, a few other examinations report a higher irradiation-saturated thermal conductivity of SiC_f/SiC, around ~3.0 W/m-K [13,17]. Fig.6 shows cladding failure probabilities with the increased thermal conductivity of SiC_f/SiC (~3.0 W/m-K) for different composite layer fractions. One can note that for the 0.57 mm cladding thickness, the thermal conductivity increase remarkably decreases the cladding failure probability. This is because of the flattened radial temperature gradient which gives a lower level of thermal and swelling-induced stresses. As can be noted in Fig.6a, the composite fraction ~0.65 gives the cladding load fracture probability ~10^{-9}, practically eliminating steady-state load fracture issues. It is noteworthy that the observed overtaking of cladding failure probabilities do not appreciably take place for the increased composite thermal conductivity, as shown in Fig.6. This is because the higher thermal conductivity works particularly favorably for structural integrity of SiC_f/SiC, which is most challenged by swelling-induced stresses due to its low thermal conductivity. That is, as SiC_f/SiC thermal conductivity becomes closer to that of CVD-SiC, use of SiC_f/SiC

becomes more advantageous than that of the monolith from the structural integrity point of view. In such a case, the outer CVD-SiC layer should assure enough thickness for volatilization of the material in steam oxidizing environments. For the 0.8mm cladding thickness, SiC_f/SiC fraction ~0.45 is found to be an optimal value.

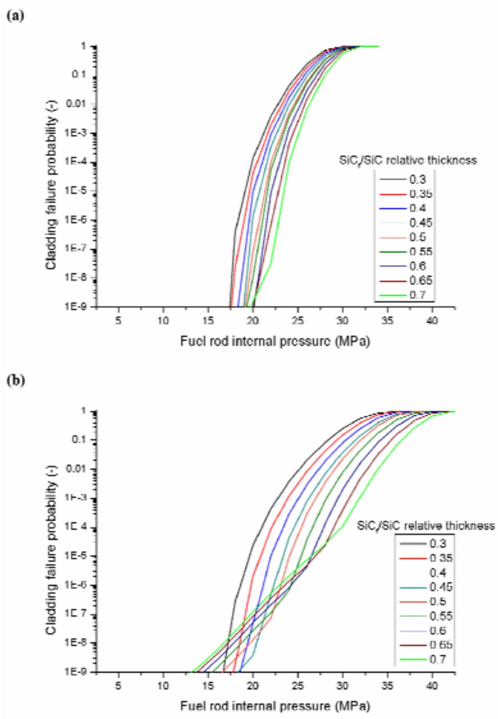

Fig.6 Cladding load fracture probability with different levels of fuel rod internal pressurization: composite thermal conductivity change from 1.5 to 3.0 W/m-K, cladding thickness (a) 0.57mm, (b) 0.8mm

4. Conclusions

The following points summarize key conclusions of this work:

- Duplex SiC cladding failure probability and its feasibility are remarkably affected by its cladding composition design.
- The sole monolithic CVD-SiC cladding is not feasible because of its unacceptably high fracture probability in steady-state operation (~10^{-2}).
- The sole composite SiC cladding is not feasible because of the dispersed matrix cracking which challenges fission gas tightness of the cladding.
- The duplex cladding with the inner composite fraction ~0.4 is found to be the optimal SiC cladding design.
- The presented methodologies can be used to optimize SiC cladding designs with different incore operating conditions and material properties.

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