Development of SiC/SiC Composite for Fusion Application

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Abstract The recent efforts to develop SiC/SiC composite materials for fusion application under the collaboration with Japan and the USA are provided, where material performance with and without radiation damage has been greatly improved. One of the accomplishments is development of the high performance reaction sintering process. Mechanical and thermal conductivity are improved extensively by process modification and optimization with inexpensive fabrication process. The major efforts to make SiC matrix by CVI, PIP and RS methods are introduced together with the representing baseline properties. The resent results on mechanical properties of SiC/SiC under neutron irradiation are quite positive. The composites with new SiC fibers, Hi-Nicalon Type-S, did not exhibit mechanical property degradation up to 10 dpa. Based on the materials data recently obtained, a very preliminary design window is provided and the future prospects of SiC/SiC technology integration is provided.
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1. Introduction

As the key material to make attractive fusion reactors with sufficient competitiveness with other power options, safety and reliability, silicon carbide (SiC) fiber reinforced SiC matrix (SiC/SiC) composites have extensively been studied and there have been significant progresses in developing SiC/SiC composites. The attractiveness of SiC/SiC for fusion applications mainly comes from its high temperature strength, pseudo-ductile fracture behavior and inherent low-induced radioactivity under fusion environment [1,2]. In this paper, recent accomplishment in SiC/SiC studies in Japan/USA collaboration program (JUPITER program) and CREST-ACE (Core Research for Evolutional Science and Technology, Advanced Composite Systems for Energy Conversion) program are presented.

2. SiC Fiber Developments

Although composite properties are not controlled only by fiber properties but by several factors, such as volume fraction of fibers, fiber/matrix interface structure, fiber architecture and matrix properties, there are key fiber properties that control radiation resistance, high-temperature strength and stability and weaveability. As reinforcements, continuous SiC fibers derived from polymer precursors, like Nicalon and Tyranno have been utilized for SiC/SiC R & D international. Not only from fusion application but also from aerospace applications, fiber improvements have been carried out in these decades. Figure 1 provides improvements of high temperature heat treatment resistance in tensile strength of polymer driven SiC fibers. From Nicalon fiber to Tyranno-SA fibers, high temperature

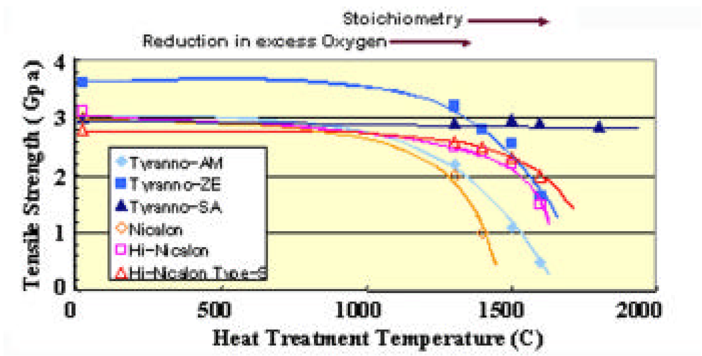


Fig.1: Stability of tensile strength under high temperature treatment

degradation of fiber strength has been improved from about 1000C to more than 1800C in air for 1 hour. This improvement was made, as shown in Fig.1, by reduction of remaining free oxygen, followed by the reduction of excess C to stoichiometry and to make high crystallinity fibers. The recent results indicate the extensive improvement of creep strength and of resistance in radiation damage and in oxidation environments seen in those advanced fibers [3,4].The transverse thermal conductivity of SiC/SiC composites reinforced with Nicalon™ CG fibers (a first generation SiC-based fiber with a mostly amorphous type structure) is about 10 and 7 W/mK at 300K and 1273K, respectively. These values are insufficient to meet the anticipated thermal conductivity requirement for current fusion reactor designs which is 30 W/mK (unirradiated value) or larger. Higher composite thermal conductance properties have been obtained by making composites with higher purity and more crystalline, low porosity SiC matrices and with improved fiber-matrix interfaces. As new, near-stoichiometric and crystalline SiC fibers are integrated into composite structures, further improvements are likely. To illustrate the progress being made in improving fiber thermal conductivity values, in Figure 2 the measured thermal conductivity values for several advanced SiC fiber types (Hi-Nicalon™, Hi-Nicalon™ type S and Tyranno™ SA-B) are compared to the values for Nicalon™ CG fiber and for dense, high-purity beta-SiC made by the chemical vapor deposition (CVD) process [5]. The thermal conductivity values for the CVD-SiC material represent the upper limit achievable for a SiC-based component. These values are approximately two orders of magnitude larger than the values for the Nicalon™ CG fiber. Likewise, the thermal conductivity values for the Tyranno™ SA-B and Hi-Nicalon™ type S fibers are more than an order of magnitude higher. Apparently, higher thermal conductivity values are obtained for the advanced SiC fibers that have crystalline structures with larger grain-sizes.

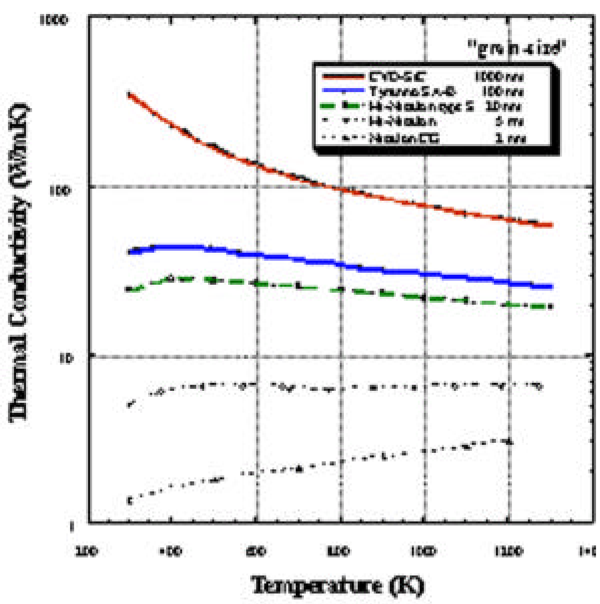


Fig. 2: Comparison of measured thermal conductivity values from 300K to 1300K for Nicalon™ CG, several advanced SiC-based fibers and high-purity, monolithic CVD-SiC.

Furthermore, the new SiC fibers promise to be more radiation resistant, in which case the rather large differential swelling/shrinking observed between the fiber and matrix components in first generation composites made with Nicalon™ CG fibers should be considerably alleviated [6]. Improved dimensional stability of the fibers, in turn, should lead to improved fiber/matrix interface conductance in SiC/SiC composites. By using such new fibers and by careful design of the fiber/matrix interface, the opportunity to make significant improvements in the thermal conductance of SiC/SiC composites now exists.

3. SiC/SiC Composite Process Development

As potential fabrication processes of SiC/SiC composites for fusion applications, forced chemical vapor infiltration (F-CVI), Reaction sintering (RS), liquid phase sintering (LPS), polymer impregnation and pyrolysis (PIP) and their combined processes have been developed in these decades. Although these efforts utilized the knowledge and technology for non-fusion application, there has been a strong incentive to make neutron radiation damage resistant and high thermal conductivity composites [3].

Firstly, the high performance SiC by reaction sintering (RS) method, which has been known as an inexpensive fabrication process but with inferior performance, becomes available by process modification and optimization with the same production cost. The representative mechanical and thermal properties of the improved RS-SiC are shown in Fig. 3. By the new RS method, extensive improvements in both strength and thermal conductivity are accomplished, as indicated as CREST-ACE accomplishment [7]. This improvement is due to the reduction in residual Si value and fine dispersion of the residual Si as shown by optical micrographs in the figure. Another important improvement recently made is the utilization of very fine SiC powder as filler to performs. By changing the SiC filler powder size to 1um with 10% of residual Si, the thermal conductivity is improved to be as high as 170 W/mK [8]. Put these efforts together, it may be possible to make SiC with high strength and high TC indicated as a pink line in the figure.

Another example to obtain high quality SiC matrix is shown in Fig. 4, for the case of

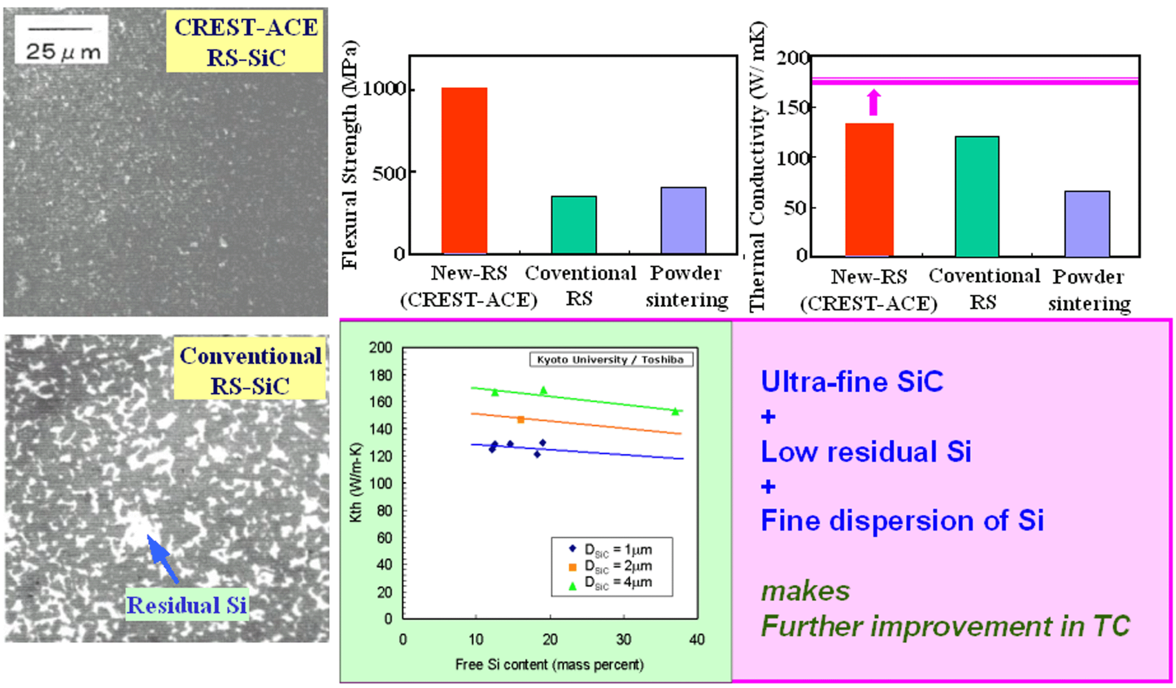
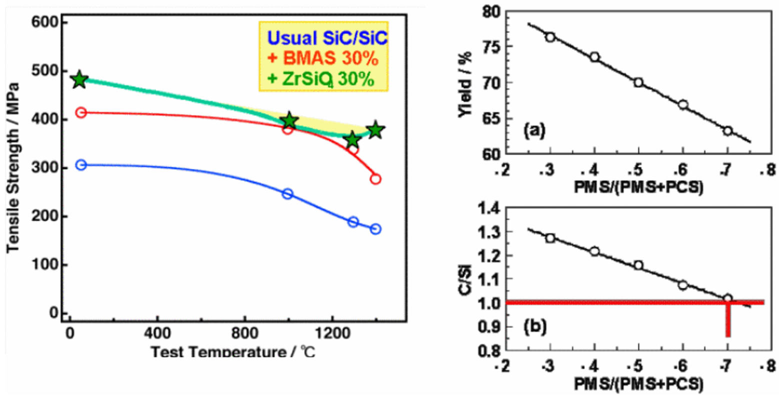


Fig.3: Improvements in thermal conductivity and strength SiC by RS process modification

PIP process. The effort to improve strength is shown as (a), where small amount of oxide addition is effective not only room temperature strength but rather effective at higher temperature. Especially for the case of 30% $ZrSiO_4$ addition, strength at 1400C still stays 400Mpa, which is more than two times higher than the SiC without oxide addition. In general, simple PIP process makes non-stoichiometry SiC and the deviation from stoichiometry becomes the origins of high temperature strength degradation, lower radiation resistance and lower oxidation resistance and others. Therefore, many efforts have been done and the success is seen in new generation stoichiometry fibers. (b) of figure 4 is an example of stoichiometry SiC matrix obtained by blending polymers for PIP [9]. By the appropriate blending of polymethylsiren (PMS) and polycarbosiren (PCS), shown red lines as the example, near stoichiometry SiC is obtained. The selection of non-PCS polymer for matrix, such as poly-vinyl-siren (PVS), is indicating a good fiber-matrix interface with PCS-SiC fibers, like Nicalon-family fibers, where appropriate fiber pull-outs after bending test were obtained without any fiber surface coating [10]. Although PIP process is still inexpensive process but the quality becomes quite high. Thus, the improved PIP process, with enough potential to make complex and large component, is an attractive option to make SiC/SiC composite for fusion.



(a) Effect of oxide filler addition to matrix on high temperature strength

(b) Effect of polymer blending on controlling C/SiC ratio in matrix

Fig.4: Improvements in PIP process

The improvement of SiC fibers, like Tyranno-SA and Hi-Nicalon type-S, in high temperature resistance opens wider capabilities in production process options. This is to apply higher temperature for processing to make composites, which enable to obtain high crystallinity matrix. By using Tyranno-SA fibers to PIP SiC/SiC production with final process temperature at 1600C, high thermal conductivity SiC/SiC has been successfully produced [11].

There has been many extensive efforts to improve SiC/SiC properties by F-CVI process at NRIM, Japan, and ORNL, USA under the CREST-ACE program and the Japan-USA collaborative

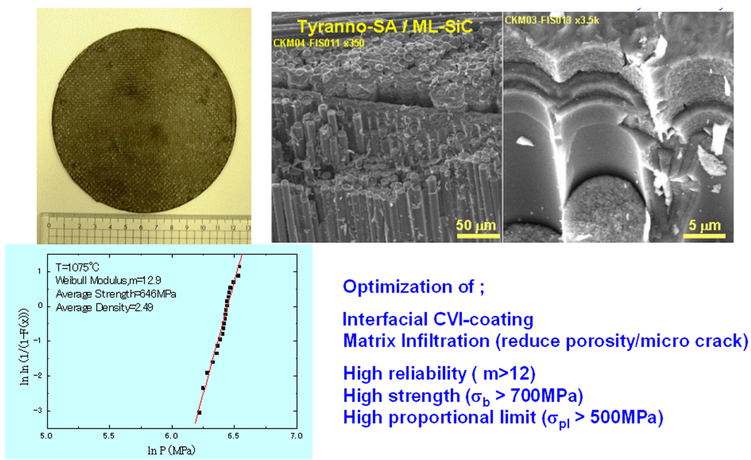


Fig. 5: Forced CVI Process Improvement

program, JUPITER program, in these years. One of the recent accomplishments is shown in Fig. 5. The direction of improving properties has been to optimize interfacial structure and to improve matrix density and its microstructure and 110mm diameter SiC/SiC disk fabricated at NRIM is shown in the figure, where multiple coating of SiC and C on Tyranno-SA fiber presents high flexural strength (S_f =723MPa), high proportional limit (S_{pl} =503MPa) and high reliability for fracture (indicated as Weibull modulus, m; m>12)[12].

4. Radiation Damage in SiC/SiC Composites

In fusion reactor environment, nuclear collisions and reactions by high-energy neutrons and particles from fusion plasma have strong impacts on materials through the production of displacement damage and transmutation damage. Degradation of material performance such as mechanical properties, thermal properties, etc., has been recognized as the key issue and extensive efforts have been conducted.

The major and the current concerns relating with SiC/SiC composites are mechanical property degradation, such as static tensile properties, static bending properties, dynamic strength properties like creep, fatigue and creep-fatigue, shape and phase stability, including swelling, under displacement damage. He transmutation effect together with displacement damage on strength and fracture toughness, fatigue, oxidation at very low O_2 and H_2O pressure, microstructural and microchemical instability at very high temperature are issues to be evaluated and be solved.

The one of the difficulties lies in radiation damage study is the insufficient accuracies of radiation condition measurement and of radiation damage condition control which has been limiting the interpretation of radiation damage mechanisms and even establishing the mutual basis to see the radiation effects in neutron irradiations. Another important difficulty has been the limitation of microstructure and mechanical property evaluation methods, especially for SiC/SiC composites. The recent advancements in multiple-beam irradiation facility, like DuET facility of Kyoto University [13] and in FIB (focused ion beam) micro pickup method for TEM inspection. Our strategy is to obtain basic understandings through well controlled and well-characterized irradiation experiments, like the one in DuET facility, and apply the knowledge to the most advanced neutron irradiation experiments (under the state of the art technology in irradiation condition control and characterization and in materials to be tested). For this purpose, DuET facility, Kyoto University, has been

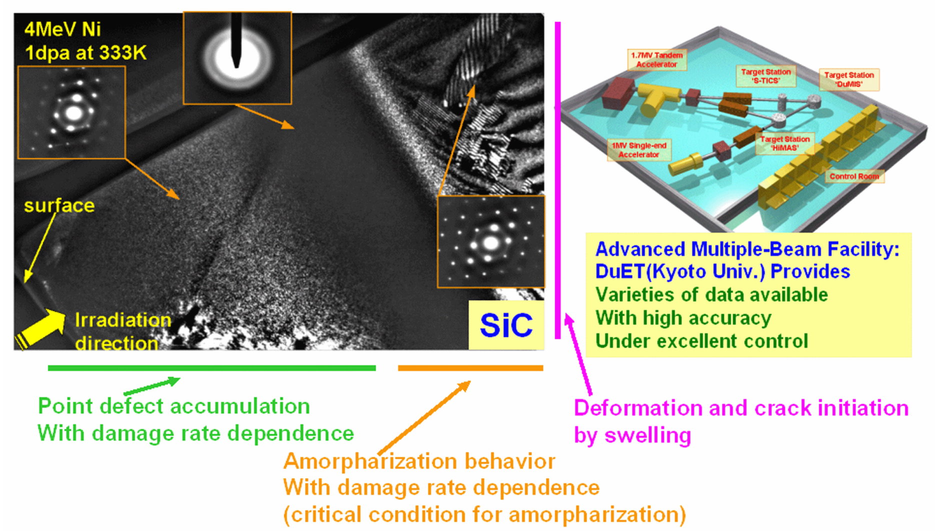


Fig.6: Advanced Multiple Beam Irradiation Experiment at DuET, Kyoto Univ.

recently established. Figure 6 is an example to give the outline of the DuET facility and a typical example of varieties of damage data from just one small sample. DuET facility has two main accelerators and three test chambers for experiments with many in-beam analysis capabilities and radiation condition monitoring and controlling devises. A TEM micrograph shows the data available from a tiny region, where point defect accumulation with damage rate dependence in crystalline phase, amorphization behavior from crystalline phase with damage rate dependence (critical condition for amorphization can be obtained) and even deformation and crack initiation caused as the result of a large swelling by amorpharization can be quantitatively defined. Although, He and H transmutation effects are not indicating in this figure, dual-ion irradiation can produce synergistic effect of displacement damage and He or H transmutation effect can be studied at DuET facility. Swelling in SiC has been an important subject to be understood for many decades and there has been a serious discussion on high temperature swelling around 1000 C where differences in irradiation conditions, other than irradiation temperature, made it difficult to establish a mutual basis. As shown in Fig. 7, the recent data from DuET experiment clearly demonstrate the negative temperature dependence of swelling and the monotonic decrease of swelling up to 1200 C and almost negligible He effect can be interpreted [14]. Also, the recent effort on in-reactor measurement of thermal conductivity degradation in SiC, SiC/SiC and other ceramics, TRIST-TC1 experiment under JAERI-ORNL and JUPITER Program, provided an important information about 50% of thermal conductivity degradation at 700C, which may suggest that neutron irradiation is not a major issue in a high temperature gas-cooled solid blanket [15]. An example of radiation induced shape instability is shown in Fig. 8, where a large fiber shrinkage, for the case of old SiC fibers, caused a inner laminar cracking to change a plate specimen to lens shaped specimen. Whereas, for the case of advanced stoichiometry SiC fibers, no dimensional change was observed under 1 dpa neutron irradiation in JMTR at 300-500 C. The same behaviors can be seen in the samples

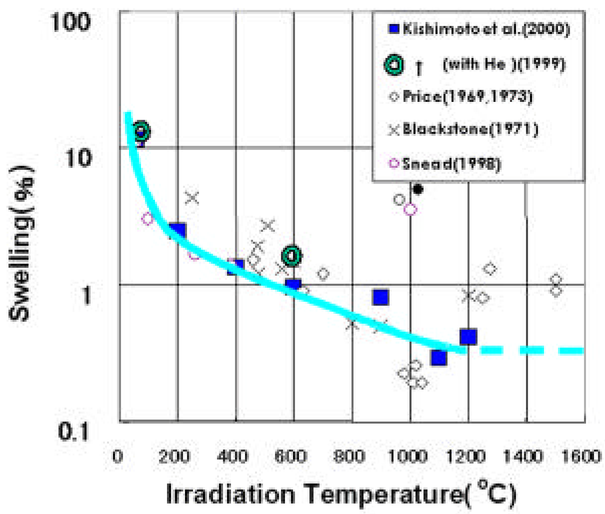


Fig. 7: Temperature dependence of swelling in SiC

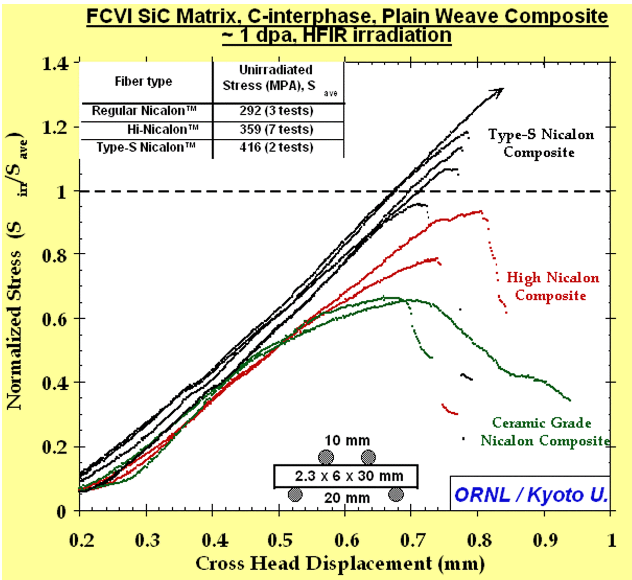


Fig. 8: Effect of neutron irradiation on bending strength

from HFIR irradiation up to 10 dpa and from JOYO up to 5 dpa (these are the only available data currently). These neutron-irradiated samples were tested by four-point bending test method, as shown in Fig. 8 together with the test result. As is clearly seen, the samples with the first generation fiber, Nicalon commercial grade, degraded greatly and the samples with the second generation fiber, High-Nicalon, only slightly degraded for 1 dpa irradiation in HFIR which is the same as the results from JMTR irradiation. However, for the case of the advanced fiber, Hi-Nicalon type-S, no mechanical property degradation can be seen or a slight increase in fracture strength can be detected [16]. The flexural strength data from neutron irradiation are summarized in Fig. 9, including the very new data from HFIR irradiation to 10 dpa. The data are normalized as the ratio of strength after neutron irradiation with the one before irradiation. The data for the first-generation fibers, CG-Nicalon, and the second-generation fibers, Hi-Nicalon, have a similar trend to decrease monotonically from 1 dpa to 10 dpa and to saturate up to almost 100 dpa. However, for the case of the advanced fibers, the strength stays or slightly increased with increasing total damage up to 10 dpa. The fracture behavior seen from fracture surface stays the same for the case of the advanced fibers. This may suggest the strong radiation resistance in strength and shape stability in composites with stoichiometry fibers and stoichiometry matrix with appropriate interfacial structure [17].

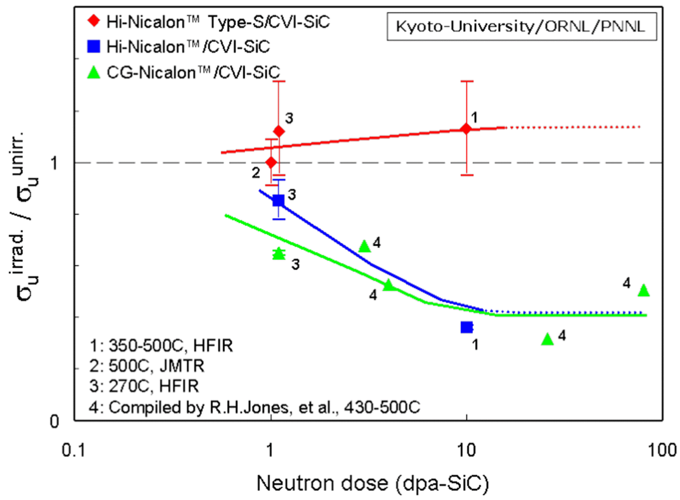


Fig. 9: Influence of neutron irradiation on flexural strength of CVI-SiC composites with Nicalon™-family SiC fibers

5. Limiting Factors of SiC/SiC Composites for Fusion Applications

As described the above, both the baseline properties and the neutron irradiation performances of SiC/SiC are drastically improved through the recent efforts in developing high performance composites. Based on the materials data recently made available, a very preliminary design window is provided, as shown in Fig. 10 [18]. This design window has been greatly improved from the previous data set compiled at the first IAE SiC/SiC working group meeting in 1996. Further efforts are being made for studying the remaining technical issues, such as helium effect on strength,

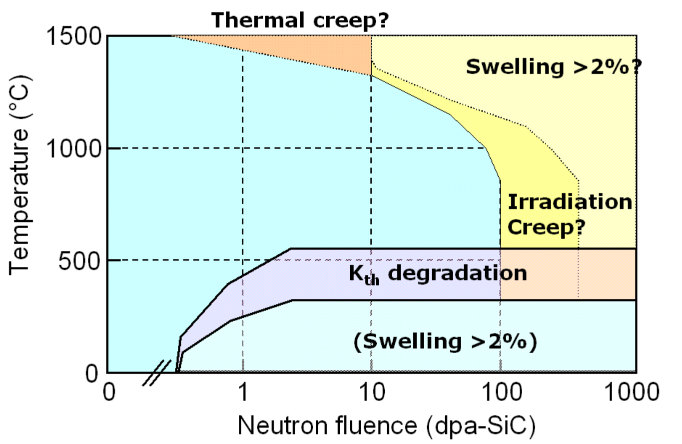


Figure 10: A very preliminary design window for SiC/SiC

fracture toughness, fatigue, oxidation at very low P_{O2} and P_{H2O} and grain growth effect at high-temperature, in order to provide a solid and reliable design window for fusion structural applications. Although, technology integration issues are not mentioned here due to the limitation of length, joining [19], hermetic coating and component fabrication technology developments are on-going. Including these efforts, there is an attempt to promote technology integration toward fusion reactor engineering and design. A typical example is the application to blanket, where compatibility with breeders and neutron multipliers under He purge gas and thermal and mechanical stresses are being studied. This is also going to be one of the emphases in the next Japan-USA collaboration program, JUPITER-II from 2001 to 2006.

6. Summaries

There has been a great improvement in SiC/SiC performance for fusion application, under CREST-ACE and JUPITER programs. The materials performances will be steadily improved and technology integration will be accelerated.

Irradiation effects of SiC/SiC are extensively being studied using fission reactors of various types and newly developed multi-beam accelerator facilities. There are significant improvements in radiation resistance of SiC/SiC and in understanding radiation effect mechanisms.

Current status and future prospects of R&D and/or application activities of SiC/SiC composites make SiC/SiC more attractive and realistic for fusion reactor application.

References

[1] A. Kohyama, H. Matsui and A. Hishinuma, Proceedings of 10th Pacific Basin Nuclear Conference(1996)883-889.

[2] P. Fenici, A. J. Frias Rebelo, R. H. Jones, A. Kohyama, L. L. Snead, J. Nucl. Mater., 258-263(1998)215-225.

[3] A. Kohyama, M. Seki, K. Abe, T. Muroga, H. Matsui, S. Jitsukawa, S. Matsuda, , J. Nucle. Mater.,(2000) in press.

[4] H. Serizawa, A. Kohyama, D.E. Johnston, G.E. Youngblood, R.H. Jones and C.A. Lewinsohn, , Ceramic Engineering & Science Proceedings, Vol.20 Issue 4(1999)443-450.

[5] G.E. Youngblood, R.H. Jones, W. Kowbel, Reiji Yamada, Paul de Heij, Toshihiro Ishikawa and Akira Kohyama," to be presented at the 4th IEA SiC/SiC Workshop, Frascati, Italy, October 12-13, 2000.

[6] G.E. Youngblood, R.H. Jones, Akira Kohyama and L.L. Snead J. Nucl. Mater. 258-263 (1998) 1551-56.

[7] S.P. Lee, Y. Katoh, W. Zhang, M. Kotani, A. Kohyama and S. Suyama, Proceedings of the Second Asian-Australasian Conference on Composite Materials (ACCM-2000), Vol.I, p.p.651-656. (2000).

[8] S. P. Lee, Y. Katoh, T. Hinoki, M. Kotani, A. Kohyama, S. Suyama and Y. Itoh, Ceramic Engineering & Science Proceedings, Vol.21 (2000) in press.

[9] M. Kotani, A. Kohyama, K. Okamura and T. Inoue, Ceramic Engineering and Science Proceedings,Vol.20, Issue 4, p.p.309-316 (1999).

[10] W. Zhang, Y. Katoh and A. Kohyama, Proceedings of the Second Asian-Australasian Conference on Composite Materials (ACCM-2000), Vol.II, p.p. 759-764(2000).

[11] Y. Katoh, T. Hinoki, T. Shibayama, A. Kohyama and H. Takahashi, Ceramic Engineering & Science Proceedings, Vol.20 Issue 4, p.p.325-332(1999).

[12] W. Yang, H. Araki, A. Kohyama, T. Noda, Ceramic Engineering & Science Proceedings (2000) , in press.

[13]A. Kohyama, Y. Katoh, M. Ando and K. Jimbo, Fusion Engineering and Design (2000) in press.

[14]H. Kishimoto, Y. Katoh, A. Kohyama and M. Ando, Proceedings of the Second Asian-Australasian Conference on Composite Materials (ACCM-2000), Vol.II, p.p.733-738(2000).

[15]R. Yamada, S. Zinkle, L.L. Snead, Y. Katoh, J. of Nucl. Mater., (2000) in press.

[16] Y. Katoh, T. Hinoki, A. Kohyama, Ceramic Engineering & Science Proceedings, Vol.21 (2000) in press.

[17] L.L. Snead, Y. Katoh, A. Kohyama, R. Shinavski, A.M. Williams, J.L. Bailey, N.L. Vaughn and R.A Lowden, Journal of Nuclear Materials, in press.

[18] T. Shibayama, H. Takahashi, G.W. He, Y. Katoh and A. Kohyama, Ceramic Engineering & Science Proceedings, Vol.20 Issue 4 p.p.161-168. (1999).

[19] M. Ferraris, M. Salvo, C. Isola, M. Appendino Montorsi and A. Kohyama, Journal of Nuclear Materials 258-263 (1998) 1546-1550.