Materials for Sliding Ring Seals of the German Spallation Neutron Source

R.-E. Schulze+, W. Delle+, H. Nickel+, H.A. Schulze+, W.F. Sommer++, K.H. Victor+++

+ KFA Jülich GmbH, D-5170 Jülich, Federal Republic of Germany

++ Los Alamos National Laboratory, H 383 MP-13, Los Alamos, NM 87545, USA

+++ Pacific Wietz GmbH u. Co. KG, D-4600 Dortmund 72, Federal Republic of Germany

Introduction

The use of sliding ring seals in pump systems of nuclear power stations as well as in the target support and drive unit of the planned German Spallation Neutron Source[1] (SNQ) require suitable ring material combinations. Several combinations have been proposed for the ceramic rings of the two SNQ sliding seals (Tab. 1). They correspond to the types of seal, which particularly differ from each other by the kinds of friction:

- type I with mixed (solid/liquid) friction for sealing of the cooling water from the internal vacuum space
- type II with technical dry (solid/solid) friction for retention of water vapour, which might develop at the first seal, from the external vacuum space[2].

The selection of promising candidates is based on industrial experience with conventional sliding ring seals as well as on results of seal tests, successfully performed in standard equipment at Pacific company. Futhermore, materials are included that are used in the pump systems of nuclear power stations.

Table 1. Material combinations for sliding ring seals in the SNQ as well as in nuclear power stations

Sliding ring seal	Material combinations for sliding rings: proposed and investigated for seals in the SNQ	used for seals in nuclear power stations	provided for seals in nuclear power stations
with mixed friction	Si-SiC/Carbon(C*)	Si-SiC/Carbon(Sb*)	Si-SiC-C/Si-SiC-C
	Si-SiC-C/Carbon(C)	WC/Carbon(Sb)	
	Si-SiC-C/Si-SiC-C		
	Si-SiC/Si-SiC		
with dry friction	Si-SiC-C/Carbon(C)		
	Si-SiC-C/Carbon(Sb)		

* impregnation: carbon and antimony resp.

Materials programme and investigations

The materials programme resulting from the selected combinations considers three categories of materials:

1. reaction-bonded silicon carbides
2. silicon carbide/graphite compound materials
3. carbon/graphite materials.

Altogether, 8 sliding ring materials are avialable for investigations (Tab. 2). Development work at

Table 2. Sliding ring materials being investigated and irradiated for the SNQ seals

Material category		Material: Designation	Ratio of the main phases*	Density ($g \cdot cm^{-3}$)
1	Reaction-bonded SiC	Si-SiC	SiC : Si = 8 : 1	3.07
2	SiC/graphite compound materials	Si-SiC-C	SiC : C_1 = 1 : 1	2.87
			SiC : C_1 = 2 : 1	2.65
			SiC : C_1 = 4 : 1	2.90
3	Carbon/graphite materials	Carbon(C)	C_2 : C_1 = 2 : 1	1.80
			C_2 : C_1 = 3 : 1	1.65
			C_2 = 100%	1.50
		Carbon(Sb)	C_2 : C_1 = 2 : 1	2.55

C_1 = carbon, predominantly ordered
C_2 = carbon, predominantly disordered

*ratio of the weight fractions (category 1 and 2)
ratio of the volume fractions (category 3)

several industrial companies as well as at KFA has contributed to this materials palette.

Identification of the phases in each material and determination of phase ratios have been performed by x-ray diffraction analysis and quantitative image analysis. The results are supported by investigation of the microstructure by use of ceramography and scanning electron microscopy.

From the first material category, Si-SiC is available which has been already proved in industrial practice and sliding rings in large SNQ dimensions (Ø ≈ 400 mm) have been successfully manufactured. Investigations show that the bearing part of the sliding ring material consisting of α/β-SiC amounts to about 85 vol.% with a ratio of α-phase to β-phase of about 7 : 1. β-SiC (cubic), due to the reaction of C and Si during silication, has been formed at the grain boundaries of α-SiC (hexagonal). The skeleton pores are filled with free Si.

From the 2nd material category three SiC/graphite compound materials are under investigation. They have been developed with regard to sliding rings and sliding bearings as well as for use as high temperature materials. They show high hardness and high resistance to wear as well as good chemical, thermal and thermal shock resistance. The main phases of the three materials are SiC and C_1 (predominantly ordered carbon, Tab. 2). Their ratios show a systematical variation. Additionally, different amounts of free Si are present. Till now, in

most cases two phases were found in SiC. In the Si-SiC-C materials with ratios of SiC : $C_1 \approx 2:1$ and $\approx 4:1$ (coat-mix based material[3]) SiC mainly consists of the cubic β-phase (95 %). Investigations of the first material have demonstrated that the structure is very homogeneous and consists of a matrix of SiC with embedded graphite grains. The free Si is evenly distributed. At present, sliding rings in SNQ dimensions consisting of SiC/graphite compound materials are not yet available. Development work at industrial companies is under way.
The main component of all the carbon/graphite materials belonging to the 3rd category, is predominantly disordered carbon (C_2). Especially with a view to dry friction three variants contain a portion of predominantly ordered carbon (C_1) for improving the self-lubricating properties. The coat-mix based material[4] with 100 % C_2 may be considered as a limiting case and is particularly suitable for obtaining a better understanding of the irradiation behaviour of the materials.
The carbon/graphite materials are divided according to their impregnation (carbon(C) and carbon(Sb)). C impregnation consists of binder coke (C_2) formed from resin during the coking process, this being the second C_2 component beside the filler coke. For impregnation of carbon(Sb), the metallic antimony is used. Both types of material distinctly differ from each other in open porosity and surface roughness, which are higher for carbon (C); accordingly, the latter exhibits more space for water storage. Because of problems with carbon(Sb) used for seals with mixed friction in nuclear power stations[5], the carbon(C) variants seem to be more suitable for this seal type in the SNQ. Sliding rings of SNQ dimensions consisting of carbon(C) and carbon(Sb) are already available.
In order to establish complete data sets for the 8 sliding ring materials, extensive measurements for the determination of physical and mechanical properties are being performed[2]. In addition, special investigations concerning the material structure are under way.

Irradiation testing and first results

During SNQ operation, the sliding seals will be exposed to spallation neutrons. The expected operation conditions are:
- time of operation: 12000 h
- max. operating temperature: for the seal with mixed friction 70°C, for that with dry friction 200°C
- max. accumulated spallation neutron fluences (in the case of ^{238}U as a target material): $\sim 1 \cdot 10^{19}$ and $\sim 1 \cdot 10^{20}$ cm^{-2} $E > 0,1$ MeV resp. for the two SNQ steps of development.

For the testing of sliding ring materials under conditions near those of SNQ operation, 2 experiments were performed in the spallation neutron environment at LAMPF. The test capsules were irradiated at different temperatures, thereby accumulating different fluences:
1st capsule: 50 - 100°C, $\sim 2 \cdot 10^{20} cm^{-2}$ $E > 0,1$ MeV
2nd capsule: 100 - 150°C; $\sim 6 \cdot 10^{19} cm^{-2}$ $E > 0,1$ MeV.
The capsules contained 20 and 19 samples machined from sliding rings of SNQ dimensions, representing the 3 different materials (categories 1 and 3):
- Si-SiC (SiC : Si $\approx 8:1$)
- carbon(C) ($C_2 : C_1 \approx 3:1$)
- carbon(Sb) ($C_2 : C_1 \approx 2:1$)

The first results for samples which allow a handling for measuring are summarized in Tab. 3.
Both materials (Si-SiC and carbon(C)) show very high dimensional stabilities and small changes in density. The thermal conductivity (λ) of Si-SiC

Table 3. First results of irradiation testing of sliding ring materials in the spallation neutron environment at LAMPF

Irradiation data	Material property/ Dimension	Irradiation-induced changes Si-SiC*	Carbon(C)**
spallation neutron	dimension	0 to + 0,04%	- 0,02 to + 0,03%
- flux: $\sim 2 \cdot 10^{13} cm^{-2} s^{-1}$ E > 0,1 MeV	density	within the limits of error	+ 3%
- fluence: $\sim 6 \cdot 10^{19} cm^{-2}$ E > 0,1 MeV	thermal conductivity	- 50%	not determined
irradiation temperature: 120 - 150 °C	Young's modulus	not determined	+ 43 to + 97%

*reaction-bonded SiC (SiC : Si = 8 : 1) **carbon/graphite material ($C_2 : C_1 = 3:1$)

decreases significantly (- 50 %), in agreement with results for graphite irradiated in high flux reactors at low temperatures[6]. The decrease can be correlated with the high scattering of phonons at irradiation-induced lattice defects at low irradiation temperatures[7]. Nevertheless, the reduced λ-value of Si-SiC is in the range of the λ-values of steels that are usually applied for structural parts surrounding the sliding ring seals. Measurements of the Young's modulus show high and anisotropic changes for carbon(C). This result is in agreement with results of fission reactor irradiations of other graphitic materials[8]. The high increase can be explained by the irradiation-induced pinning of mobile dislocations in the well-ordered regions and the reduced recombination probability of point defects at the low irradiation temperature[9]. As expected for the range of low temperatures, x-ray diffraction analyses showed no evidence for any irradiation-induced graphitization[10] of the predominantly disordered C_2 phase.
The results for carbon(C) are in good agreement with the results of carbon materials irradiated in the Dounreay Fast Reactor at 250°C up to a fluence of $1.8 \cdot 10^{20} cm^{-2}$ $E > 0.1$ MeV[11].
For irradiation testing of all the sliding materials (summarized in Tab. 2) another experiment at LAMPF is being prepared.

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