13<sup>th</sup> International Workshop on Spallation Materials Technology 31<sup>st</sup> Oct., 2016@ Chattanooga

#### Investigation of target material for muon production under high power proton beam irradiation



J-PARC Center, High Energy Accelerator Research Organization, KEK

Shunsuke Makimura

**Challenger: silicon carbide** 

against king of Low-Z target material, polycrystalline graphite



#### Many thanks to everyone, (^\_^)/

## **Collaborators**



This program for SiC/SiC is supported by JSPS Kakenhi "16Ho3994".

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## **Graphite Target at J-PARC and RCNP/Osaka**



Muon Rotating target at MLF 3 GeV, 1 MW, thickness 20 mm Thermal radiation in vac. 950 Kelvins @ 1MW

Neutrino target 30 GeV, 750 kW, thickness 900 mm He-cooling: 1010 Kelvins @ 750 kW

> Graphite target IG-430U 26mmf x ~900mm Inner tube (graphite)

> > Outer tube / beam window (Ti-6Al-4V)

MuSIC target at RCNP 400 MeV, 400 W, thickness 200 mm Thermal rad. In vac. 600 K

**COMET** target 8 GeV, 4 kW, thickness 400 mm Thermal rad. In vac. 500 K

MuSIC target



## Polycrystalline graphite

King of Low-Z target material, especially for muon/pion production

#### <u>PROS</u>

- High thermal resistance (1600 degC @Vac.)
- Mech. properties (Low Young's modulus, Low thermal expansion, <u>High resistance to thermal shock</u>)
- Experience as irradiation material (Nuclear fission reactor)

#### <u>CONS</u>

- Low density (Volume of muon/pion source should be small for efficient transport.)
- <u>Easy oxidation at high temperature</u> (Unexpected air introduction)

Silicon carbide (SiC) as a new candidate of target material



## WHY SILICON CARBIDE ?

#### Comparison with graphite

What kinds of SiC will be investigated?

#### <u>PROS</u>

- Density: 1.76 times higher than graphite (3.2 g/cc vs 1.82 g/cc), Higher efficiency of transportation due to small spatial volume of pion/muon source
- (2) High oxidation resistance ~ dependence on partial pressure of oxygen, Protection against unexpected air introduction
   <u>CONS</u>

③ Higher residual radiation dose, Gamma emitter SiC has 8 times larger risk for thermal stress. (Large Young's modulus and beam loss), Furthermore, SiC is brittle.

Monolithic silicon carbide cannot be used as target material.

SiC coated graphite

1: Not satisfied, 2: Satisfied

3: Not so bad



SiC composite material (NITE-SiC/SiC)

1: Satisfied, 2: Satisfied

3: Very bad

JSPS Kakenhi

## SiC coated graphite & NITE SiC/SiC

#### SiC coated graphite

- Commercially available at Graphite manufacturers
- CVD-SiC coating (Dense coating)
- Study for fission nuclear reactor with higher oxidation resistance

#### **NITE SiC/SiC**

Nano-powder Infiltration and Transient Eutectic

- Developed at Muroran T. I./OASIS
- Composed of SiC-fiber and matrix
- Controllable thermal & mechanical properties
- Pseudo ductility
- **D** Resistance to irradiation



OASIS 1/3 model for muon target



Neut

Press

BWR

Neutr

Fuel Shroi

Dowi Diffe

meas Inlet thern

Irradiation tests at Harden/ Norway

### PROS 1

## <u>More efficient transport of Pions/Muons</u> <u>due to higher density</u>

Meaning of existence for Accelerator Target 3.2 g/cc vs 1.82 g/cc

From physics requirements, especially fundamental physics, the most important.
 If 1.3 times more efficient, 5-years experiment will be 4-years experiment.

Efficiency doesn't depend only on the density of target material but also on the purpose of physics, the dimensions, the beam energy, the optics of the transport and so on.

## Efficiency of SiC at MLF for DeeMe Project

by Aoki at Osaka Univ.

#### Muon-electron Conversion Search on the target material Representative; M. Aoki, Osaka University

 $\mu^{-} + A(Z,N) \rightarrow e^{-} + A(Z,N)$ 

- Forbidden in the Standard Model(SM) of particle physics.
- No signals found yet.
- Discovery of the signal
  - $\rightarrow$  a proof of the physics beyond SM.
    - Complementary to high-energy frontier experiments: LHC, ILC.
    - Can explain the neutrino oscillation phenomena (Seesaw Mechanism).

Efficiency strongly depends on the atomic number of target material in this experiment.



Efficiency of SiC will be 6 times larger than graphite!!







Proton beam line

1m

#### by Mihara at COMET/KEK.

## **COMET Pion Production Target**

- Base design of the target
  - 26mm (r) x 700 mm (L), Graphite for 8GeV 3.2kW proton beam
- SiC target (with same interaction length as Graphite) is expected to provide higher muon stopping rate due to the smaller target image at production
  - 30% increase of muon yield

Efficiency is not always increased. But in general, efficiency will be higher due to higher density of SiC.



## <u>PROS 2</u> Oxidation resistance of Silicon carbide

Accidental Loss of Vacuum during beam operation
 Loss of target material through O<sub>2</sub>, impurity during normal beam operation

Resistance to unexpected air introduction

Oxidation tests of NITE SiC/SiC are on-going by OASIS.

Oxidation tests of SiC coated graphite are planned by Hitachi co., Itd.

Background >
Oxidation of Graphite and SiC







<Current study: Under preparation>

# Verification of oxidation resistivity

- Objectives
  - Verification of oxidation resistivity of NITE-SiC/SiC composites under LOVA (Loss of vacuum accident).
    - Effects of oxygen concentration
    - Effects of test temperature
- Materials:
  - Carbon (T-6, IG-430U) as reference material
  - <u>NITE-SiC/SiC composites</u>
  - <u>CVD-SiC coated NITE-SiC/SiC composites</u>
- Experimental condition
  - Equipment: Infrared gold image furnace
  - Temp: ~ max. 1100℃
  - Env. gas : Air(O<sub>2</sub> 21vol%), N<sub>2</sub>+O<sub>2</sub> (O<sub>2</sub> 1vol%)
  - Gas flow rate: ~ max. 500 ml/min



<Current study: preliminary results>



## Oxidation test results (Graphite)



Time [min]

<Current study: Under preparation>





## Environmental Barrier Coating (CVD-SiC coated NITE-SiC/SiC tube)

For further improvement of oxidation resistivity of SiC/SiC composites



Preliminary PHITS simulations by Matoba

## <u>CONS 3</u> Residual radionuclides of SiC & SiC/SiC

Irradiation tests at RCNP (Low flux)
400 MeV, 1 uA, 1 hour
t = 1 mm, D = 10 mm, plate
Irradiation at MLF beam operation (High flux)
3 GeV, 333 uA, 5000 hours
t = 20 mm, D = 70 mm, plate

#### Produced radionuclides on simulation for RCNP

#### Low flux proton beam

Low flux proton bear	n			□ Half life
Dose rate @5 cm mSv/h	SiC	SiC comp. material SiC+Al 3%+Y 3%	Graphite	H3 : 12.3 years Be7 : 53 days
Fresh	3.65	3.80	1.35	C11: 20 minute
After 1 day	0.0417	0.104	0.00104	Na22: 2.6 years Na24: 15 hours
After 3 days	0.00625	0.0318	0.000938	Ge71: 11 days
After 7 days	0.00208	0.0104	0.000833	Sr83: 1.3 days

Radionuclides Bq	SiC		SiC comp. m SiC+Al3%+Y3	naterial %	Graphite		
Fresh	Total	4.11E+07	Total	4.11E+07	Total	1.98E+07	
	Si27	1.20E+07	Si27	1.25E+07	C11	1.77E+07	
	C11	9.90E+06	C11	8.33E+06	C10	1.09E+06	
	Al26m	5.73E+06	Al26m	5.73E+06	Be7	2.71E+05	
After 1 day	Total	4.01E+05	Total	9.90E+05	Total	2.76E+05	
	Na74	1.98F+05	Be7	1.88F + 05	Be7	<u>2 66E+05</u>	

Residual radiation dose of SiC is higher than Af graphite and is very slowly decayed due to the effect of Na22.

Af This will be confirmed with low flux proton beam at RCNP.

> 7.29E+03 H3 |

Sr87m | 1.61E+04

PHITS by Matoba

20 minutes 2.6 years 15 hours

+03

+05

+05

+03

+05

+05

+03

2.8 hours

PHITS by Matoba

MLF Target:
 Irradiation at MLF beam operation
 3 GeV, 333 uA, 5000 hours
 t = 20 mm, D = 70 mm, plate

	SiC	SiC+A13%Y3%	Graphite
Loss (W)	5773	5739	3096
Loss rate (W/cc)	75	75	40
DPA	43	48	5.1

Surface dose (5cm)						
	SiC	SiC+AlY	Graphite			
Fresh	2.5 kSv/h	2.6 kSv/h	763 Sv/h			

Residual radiation dose of SiC is much higher than graphite and is slowly decayed due to the effect of Na22. Effects of additives for composite material is not so large. These amounts will be validated at irradiation tests of RCNP. dominant for actual maintenance.



## <u>Plans for irradiation tests of the SiC coated</u> <u>graphite: Under RaDIATE collaboration & BLIP</u>

**Ra**diation Damage In Accelerator Target Environments

radiate.fnal.gov

Introduction by P. Hurh in this session



Purpose: Investigation of irradiation effects
 Irradiation at BLIP facility at BNL from next January.





SiC coated graphite will be included at CERN capsule.
 Confirmation through Microstructural analyses at PNNL whether exfoliation will happen.
 Comparison of three kinds of graphite

Precious opportunity for high-energy proton irradiation

By Claudio. L.T. Martin at CERN

## Thermal Analysis – Si Capsule

#### Temperatures



Max T Si samples: 216 °C Max T Graph/SiC: 220-240 °C Max T Sigraflex: 193 °C Max T SS window: 71 °C Max HF SS window-Water: 28 W/cm2



#### **Thermal Expansions:**

Initial lateral gaps Samples-Fillers = 0.1 mm Initial lateral gap Fillers-SS capsule = 0.2 mm

Remaining gap Graph/SiC –Filler: **94 um** Remaining gap Si samples – Si Filler: **80 um** Remaining Fillers– SS Capsule: **200 um** (remains the same)



\*Assumed TCC in Back-up slides

## Plans for irradiation tests of the SiC/SiC composite: at RCNP, Osaka Univ.





- Specimens can be handled by hands-on.
- Chemical analyses is available at RI building of RCNP.
- Transport from N0 beamline to the RI building, going through only a radiation controlled area
- Validation of the simulation for residual radionuclides
- Support by T. Shima and T. Suzuki

Irradiation tests of Graphite, SiC, and SiC/SiC was approved by PAC with "Grade A".

By K. Ninomita at Osaka univ.

#### Residual radionuclides on graphite sample



By K. Ninomita at Osaka univ.

## Residual radionuclides on SiC sample



#### Summary and Acknowledgement

Possibility of SiC as Low-Z target material is investigated.
 SiC coated graphite and SiC/SiC composite are candidates.
 Higher efficiency of beam transport can be mostly expected at NITE SiC/SiC.
 Higher oxidation resistance can be expected for both cases.
 Residual radiation dose will be higher at NITE SiC/SiC.
 Oxidation tests are on-going.

Irradiation tests at BLIP and RCNP are on-going.

Thanks to RaDIATE, Osaka Univ. /RCNP, Muroran Institute Technology /OASIS collaborations

#### 生成放射性核種(Fulka計算との確認用・7時間照射)

言っていた。

				生	成	放射性	<b>Ł核</b> 種	/Bq		
		照	射後	黒	鉛					
半減期	半減期	直	後	全	:体:	3.5	E11			
H3 :	12.3	3年			С	11:	3.2	2E11		
Be7:	53E	E			С	10:	1.5	5E10		
Na22	207 : 26:				B	e7:	5.1	E8		
Na24	: 158	, 寺間			Η	3:	1.1	E7		
		1 ป	1週		[体:	4.7	7E8			
				B	e7:	4.6	6E8			
				Η	3:	1.	1E7			
Ta	rget :	graph	ite, V	/=2.4	-0 ]	E2 cr	$m^3$	修正済:標的の	)表の値が約67	桁間違ってい <i>た</i>
							t = (	) s	t = 1	week
	А	Sym.	Z	T 1/2	2	В	q	Bq/cm <sup>3</sup>	Bq	Bq/cm <sup>3</sup>
	3	н	1	12.32	2 y	9.2E	+06	3.8E+04	9.2E+06	3.8E+04
	7	Be	4	53.12	d	6.0E	+08	2.5E+06	5.5E+08	2.3E+06
	11	С	6	20.33	m	5.0E	+11	2.1E+09		

## Residual radionuclides for COMET

	_			Half Life	gamma_energy (MeV)	B	3q	Sv∕h <b>*</b> mີ2
SiC/SiC	fresh						6.59E+11	1.23E+05
		C 11	1.02E+11	20m				
		Si 27	8.98E+10	4.2s				
		Al 26m	7.80E+10	6.3s				
		F 18	6.09E+10	1.8h				
		Be 7	5.95E+10	53d		0.478		
	1w						6.84E+10	2.11E+03
		Be 7	5.66E+10	53d		0.478		
		Н 3	7.11E+09	12.3y		0.02		
		Na 22	4.67E+09	2.6y	1.27, 0.543(e+)			
		Y 88	1.63E+09	107d		1.86		
		Sr 85	1.40E+09	65d	0.878, 0.514			
		Rb 83	1.08E+09	86d		0.525		
	1y						1.15E+10	1.38E+03
		Н 3	6.75E+09	12.3y		0.02		
		Na 22	3.52E+09	2.6y	1.27, 0.543(e+)			
		Be 7	5.16E+08	53d		0.478		
		Y 88	1.99E+08	107d		1.86		
		Rb 83	6.07E+07	86d		0.525		

#### Residual radionuclides

20 minutes

#### Residual radionuckides /Bq

Fresh	С	SiC		SiC+AlY	
H3	-	4.82E+11	6.34E+11	6.37E+11	Half life
Be7		3.71E+12	3.33E+12	3.17E+12	H3 : $12.3$ years
C11		1.02E+13	6.57E+12	6.27E+12	Be7: 53 days
Si27			7.39E+12	6.98E+12	C11: 20 minute
Al26m			3.33E+12	3.35E+12	Na22: 26 years
1 week	С	SiC		SiC+AlY	
Н3		4.82E+11	6.33E+11	6.37E+11	
Be7		3.39E+12	3.04E+12	2.89E+12	
Na22			3.85E+11	3.72E+11	
1 month	С	SiC		SiC+AlY	
H3		4.80E+11	6.31E+11	6.34E+11	
Be <mark>7</mark>		2 51E±12	2 25E±12	2 15E±12	

Na Residual radiation dose of SiC is much higher than graphite and is slowly decayed due to the effect of Na22. 1 Effects of additives for composite material is not so large. H3 Be These amounts will be validated at irradiation tests of RCNP. Na

## Back-up slides: TCC correlations (1)

- For Metal Interface: Mikic Correlation\*  $h_m(p) = 1.55 \frac{k \cdot m_r}{A_r} \left(\frac{\sqrt{2} \cdot p}{E \cdot m_r}\right)^{0.94}$ • For Graphite-Metals Interface:
- E. Marotta Correlation\*\*  $h_g(p) = 1.49 \frac{k \cdot m_r}{A_r} \left(\frac{2.3 \cdot p}{E_g \cdot m_r}\right)^{0.935}$

Input Parameters of the materials : Roughness (RA) Poisson Ratio Young Modulus Thermal Conductivity

"The assumptions included nominally flat contacting surfaces, uniform pressure distribution at the interface, elastic deformation and a vacuum environment..."

\*HANDBOOK OF HEAT TRANSFER (Warren M. Rohsenow, James R Hartnett, Young I. Cho), chapter 3, page 3.58.

\*\* E. Marotta and L. S. Fletcher. "Thermal contact conductance of selected polymeric materials", Journal of Thermophysics and Heat Transfer, Vol. 10, No. 2 (1996), pp. 334-342

Convective Coefficient of cooling water assumed to be: 6000 W/m2K



## Back-up slides: TCC Assumed- Low Density Capsule

	Reference Analysis (Real		
Contacts			
Name	Pressure assumed (MPa)	Ra assumed (um)	TCC [W/m2K]
SS_Graphite Sigraflex	0.	2 5	5000
Sigraflex_Si	0.	2 10	10000
Si_Makimura Graphite	0.	2 5	2000
SS_Makimura Graphite	0.	2 50	400
Filler_SS radial Gap	(No contact, just radiation)		20
Sigraflex Thermal Conductivity	5/150 W/mK		

#### Reference Analysis

	Conservative Analysis (Very low pr	essure Contacts)	Degradation of Sigraflex	Contact loss due	to degradation
Contacts		•	Contacts		
Name	Pressure assumed (MPa)	TCC [W/m2K]	Name	Pressure assumed (MPa	TCC [W/m2K]
SS Graphite Sigraflex	0.06	3400	SS_Graphite Sigraflex	0.2	30
Sigraflex Si	0.01	4500	Sigraflex_Si	0.2	30
Si Si samples	0.04	150	Si_Si Samples	0.4	230
Si Makimura Graphite	0.01	400	SS Makimura Graphite	0.06	400
SS Makimura Graphite	0.06	400	Filler_SS radial Gap	-	20
Filler SS radial Gap	-	20	Sigraflex Thermal Conductivit	5/150 W/mK	
Sigraflex Thermal Conductivity	5/150 W/mK		Results		
	5,155 W/ III			Max Temp [C]	
Results			SS Capsule	77	
	Max Temp [C]	Max Stress [MPa]	Sigraflex	1250	
SS Capsulo	61	190	Si_1 Samples	1150	
33 Capsule	04	180	Si_2 Samples	765	
Sigraflex	174	67	Makimura Graphite	692	
Si 1 Samples	200	50 in Samples / 35 in t			
<u></u>			Interfaces	Expansion [um]	
Si_2 Samples	478	50	Si samples - Si filler	-	
Makimura Graphite	420	4 in samples / 9 in fille	Si filler_SS capsule	-	
•			Makimura Graphite Samples/F	-	



#### "Silicon Carbide"

#### Major structural material for fusion applications

#### Crystal structure

More than 200 polytypes are currently reported. The fundamental structure unit in SiC is <u>a covalently bonded</u> primary co-ordination tetrahedron (SiC<sub>4</sub> or CSi<sub>4</sub>).

The most common polytypes are 3C, 4H, 6H, and 15R. The 3C-SiC are know as  $\beta$ -SiC, and the others are classified as  $\alpha$ -SiC.

The  $\beta$ -SiC is believed to be more stable, although some studies dispute this findings.

#### Fabrication of Silicon Carbide

<u>Various fabrication techniques</u>; CVD, Reaction bonding, Sintering, Polymer pyrolysis <u>> CVD</u>− highly crystalline, pure, high density. But Difficulty for fabrication of Large components, and expensive

<u>Reaction bonding</u> – Mixture of SiC and carbon particles with metallic silicon in the form of vapor or liquid. Severe degradation of bulk performance by Residual silicone.
 <u>Pressureless sintering process</u> – SiC particles with <u>boron</u> and carbon. <u>Flexibility in complex components shape</u>. But <u>degradation of mechanical properties</u>

➢<u>Hot-pressing liquid-phase sintering</u> – Robust and dense. But just simple shape and degradation of mechanical properties

➢ Polymer pyrolysis – Continuous SiC fibers and porus SiC

First candidate of SiC rotating target;

Pressureless sintering process, KYOCERA SC-1000

#### "Material Properties of Silicon Carbide"

- Density; 3.2 g/cc (1.82 g/cc)
- Heat generation by proton beam; 8kW (4kW), 2 times larger than Graphite
- Thermal Conductivity; 200W/m/K @ R.T.
  - (130W/m/K @R.T., 50W/m/K @1000°C)
  - In our case, radiation damage must be considered; 20W/m/K
- Upper temperature limit; 2000 $^{\circ}$ C (1700  $^{\circ}$ C) on the viewpoint of vacuum 1000  $^{\circ}\mathrm{C}$  on the v.p. of heavy radiation damage, dislocation loop
- Bending Strength; 450MPa (45MPa), 10 times larger than Graphite. (By radiation damage; 350MPa @4dpa 500 degC
  - 280MPa @25dpa 800 degC for typical sin. material [1])



- Young's Modulus; 440GPa (11GPa), 40 times larger than Graphite.
  - Emissivity; 0.8-0.9 (0.94; calibrated by T.C.)
- Thermal stress depends on Young's modulus x Heat
  - [1] G.W. Hollenberg et al. Journal of Nuclear Materials 219 (1995) 70-86
- "10 times Strength" vs "80 times Thermal stress"

For thermal stress, 8 times larger risk than Graphite.

## Oxidation resistance of silicon carbide

Active oxidation (low O<sub>2</sub> pressure) VS Passive oxidation (high O<sub>2</sub> pressure) J. Am. Ceram. Soc., 74 [10] 2583-86 (1991), J. Am. Ceram. Soc., 72 [8] 1386-90 (1989)



Temperature dependence of the transient oxygen partial pressure from active to passive oxidation



Passive oxidation 1500 degC, 1 atm, 5000 h Thickness gain: 15 mm



Active oxidation 870 degC, 1Pa, 5000 h, Thickness reduction: 1.8 mm 870degC, 10<sup>-3</sup> Pa, 5000 h, Thickness reduction: 1.8 mm

#### **Resistance to unexpected air introduction**

Oxidation tests of NITE SiC/SiC are on-going by OASIS.

Oxidation tests of SiC coated graphite are planned by Hitachi co., Itd.