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Zakład UZ3 – kierownik prof. M.Dąbrowski

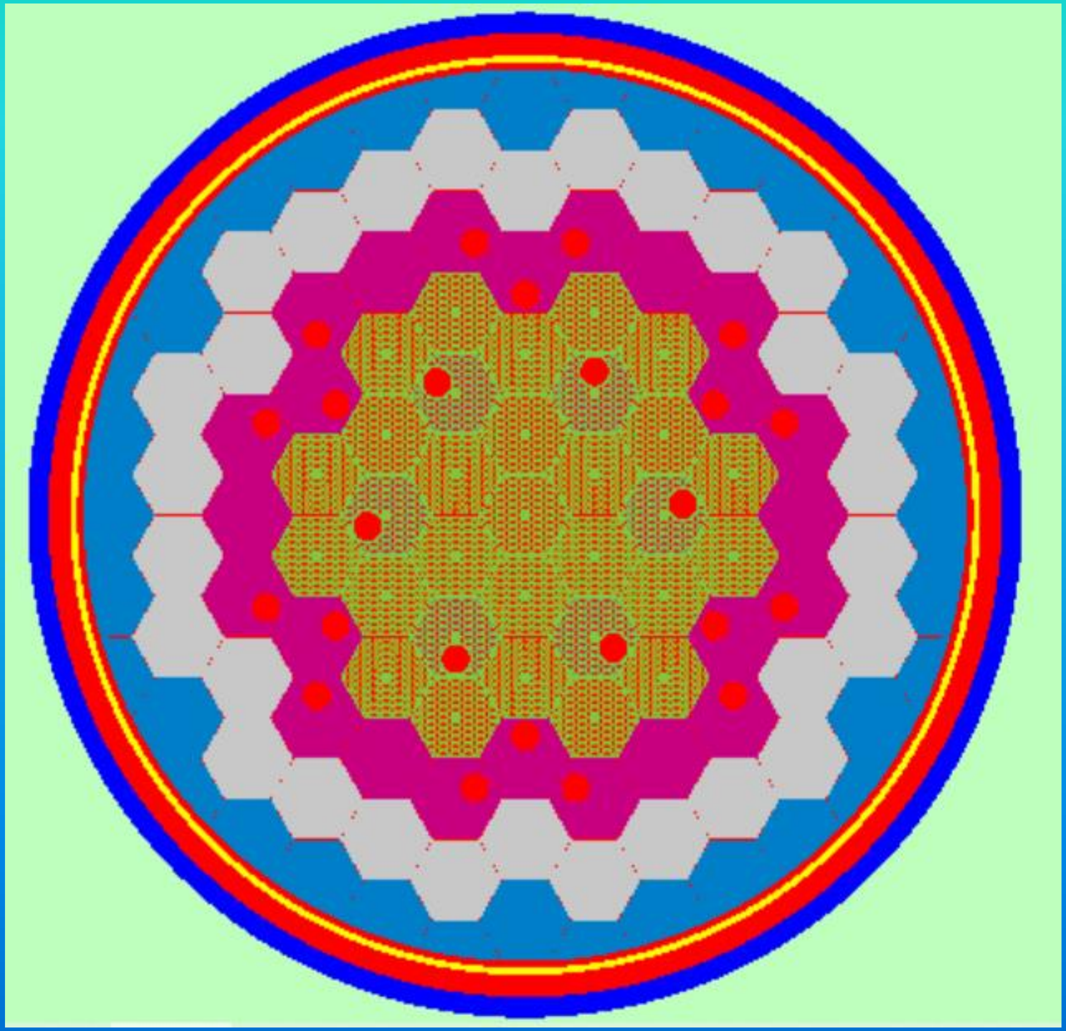
Ograniczenia bezpieczeństwa reaktorów z moderatorem grafitowym

December 2021

Outline

- Geometry and materials of GoHTGR and TERESA reactors
- Oxidation rate of nuclear graphites
- Informations about RBMK reactors
- Deformation and cracking of graphite blocks
- How increase safety of graphite reactors? - propositions

TERESA core+reflector- based on the GEMINI reactor

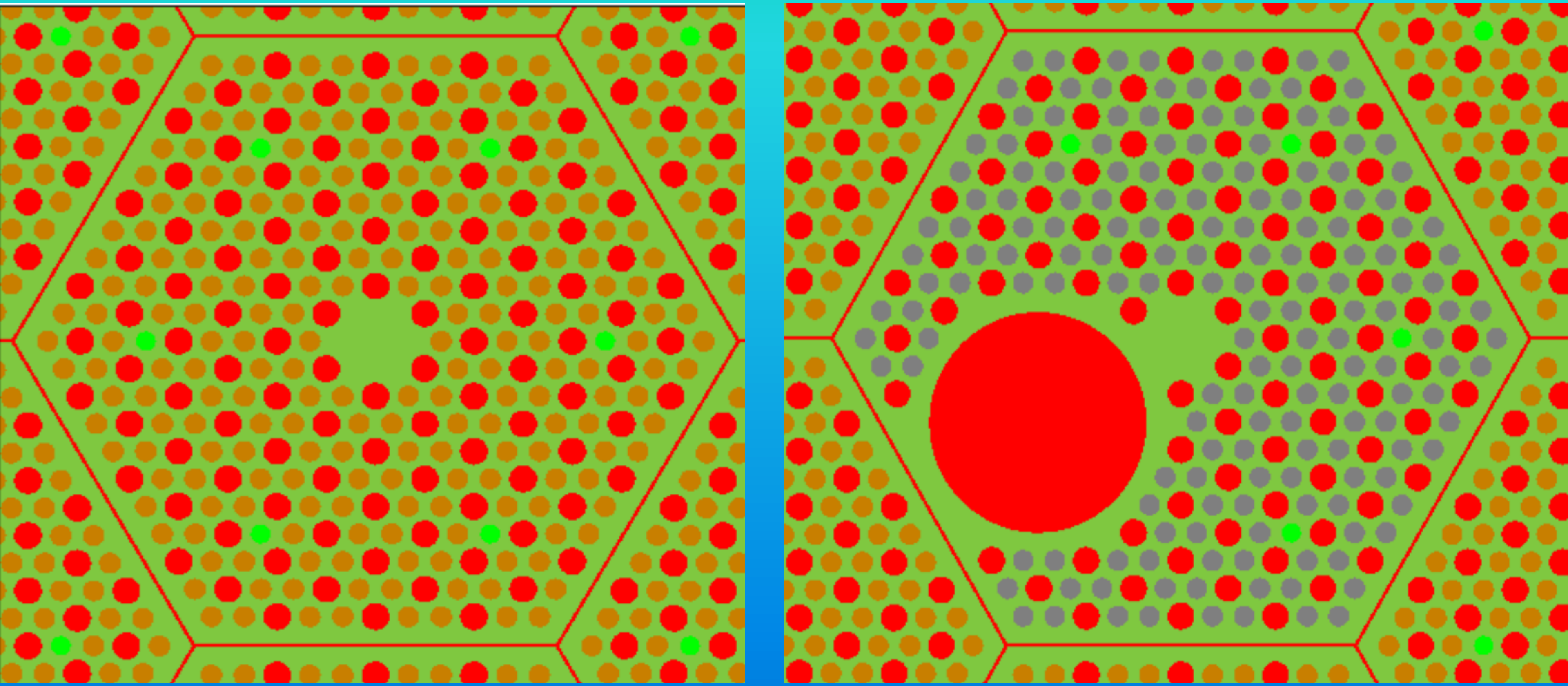


Cross section

Colors: red-helium, bronze and pink and white and light blue -graphite NBG-17, blue-steel S508, yelow-steel 800H.

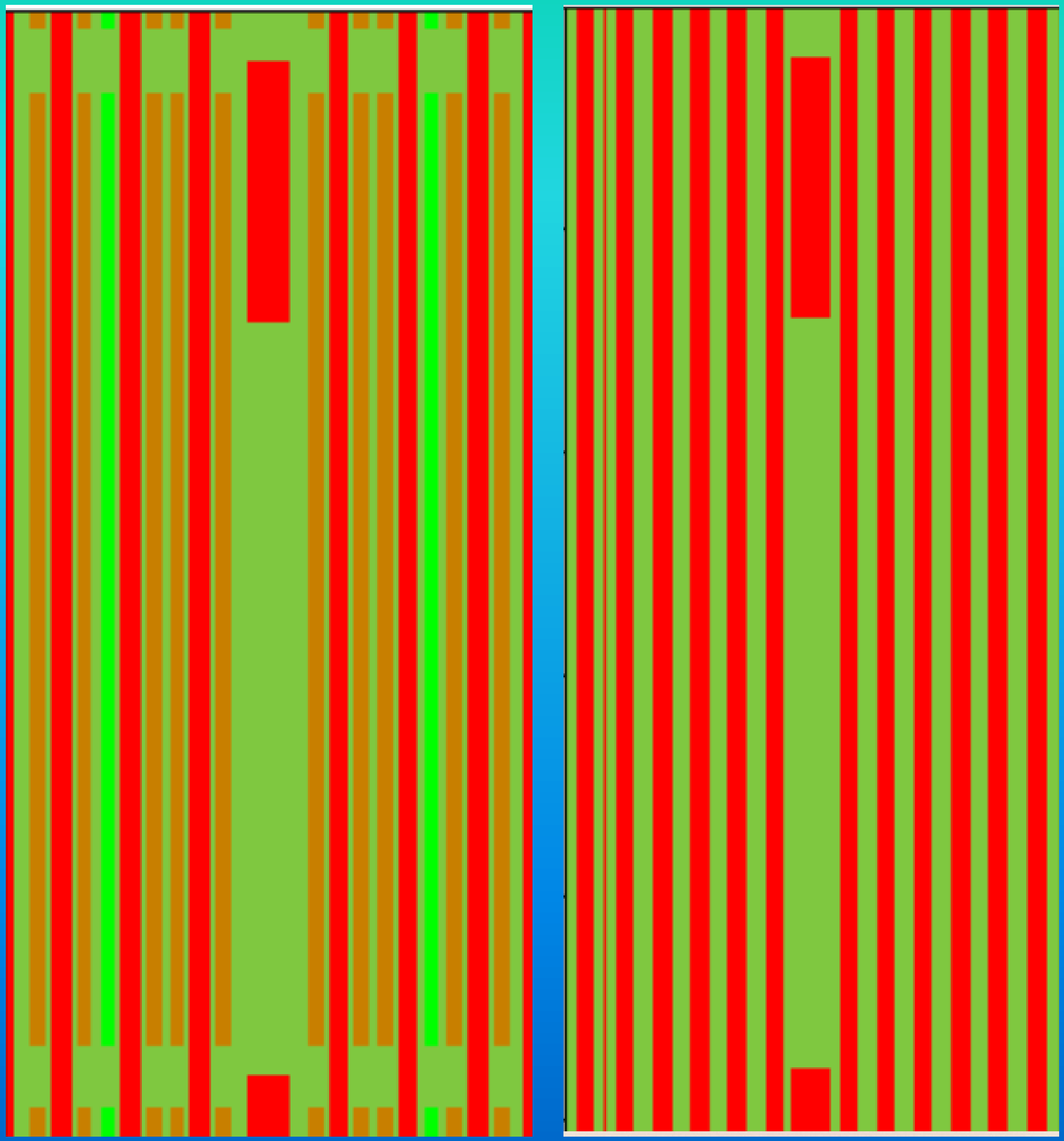
Keff=1.04 from MVP code
For temperature 300K

Fuel blocks – cross section



Fuel blocks: without CR (left), with CR (right).
Colors: red - helium, green - graphite NBG-17, bronze and blue – fuel rods, yelow - BP rods

Fuel block - longitudinal section



Longitudinal section XZ (left) and YZ (right) of fuel block without CR)

Colors: red-helium,
green- graphite NBG-17,
bronze – fuel,
Yelow - BP rods

Fuel rods –
graphite NBG-17 +TRISO
Packing 15%
TRISO diameter 460μm.

Project is based on the GENINI reactor

Reflector blocks –NGB-17
Fuel blocks –NGB -17
Fuel rods – NGB-17
TRISO particle – different kind of pyrolytic graphite

Safety technology

- Purpose: Inhibition of graphite component oxidation during accident
- Method : Development of nuclear grade fine grained isotropic graphite
- Results: Dense and few internal pores. Impurity which catalyzes the oxidation (burning) is extremely low.
- Oxidation-retardant (flame-retardant) even if the high temperature condition happens during air ingress accident.

High Temperature Gas-coolled Reactor ,
Safe and Eco-friendly Nuclear Reactor with
Meltdown-proof design
Japan Atomic Energy Agency (JAEA

Oxidation rate

Journal of Nuclear Materials 500 (2018) 64–71

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journal homepage: www.elsevier.com/locate/jnucmat

Comparison of NBG-18, NBG-17, IG-110 and IG-11 oxidation kinetics in air ☆

Jo Jo Lee [a, *], Tushar K. Ghosh [b], Sudarshan K. Loyalka [b]

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A R T I C L E I N F O

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A B S T R A C T

The oxidation rates of several nuclear-grade graphites, NBG-18, NBG-17, IG-110 and IG-11, were measured in air using thermogravimetry. Kinetic parameters and oxidation behavior for each grade were compared by coke type, filler grain size and microstructure. The thickness of the oxidized layer for each grade was determined by layer peeling and direct density measurements. The results for NBG-17 and IG-11 were compared with those available in the literature and our recently reported results for NBG-18 and IG-110 oxidation in air. The finer-grained graphites IG-110 and IG-11 were more oxidized than medium-grained NBG-18 and NBG-17 because of deeper oxidant penetration, higher porosity and higher probability of available active sites. Variation in experimental conditions also had a marked effect on the reported kinetic parameters by several studies. Kinetic parameters such as activation energy and transition temperature were sensitive to air flow rates as well as sample size and geometry.

Oxidation rate of nuclear graphite

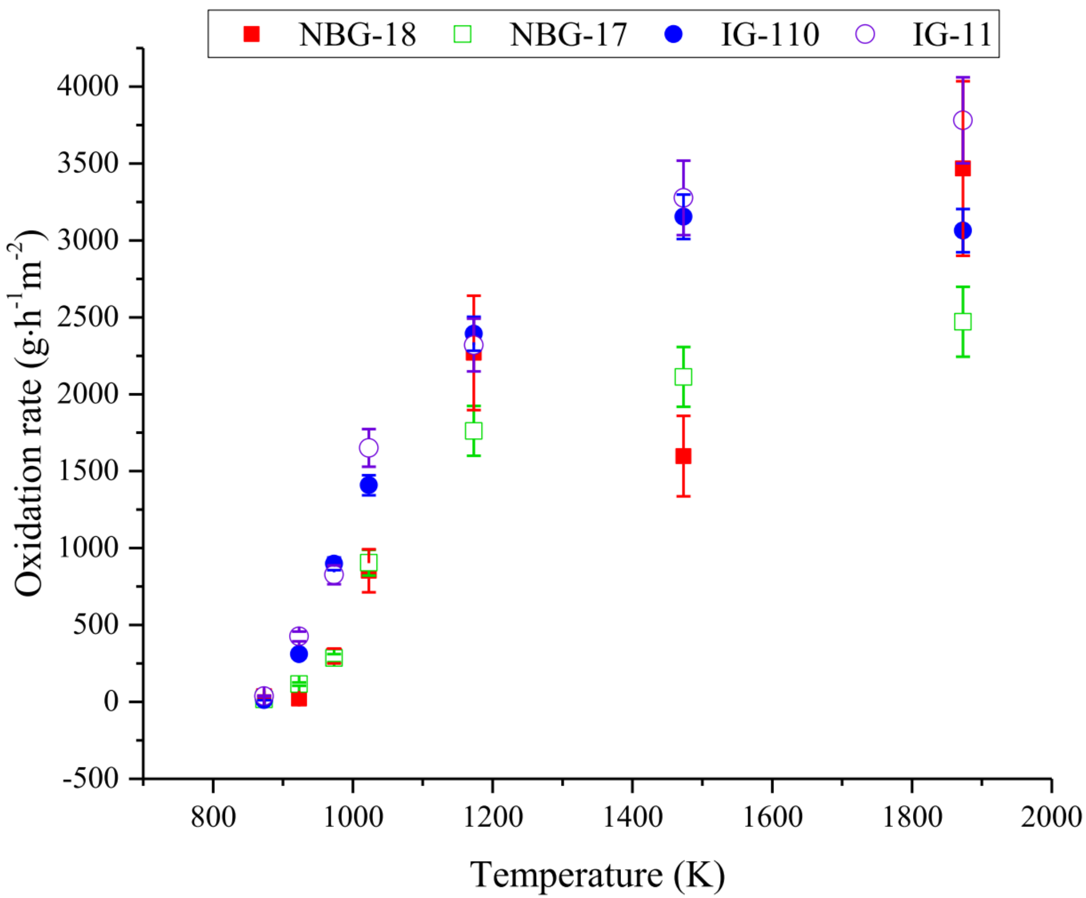

Fig. 1. Comparison of the oxidation rates of NBG-18, NBG-17, IG-110 and IG-11 in medical air, average of two trials.

Not irradiated graphite

Unit [gh-1m-2]	kW/m-2
3000	27.3
2000	18.2
1000	9.1

Jo Jo Lee , Tushar K. Ghosh , Sudarshan K. Loyalka ,
Comparison of NBG-18, NBG-17, IG-110 and IG-11 oxidation kinetics in air,
Journal of Nuclear Materials 500 (2018) 64-71,
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Graphite oxidatation

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Nuclear Engineering and Design

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Effect of temperature on graphite oxidation behavior

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Abstract

The temperature dependence of oxidation behavior for the graphite IG-11, used in the HTR-10, was investigated by thermogravimetric analysis in the temperature range of 400–1200 °C. The oxidant was dry air (water content <2 ppm) with a flow rate of 20 ml/min. The oxidation time was 4 h. The oxidation results exhibited three regimes: in the 400–600 °C range, the activation energy was 158.56 kJ/mol and oxidation was controlled by chemical reaction; in the 600–800 °C range, the activation energy was 72.01 kJ/mol and oxidation kinetics were controlled by in-pore diffusion; when the temperature was over 800 °C, the activation energy was very low and oxidation was controlled by the boundary layer. Due to CO production, the oxidation rate increased at high temperatures. The effect of burn-off on activation energy was also investigated. In the 600–800 °C range, the activation energy decreased with burn-off. Results of low temperature tests were very dispersible because the oxidation behavior at low temperatures is sensitive to inhomogeneous distribution of any impurity, and some impurities can catalyse graphite oxidation.

Oxidation of graphite IG-11

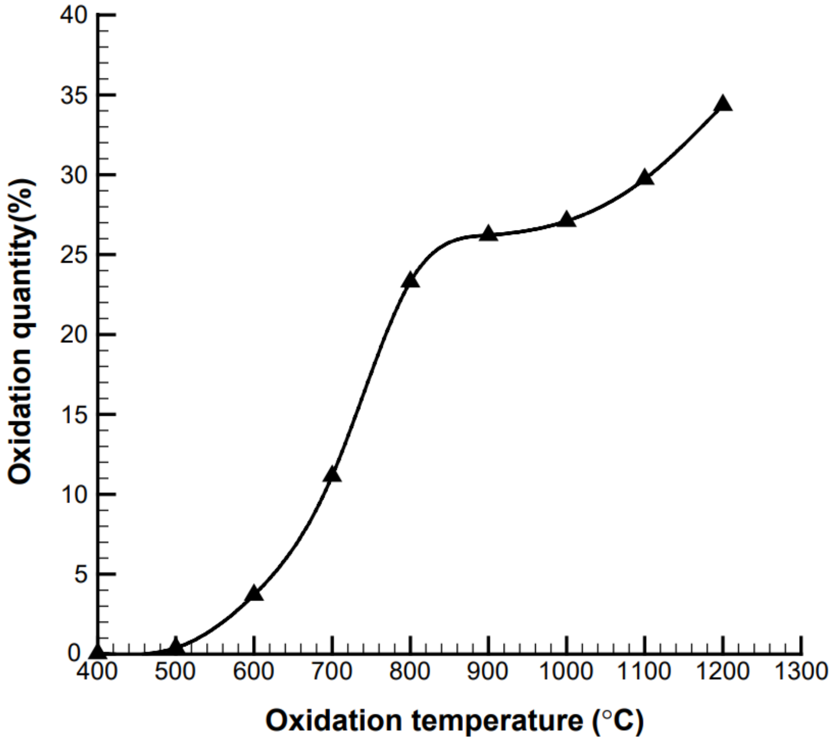

Fig. 1. Graphite oxidation extents at different oxidation temperatures.

The oxidation quantities were calculated from the weight decrease of graphite specimens and were normalized by comparison with the starting oxidation weight of the specimens

	kW/kg
10%	0.227778
20%	0.455556
30%	0.683333

The air was then piped at a flow rate of 20 ml/min and kept for 4 h

The specimens were cylinders of 10 mm diameter and 10 mm height, mass=1.3816g

Luo Xiaowei a, Robin Jean-Charles b, Yu Suyuan a
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Nuclear Engineering and Design 227 (2004) 273–280
a Institute of Nuclear Energy Technology, Tsinghua University, Beijing 100084, China
b DER/STR/LCEP, C.E.A. Cadarache, France

Oxidation of graphite IG-11

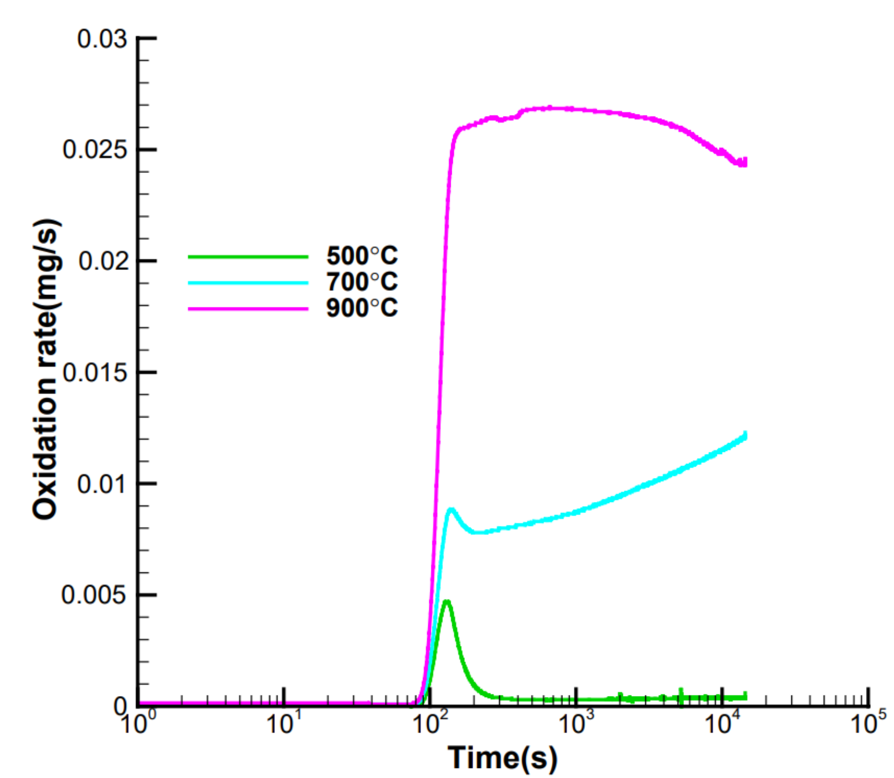

Fig. 3. Variation of oxidation rate with oxidation time at different temperatures.

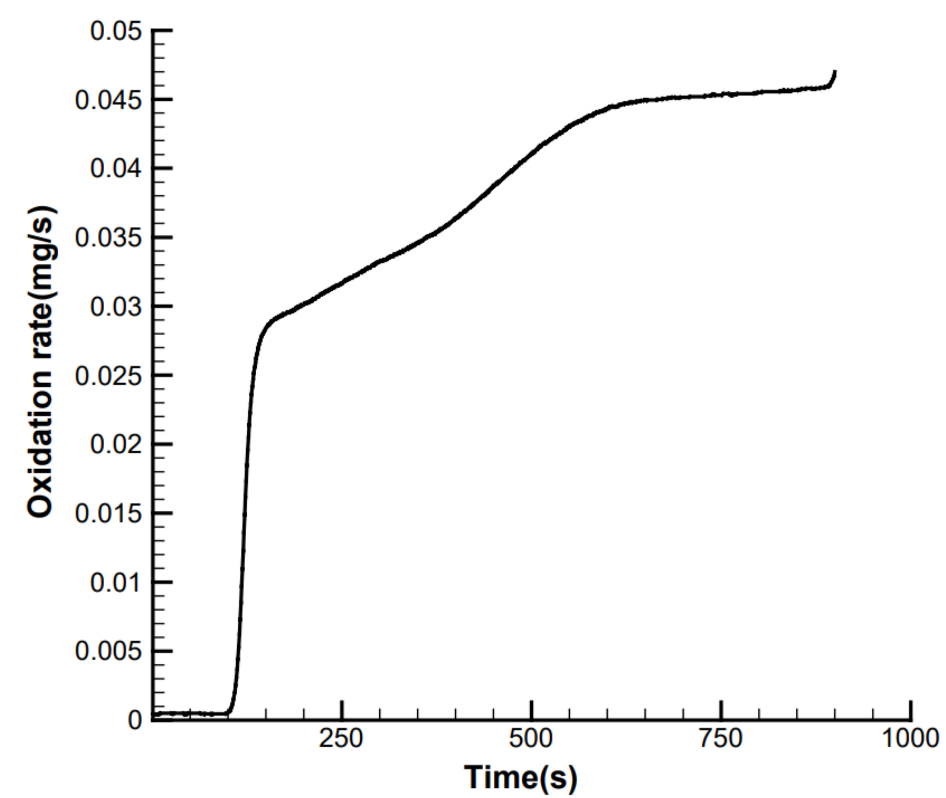

Fig. 6. Graphite oxidation at 1500 °C.

Luo Xiaowei a, Robin Jean-Charles b, Yu Suyuan a
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a Institute of Nuclear Energy Technology, Tsinghua University, Beijing 100084, China
b DER/STR/LCEP, C.E.A. Cadarache, France

mg/s	kW/kg
0.01	0.237406
0.02	0.474812
0.03	0.712218

Graphite oxidation

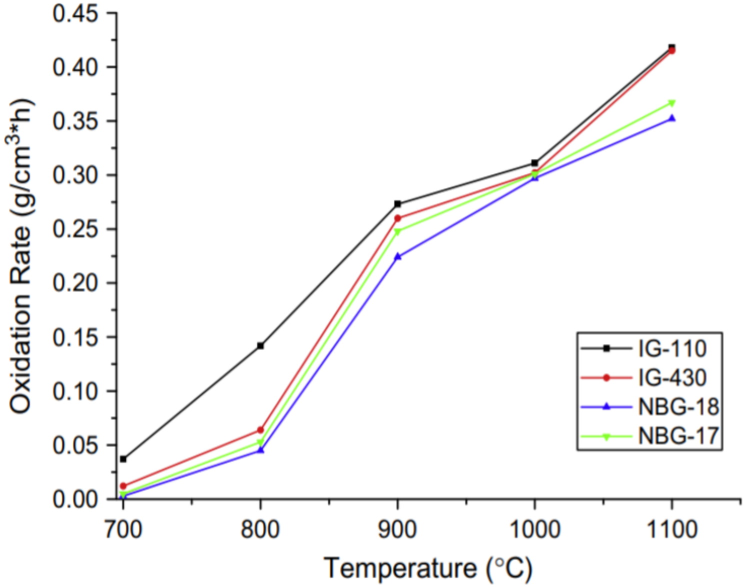


Fig. 6. The results of the measured oxidation rate for IG-110, IG-430, NBG-17 and NBG-18 graphite samples at various temperatures, showing pitch-based graphite (IG-430, NBG-17 and NBG-18) perform significantly better than that of petroleum-based graphite (IG-110) at 700–800 °C. All the oxidation rates for IG and NBG grade graphite converge at 1000 °C. However, open porosity at surface of materials results in the difference in oxidation rate at 1100 °C.

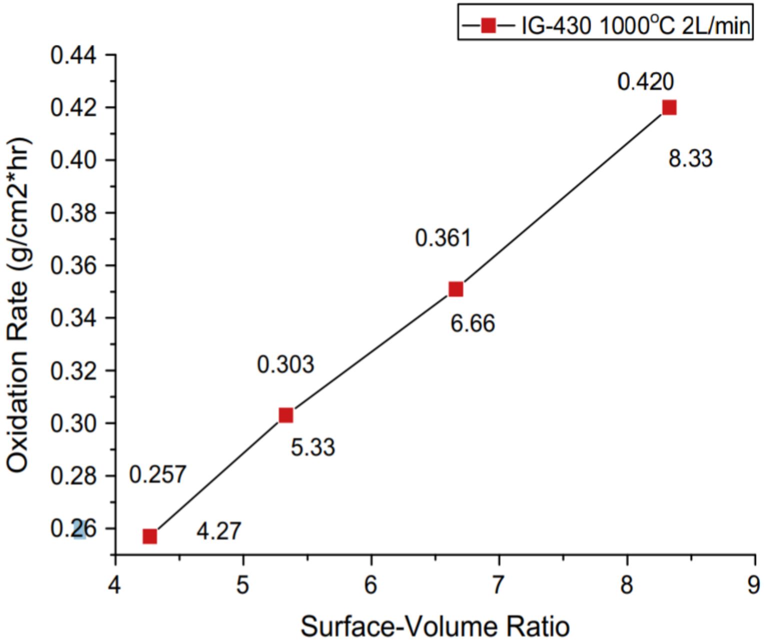


Fig. 11. The figure shows that oxidation rate is proportional to surface to volume ratio at 1000 °C, 2 L/min dry air. The result indicates that each unit graphite volume which contains more surface area for oxidation at boundary layer controlled regime results in higher oxidation rate.

Wei-Hao Huang , Shuo-Cheng Tsai , Chia-Wei Yang a, Ji-Jung Kai

The relationship between microstructure and oxidation effects of selected IG- and NBG-grade nuclear graphites,

Journal of Nuclear Materials 454 (2014) 149–158

Outer layer of TRISO –pyrolytic

Surface/Volume=250 [cm-1]

Oxidatation rate =15[g*cm-2h-1

Oxidation time =1.8[s] for IG-430

Graphite oxidation

- The results showed that the sleeve became 21.1% thinner in 5 h and that the fuel pellets lost 22% of their weight in 1 h.
- These experimental results show the concern of air oxidation on TRISO fuel particles over several hours, under the stated experimental conditions.

Journal of Nuclear Materials 454 (2014) 149–158

Extraordinary events

Unpredictable events that could destroy the cooling system and lead air to the reactor core

- Deliberate human action
  - war
  - terroristic attack (WTC)
- Failures of machines and devices
  - planes
  - rockets
  - artificial satellites and orbital stations
- Nature forces
  - meteorites and asteroids Chelabinsk
  - earthquakes Fukusima

More than 2,000 HTGR reactors are planned to be built.

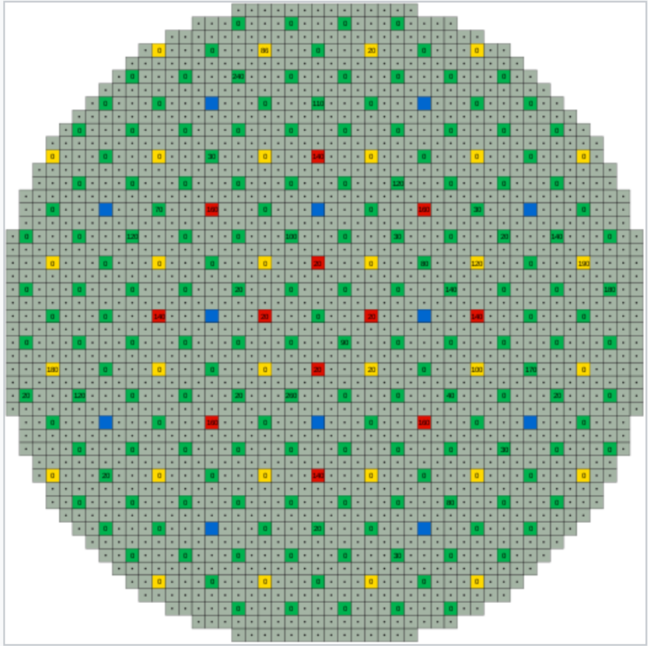
We cannot build a reactor that cannot be destroyed, but we can build a reactor that will not ignite even if it is seriously damaged.

RBMK reactors

Реактор большой мощности канальный (РБМК)

Number of RBMK reactors

- 9- works
- 7- shut down
- 1 – destroyed (Charnobyl)
- 9 –construction cancelled



Schematic plan view of core layout, Chernobyl RBMK reactor No. 4. (Quantity of each rod type in parenthesis):

- startup neutron sources (12)
- control rods (167)
- short control rods from below reactor (32)
- automatic control rods (12)
- pressure tubes with fuel rods (1661)

The numbers in the image indicate the position of the respective control rods (insertion depth in centimetres) at 01:22:30am [19] 78 seconds before the reactor exploded.

MKER (ros. *МКЭР, Многопетлевые Канальные Энергетические Реакторы*

MKER-800, MKER-1000, MKER-1500 – IV generation

-negative void reactivity coefficient

-natural coolant circulation

-economical, fuel natural uranium or enrichment 2%

The MKER-800 was under development in a joint project of Westinghouse and NIKIET

Petersburg,
Kursk – prototype MKER-800

Graphite problems

Восстановление графитовой кладки на ЛАЭС
Дата: 22/12/2014
Тема: Атомная энергетика

Успех-2014. В январе ЛАЭС вывела 1-й блок с восстановленной графитовой кладкой на полную мощность

В.И.Перегуда, директор Ленинградской АЭС

Из-за простоя первого энергоблока Ленинградской АЭС Концерн «Росэнергоатом» в 2012 г. недополучил порядка8-10 млрд руб. выручки, а также недосчитался 30- 40 млрд руб. заемных средств на инвестиции (директор по стратегии «Росэнергоатома» Павел Ипатов). Всего в России построено 11 реакторов типа РБМК, на них приходится примерно 40% выработки всего концерна. Причиной этого кризиса стало преждевременное старение графита, выявленное на первом энергоблоке Ленинградской АЭС в начале 2012 г. Не удивительно, что поиск технического решения этой проблемы стал для ГК «Росатом» задачей № 1.

Проблемы с графитом

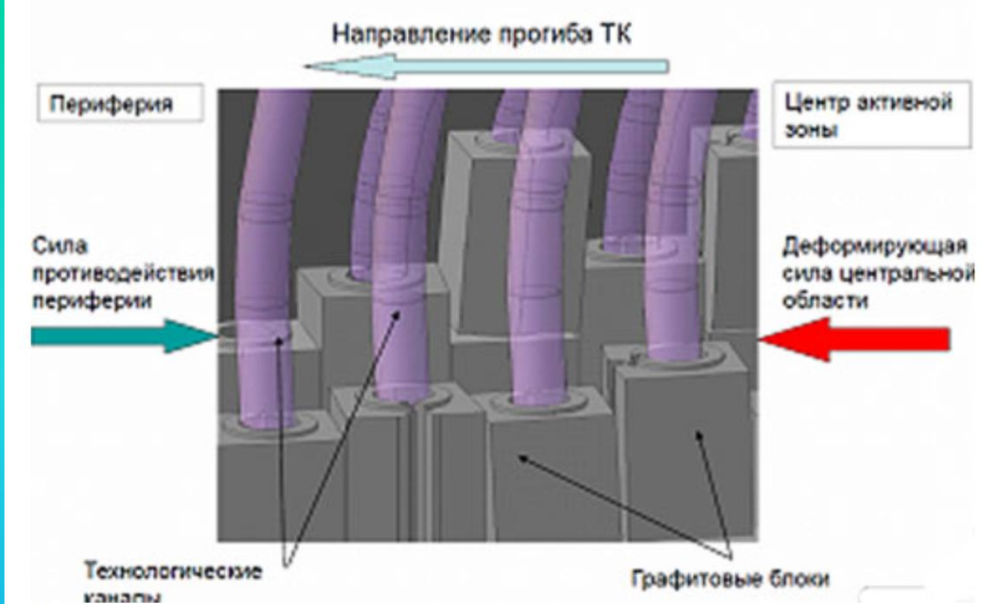


Рис.1б Механизм формоизменения графитовых колонн

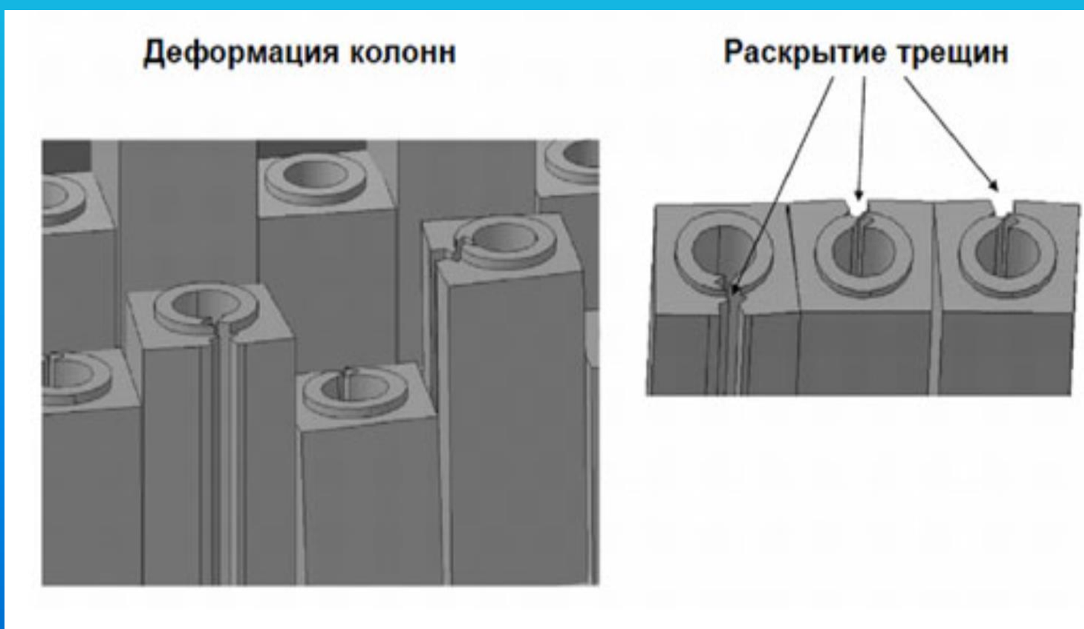


Рис.1а Механизм деформации колонн и раскрытия трещин

Графитовая кладка,исходно имевшая форму цилиндра (диаметром 12 м и высотой 9 м), состоящего из отдельных кирпичиков размером 25 × 25 см в горизонтальной плоскости и примерно 60 см– вертикальной, со временем приобретает бочкообразную форму с большим диаметром по центру, а сверху и снизу (где суммарное энерговыделение невелико) изменений почти нет.

Из-за деформации ГК могло произойти заклинивание тепловыделяющих элементов (ТВЭЛов) и стержней аварийной защиты.

[26] Восстановление графитовой кладки на ЛАЭС

Распухание графита

В 2011 году очередное обследование состояния реактора первого энергоблока ЛАЭС выявило преждевременное искривление графитовой кладки, вызванное радиационным распуханием графита и его последующим растрескиванием.[26] В 2012 году, на 37-м году эксплуатации, реактор был остановлен в связи с достижением предельных величин смещения кладки. В течение 1,5 лет были найдены технологические решения, позволившие уменьшить деформацию кладки путём пропилов в графите, компенсирующих распухание и формоизменение.[27

- В 2013 году реактор вновь был запущен, однако увеличивающиеся темпы накопления дефектов потребовали проведения практически ежегодных работ по коррекции кладки.

[26] Восстановление графитовой кладки на ЛАЭС

В.И.Перегуда, директор Ленинградской АЭС, http://www.proatom.ru/modules.php?name=News&file=print&sid=5746

Распухание графита

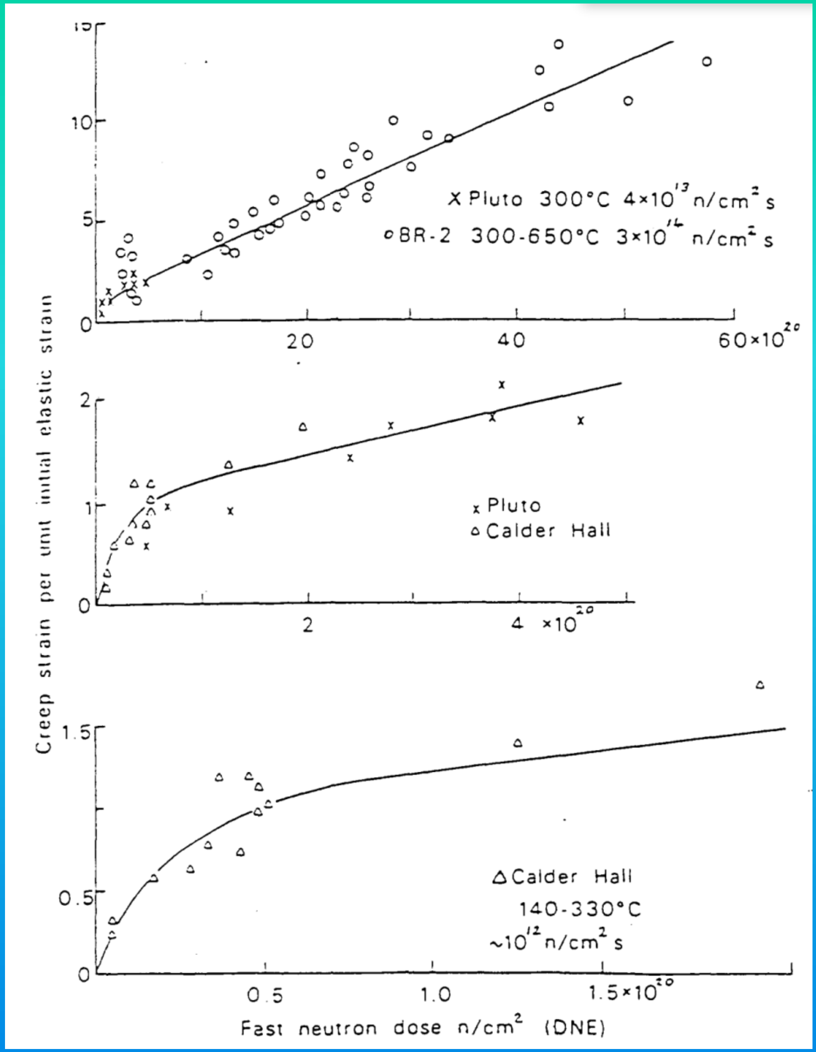
- Из-за простоя первого энергоблока Ленинградской АЭС Концерн «Росэнергоатом» в 2012 г. недополучил порядка 8-10 млрд руб.(300 mln USD) выручки, а также недосчитался 30- 40 млрд руб. (1mld USD) заемных средств на инвестиции

Цель работ по ВРХ - достижение значений стрел прогиба топливных каналов (ТК) и рабочих каналов системы управления и защиты (РК СУЗ) менее 50 мм,а также снижение скорости нарастания стрелы прогиба при эксплуатации до 15 мм/год, была достигнута.

Срок службы **первого энергоблока ЛАЭС** был продлен до 2018 г

Irradiation creep

sometimes called **cold flow**



creep (sometimes called **cold flow**) is the tendency of a solid material to move slowly or deform permanently under the influence of persistent mechanical stresses

COMPARISON OF CONSTANT STRESS IRRADIATION CREEP DATA ON GRAPHITE IN DIFFERENT FACILITIES

Young's modulus[GPa]: graphite 4.1-27, Berylium-288, SiC-90-137,

Poisson's ratio in creep was found to be 0.3

$$e_c = 0.23\ \sigma\gamma/E_o + \sigma/E_o(1-\exp(-4\gamma))$$

where σ (MPa)is the stress, *y* is the dose [n/cm2] (fluence) and *Eo* is the static Young's modulus prior to irradiation

The first term is the secondary creep strain and the second the transient creep strain.

IRRADIATION DAMAGE IN GRAPHITE

The works of Professor B.T.Kelly

BJ. MARSDEN
Graphite Section, AEA Technology, Risley, Warrington, Cheshire, United Kingdo

Creep strain

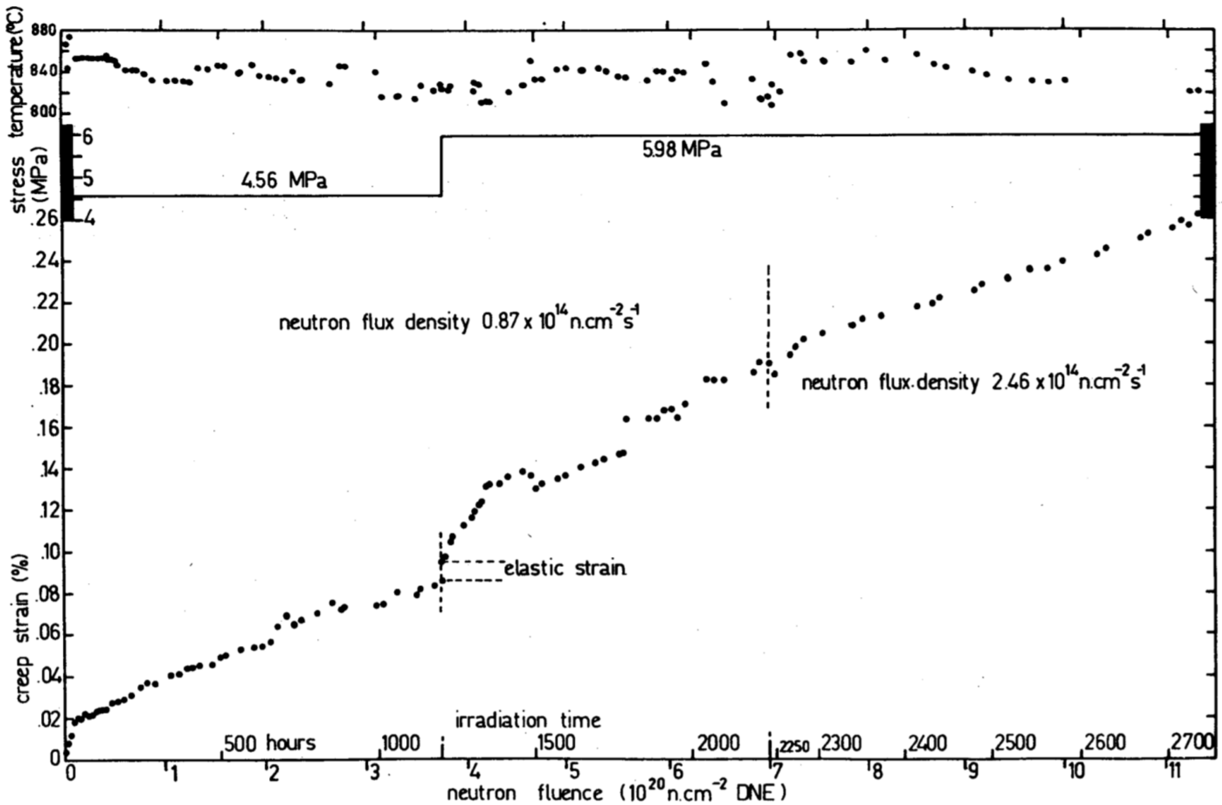


Fig. 11. Creep strain record of experiment R-135 (from Ref. 21; fluence values revised)

Creep depends on
- stress
- material properties
-- Young's modulus
-- Poisson's ratio
- temperature
- neutron flux density
- neutron fluence

R.JPRICE, Irradiation-induced creep in graphite: a review.
Prepared for the San Francisco Operations Office Departament Energy

How to choose a new moderator?

The following material properties should be considered:

- Low value of oxidation rate
- Not toxical
- Low atomic mass number
- Low value of cross section for neutron absorption
- Melting point (solid state, liquid)
- Young's modulus
  - high value - solid state
  - low value – gravel, sand
- Poison's ratio
  - high value - gravel, sand
  - low value – solid state
- Price

Solid state moderators- discussion propositions

Name	Melting point [C]	Young's modulus
• SiC	2800	90-137
• SiO_2	1700	
• Si_3N_4	1900	
• BeO	2500	345
• Be	1287	288
• Be_2C	2100	
• (Be_3N_2)		

dual fluid (prof.Czerski)

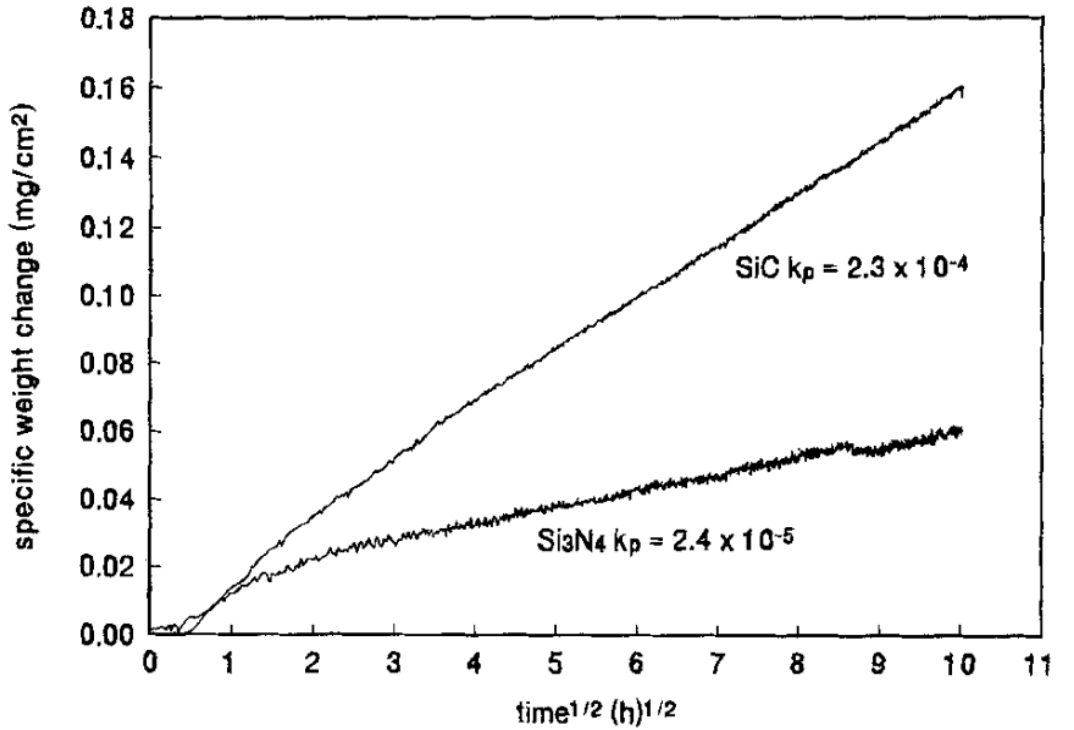


Fig. 4. Oxidation kinetics of CVD SiC and CVD Si_3N_4 in 1 atm oxygen at 1300°C as determined by thermogravimetry.

0.02mgh-1cm-2=0.2gh-1m-2

A Comparison of the Oxidation Kinetics of SiC and Si3N4,
Linus U. J. T. Ogbuji a, Elizabeth J. Opila b,
J. Electrochem. Soc., Vol. 142, No. 3, March 1995 9 The Electrochemical Society, Inc
a NYMA, NASA Lewis Research Center, Cleveland, Ohio 44135
b Department of Chemical Engineering, Cleveland State University, Cleveland, Ohio 44115

Conclusion

- Graphite has the following properties

- Relatively high rate of graphite oxidation in high temperature (burn)
- Deformations- swelling, creeping and cracking of graphite blocks

Graphite limits :

- Safety of reactor
- Economic rationale connected with long-term renovation and maintenance

Nuclear graphite research has been carried out for 80 years

Graphite HTGR may not be in competition with graphite-free HTGR

A graphite-free HTGR project should be made in NCBJ (e.g. Dual Fluid or Gas Fluid)

Thank you for attention

Main problems of RBMK reactor

- Positive reactivity coefficient (void reactivity)
- Relatively high rate of graphite oxidation in high temperature (burn)
- Deformations, Swelling and craking of graphite
- Limited economic rationale connected with long-term renovation and maintenance