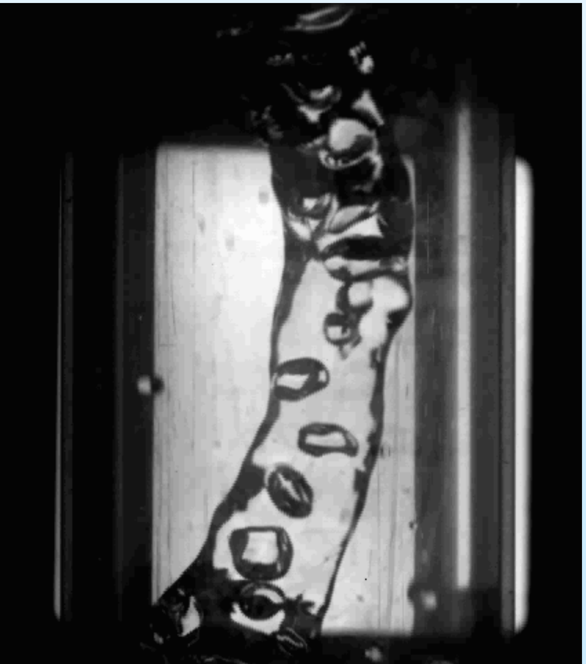
日時　　平成22年2月12日（金） 13:30～17:00
場所　　独立行政法人　原子力安全基盤機構（JNES） G会議室

平成21年度日本原子力学会 熱流動部会
「熱水力安全評価基盤技術高度化検討」
サブワーキンググループ　第2回会合
基礎研究開発に係る課題
『放射線誘起表面活性効果（RISA）』



賞雅 寛而
東京海洋大学

TUMSAT
Power and Energy Lab.

Contents

What is Radiation Induced Surface Activation?

1. Background and motivation
   - Road map
   - UV-light catalysis
2. Present studies of Radiation Induced Surface Activation
   -- Thermo-hydraulic incidents caused by RISA --
   1) Surface wettability in room temperature
   2) Leidenfrost temperature
   3) CHF in pool
   4) Reactor experiments for CHF/RISA
3. Mechanism behind RISA

熱水力開発･安全関連技術　導入シナリオのまとめ

産　官　学　学協会

2008 | 09 | 10 | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 | 25 | 30

海外展開
米国の標準設計認証（DC）等への対応、など
- US-APWR、AP1000、ABWR、ESBWR 等

FS → 概念設計 検討 → 概念設計 → 基本設計 → 詳細設計 → 許認可 → 建設 → 運開

新型炉　開発･設計

次世代軽水炉（国プロ）
- P　先進安全系（SG積極減圧）
- B　先進安全系（シビアアクシデント対策など）
- B　省ウラン（スペクトルシフト燃料）
- B　大型燃料（170-180万kW）

新型炉
- P　静的シビアアクシデント対策(IVR等)

炉規制国際動向　MDEP、OECD/WGNR等
- P+B 規制高度化　パッシブ安全系、世界標準（シビアアクシデント等）対応など　（協官）

自然循環炉
- P　高温高圧二相自然循環炉　（産官学）
- B　気水分離特性　チムニー内流動　（産官）

新型炉　現行炉　の　共通課題　安全性と信頼性の確保･向上

安全解析手法の高度化	P+B　BEコード＋統計手法、CFDの導入　（官産学）
事故後長期冷却	P+B　ストレーナ閉塞　（官産学）
シビアアクシデント対策	ヨウ素挙動、水素挙動、FCI、MCCI → 解析評価手法　（官産学） 現実的ソースターム評価など　（協官）
非凝縮性ガス対応	B　ガス燃焼（産官学） → 事例規格化（協官）
火災	P+B 火災影響評価（火災PSA評価法確立）　（官産）
炉心健全性	B Post-BT, RISA　（官産学）
高経年化	P　加圧熱衝撃時の高精度熱水力解析　（官産学）

現行炉　炉利用の高度化

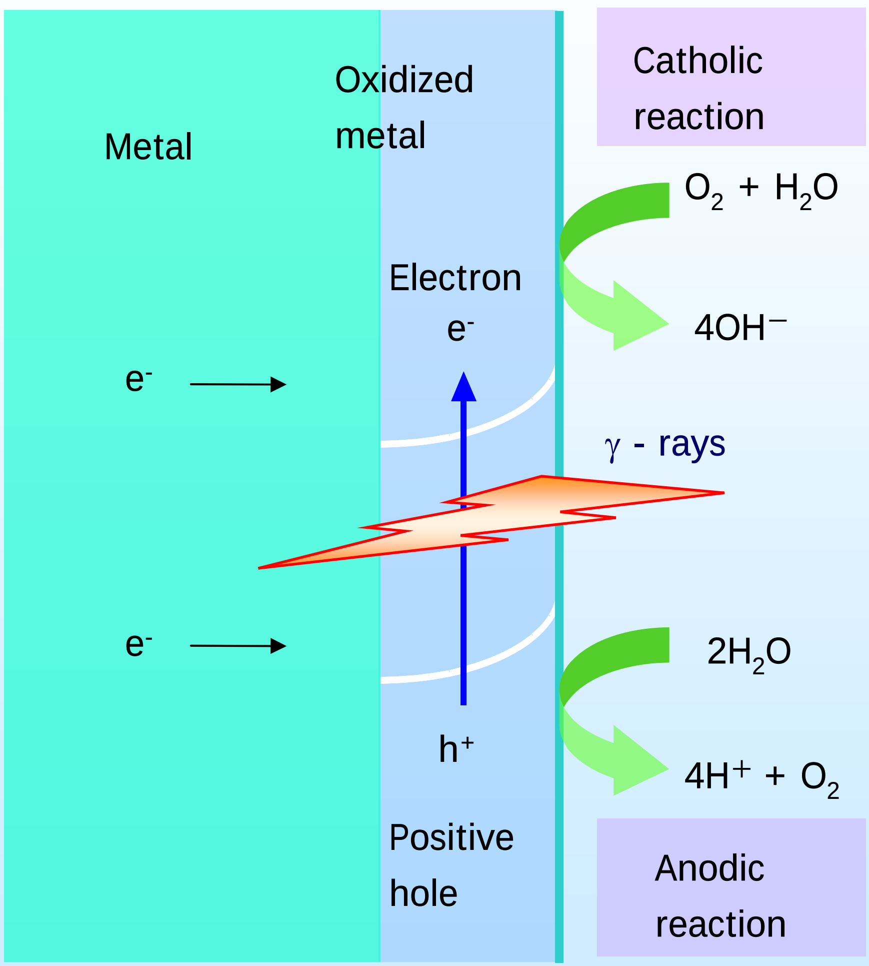
出力向上	出力向上の導入（H16版RM参照）　B　流力振動、炉心安定性、Post-BT　P+B　流量計精度保証など、RISA　（官産学）
燃料高度化	P+B 高燃焼度　B RIA過渡ボイド、Post-BT、柔軟運転サイクル　（官産学）

基盤の確保･整備

知識	国民の理解獲得に必要な知識（情報）基盤の検討と整備　（協学官）
人材	技術の継承と発展に必要な研究･開発･評価の能力を備えた人材の継続的養成　（急務）　（学産官）
施設	研究基盤施設（LSTF、THYNC、等）の維持･活用と廃止決定法を含む在り方の検討　（官産）
制度	技術評価プロセスに関する検討、産官学協力の在り方に関する検討、指針の体系化に関する検討　（協官）

（注）
役割分担（色分け）およびカッコ内は、関与者の表示まで

Radiation-induced surface activation (RISA)
放射線誘起表面活性



Assumed mechanism behind RISA

Catholic and anodic reactions by surface irradiation of oxidized metal with radioactive rays.

Activating the surface and increasing surface wettability

Improving heat transfer

Thermal Science & Engineering Vol.12, No.2, (2004).

RISA reaction
→Corrosion control
Radiation measurement

Superhydrophilicity caused by γ-ray irradiation

Improvement of the critical heat flux (CHF) requires that the cooling liquid can contact the heating surface, or a high-wettability, highly hydrophilic heating surface, even if a vapor bubble layer is generated on the surface.

To solve these problems why don't we use γ-ray irradiation ?

> Very low efficiency for surface activity

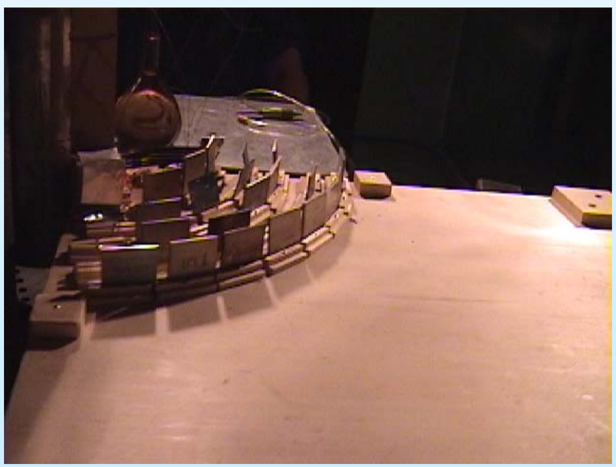
> discrepancy between its wave energy and the valence electron band for TiO_2 and other metal oxides

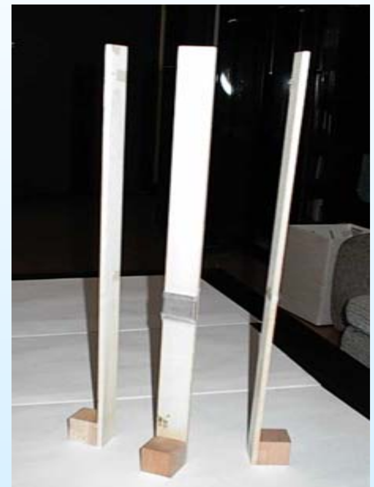
First RISA study (2000-2002)

Takamasa, Hazuku, Mishima, Okamoto, Furuya

1. Improvement of surface wettability by use of an oxide-coated material under a radiation environment.
2. Improvement of thermodynamic properties.

1) Surface Wettability in Room-Temperature (2000)

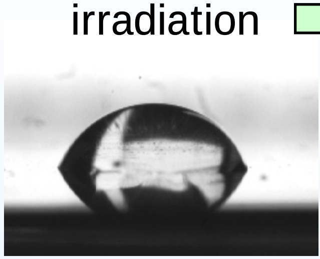
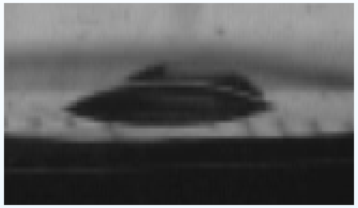
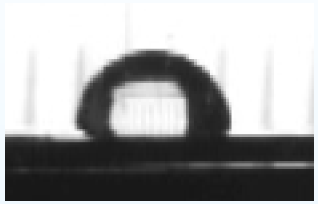
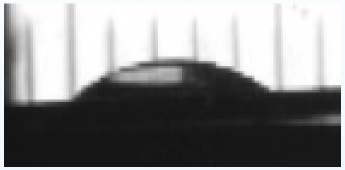




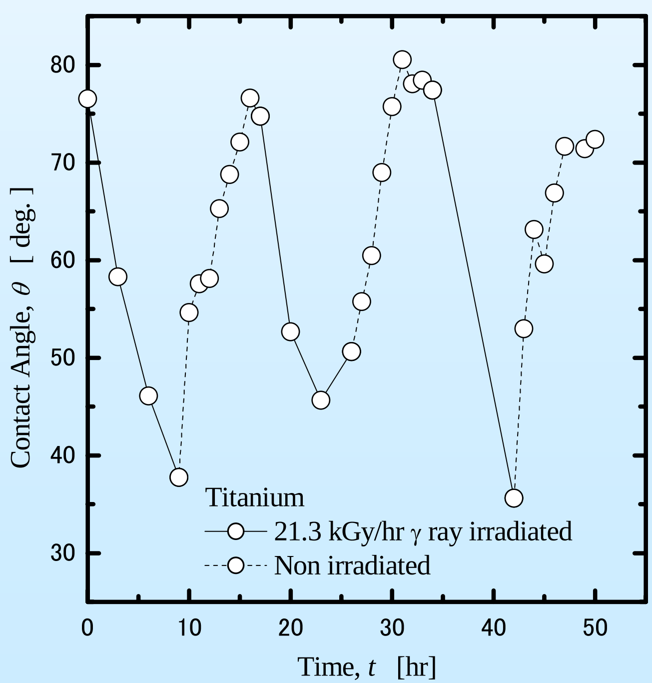
Two ^{60}Co γ-ray facilities at the University of Tokyo and Kyoto University (Radiation ray intensity: 0.1 – 20 kGy/hr)

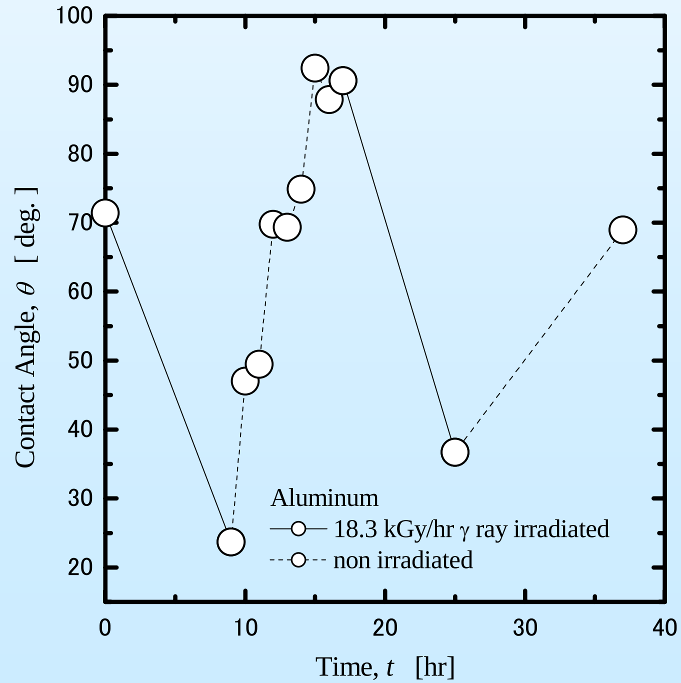
Cyclic change of contact angle

Before irradiation → After irradiation (160 kGy) → 8hr non irradiation → After irradiation (140 kGy) → 12hr non irradiation

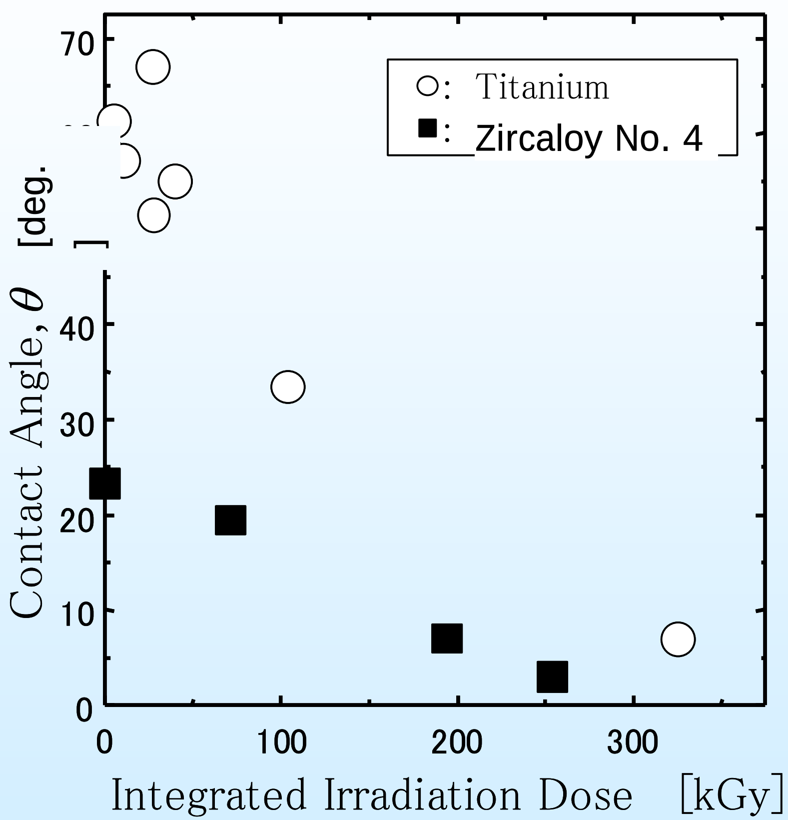






Contact Angle : 71.4° → Contact Angle 23.7° → Contact Angle : 90.6° → Contact Angle : 36.7° → Contact Angle : 68.9°

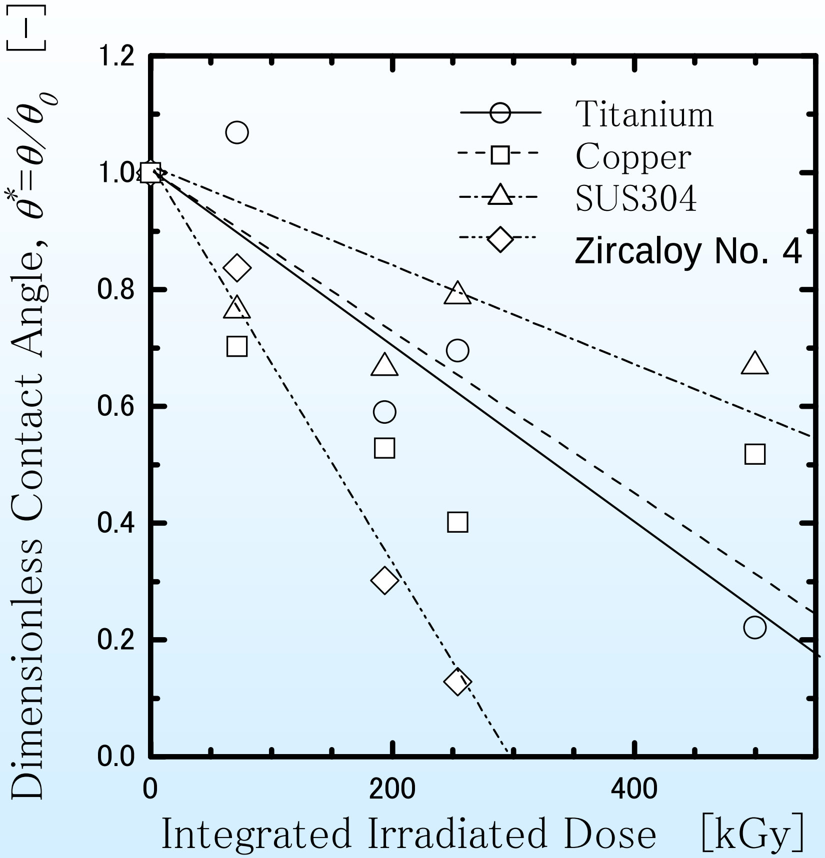




Changes of contact angle by γ-ray irradiation



Change of contact angle by γ-ray irradiation

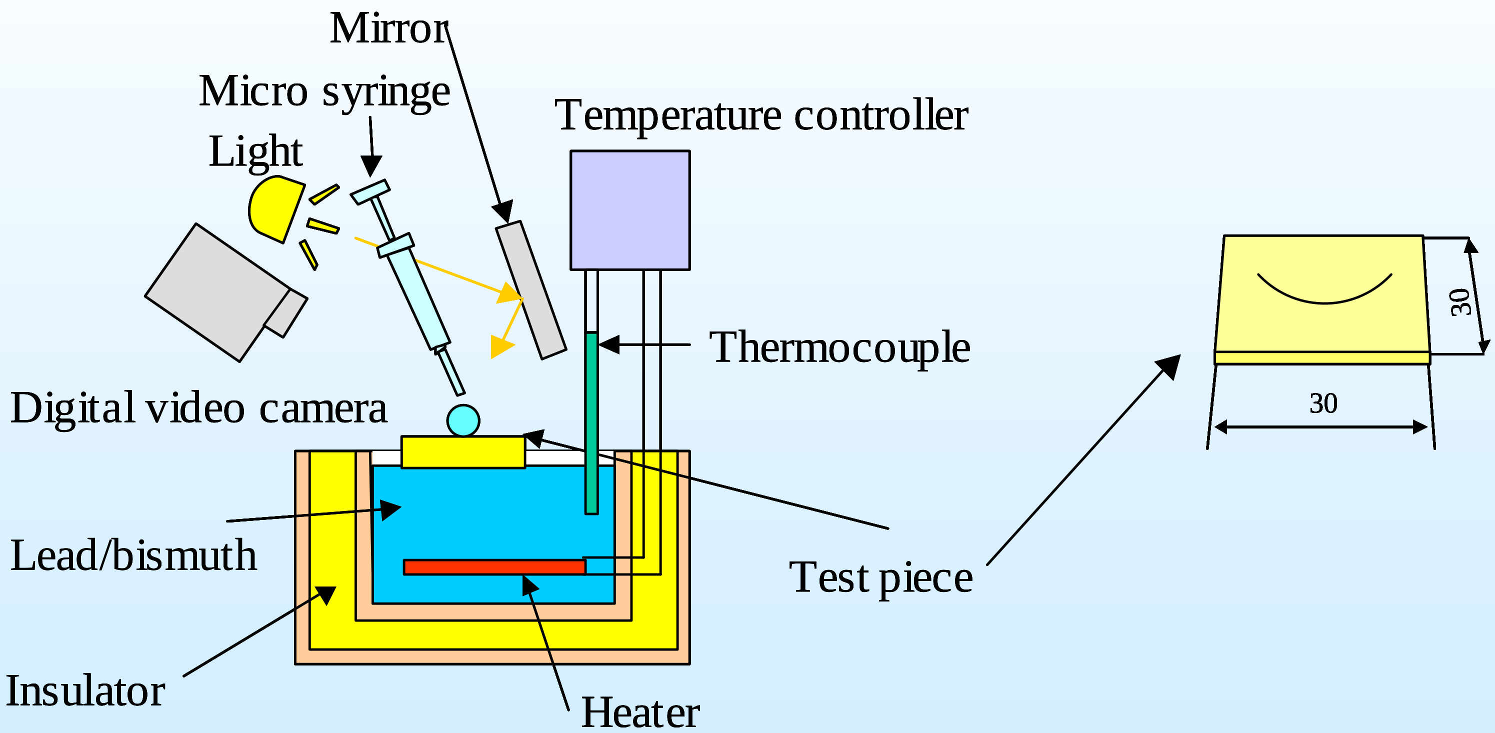


Dimensionless contact angle of test pieces

Radiation Induced Surface Activation (RISA) exists when γ-rays irradiate the surface of metal oxides.

TUMSAT
Power and Energy Lab.

2) Leidenfrost Temperature



Apparatus for Leidenfrost temperature measurement

Droplet Behavior near Leidenfrost Condition

Titanium (300℃)

Before γ-ray irradiation

Over wetting limit temperature

After 260KGy γ -ray irradiation

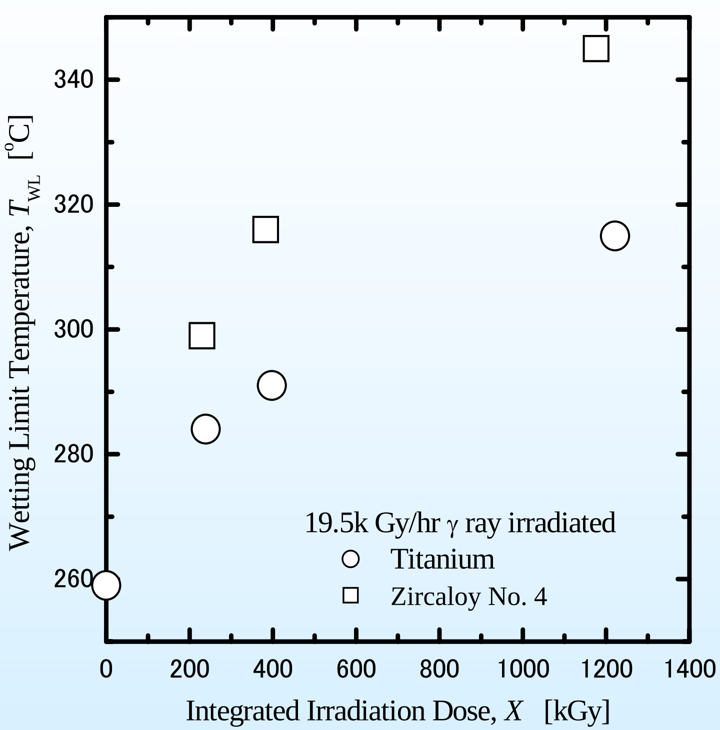
Under wetting limit temperature

Leidenfrost condition from the observation of wetting limit temperature.

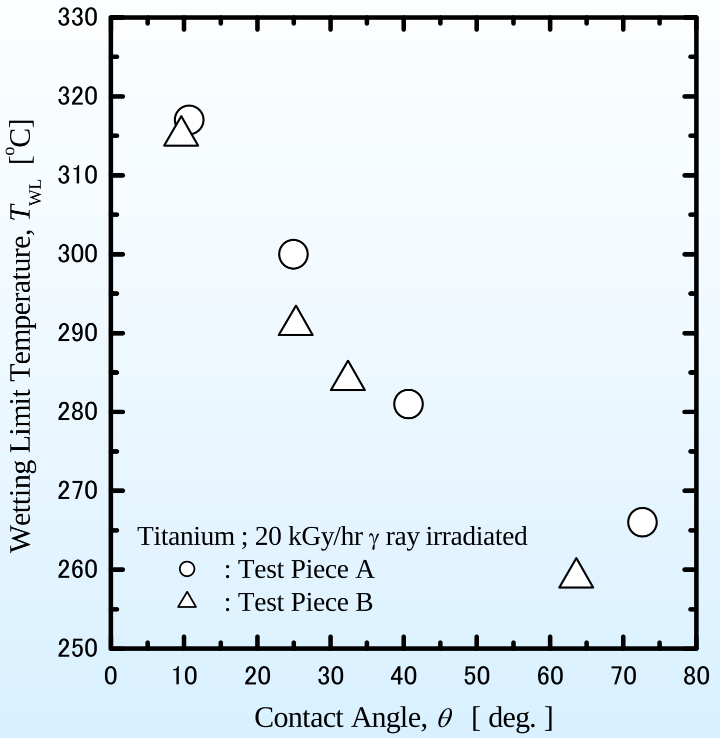
Wetting limit temperature was defined as the maximum temperature of heating surface when splashed droplets contact again with heating surface.

Takata et al., (1990)

Wetting Limit Temperature



Effect of integrated irradiation dose on wetting limit temperature (19.5 kGy/hr)



The wetting limit temperature against contact angle

Enhancement of surface wettability contribute to the improvement of Leidenfrost condition.

TUMSAT
Power and Energy Lab.

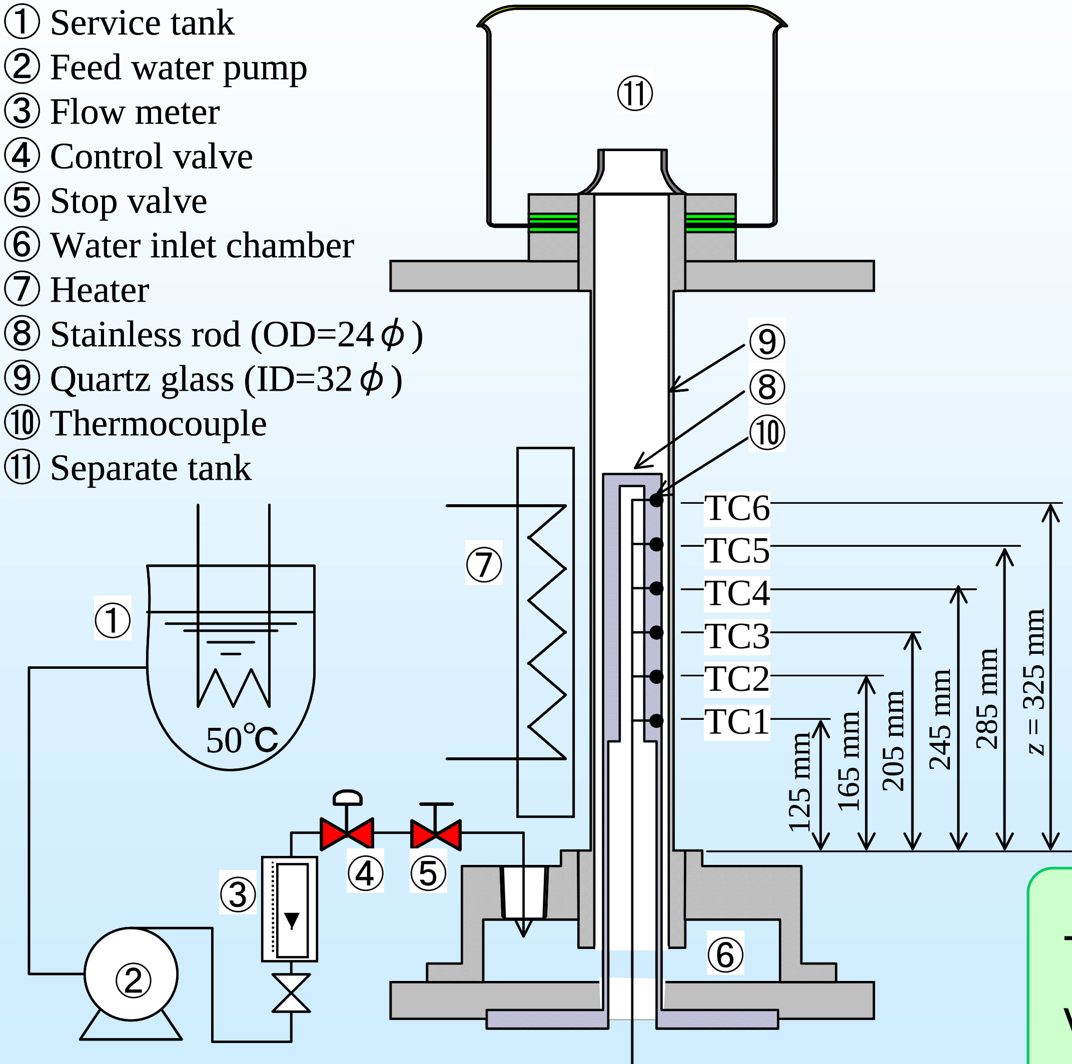
METI-RISA Project 2003-2007 (Japan)
Innovative Nuclear Technologies
Based on Radiation Induced Surface Activation (RISA)

The aim of this project is to get corrosion protection of structural materials in BWR by lowering their corrosion potential and to increase critical power of BWR fuel bundle and refloding velocity under LOCA events, resulting in excellent BWR with higher cost performance and safety.

This project investigates basic technologies of the RISA such as film forming, electrochemical dynamics, film durability in the reactor and improvement of heat transfer performance in reactor core in order to apply them to BWR.

Kobe Steel Limited
Tokyo Univ. Marine Sci. & Tech.
University of Tokyo
Kyoto University
Japan Atomic Energy Agency
National Maritime Research Institute
Central Research Institute of Electric Power Industry
Institute of Research and Innovation
Toshiba Corporation

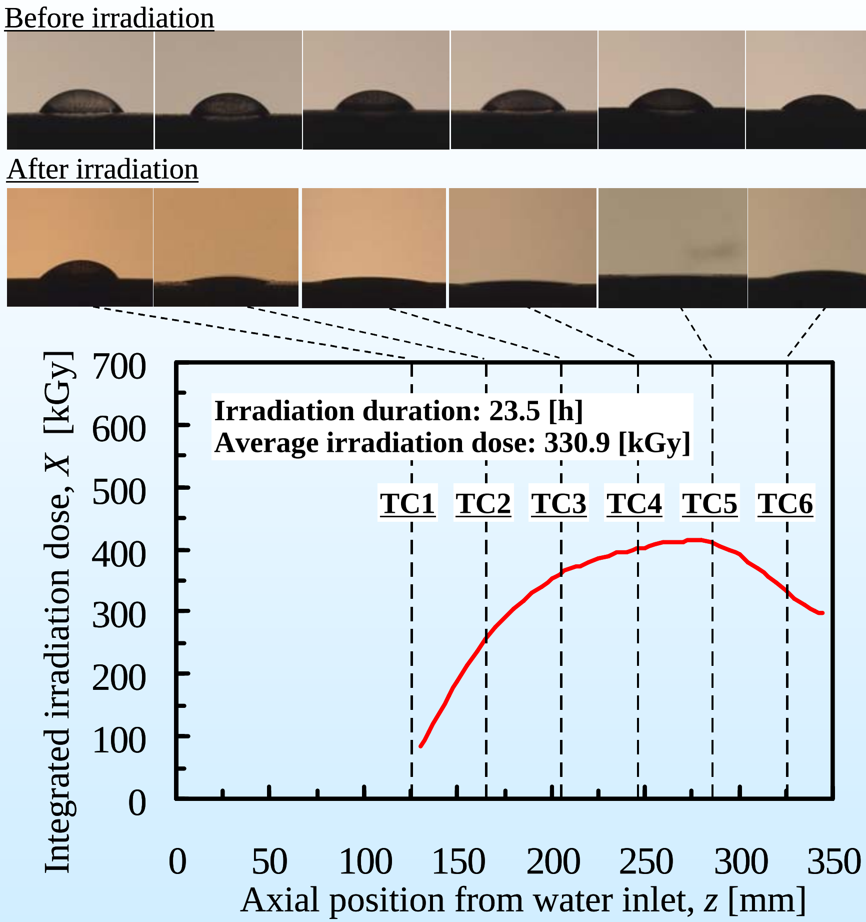
b. Quenching Experiment



Thermal Science & Engineering
Vol.12, No.2, (2004).
Takamasa, Mishima, et al

Apparatus for reflooding experiment

Rod Surface Wettability Change due to γ-ray Irradiation



Contact condition of a water droplet on stainless rod before and after γ-ray irradiation

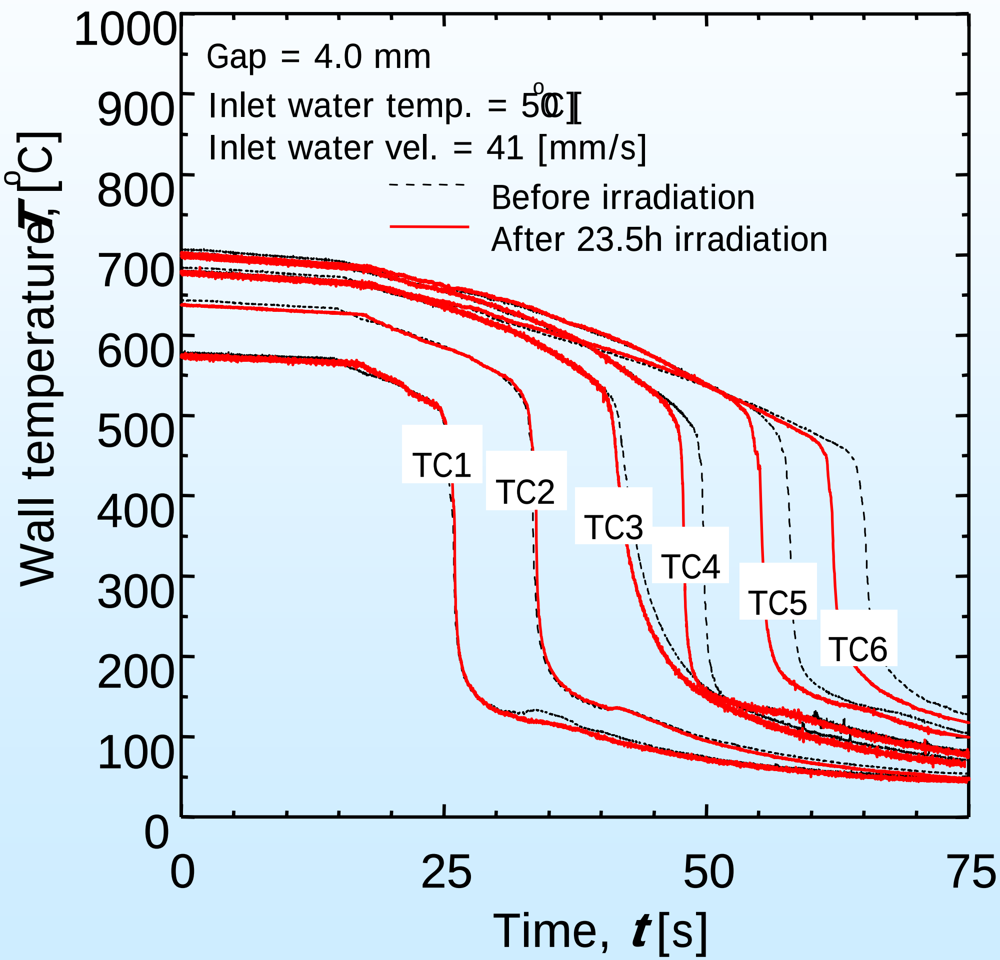
· Profile of integrated-irradiation dose on the rod forms centered peaked along the rod axis.

· Superhydrophilic condition of oxidized metal surface can be achieved after integrated irradiation dose of 300-500 kGy, located at the rod center, z = 248 mm (TC4) and 285 mm (TC5).

·Surface wettability of rod end is consistent before and after irradiation.

TUMSAT Power and Energy Lab.

Typical Result of the Surface Temperature Record



· No discrepancy exists in temperature records between before and after irradiation at TC1 and TC2 where no changed wettability was observed.

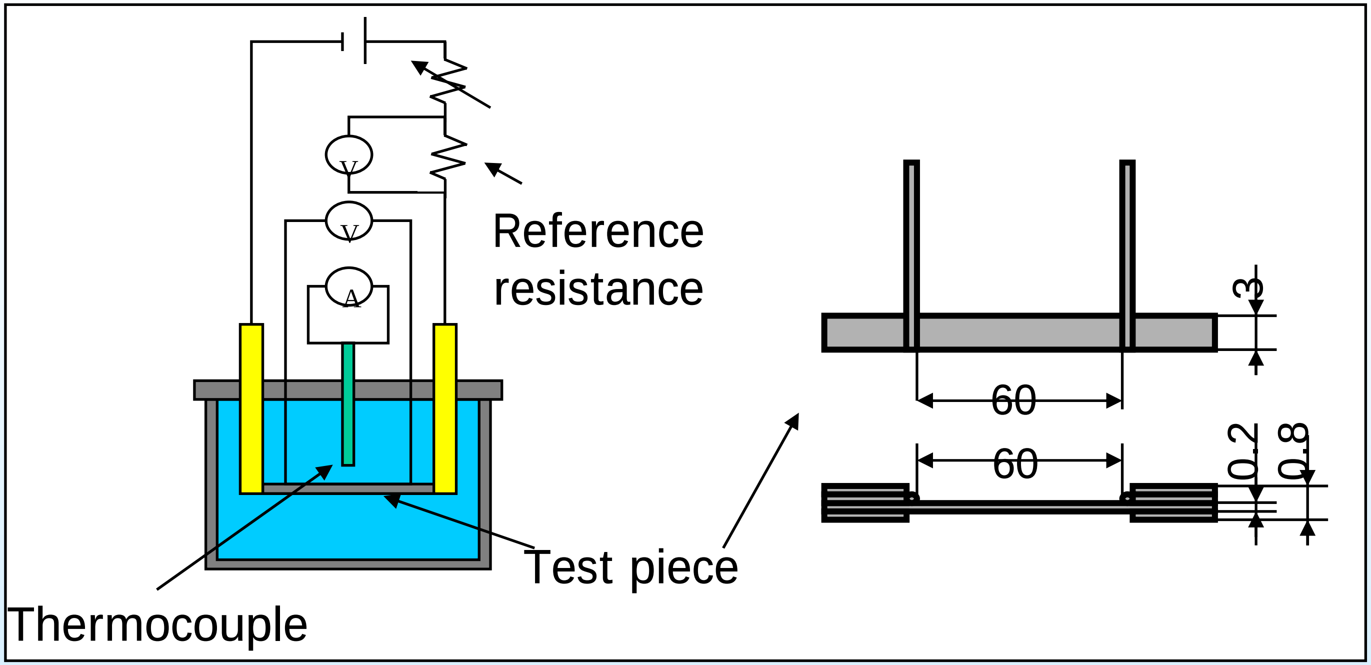
⇒ Reproducibility of the test

· Large increased quenching velocity, 7.1 mm/s, was observed at the middle elevation of rod (TC3 and TC4) after γ-ray irradiation.

·The quenching velocities were increased up to 20-30 % after 300kGy ^{60}Co γ-ray irradiation.

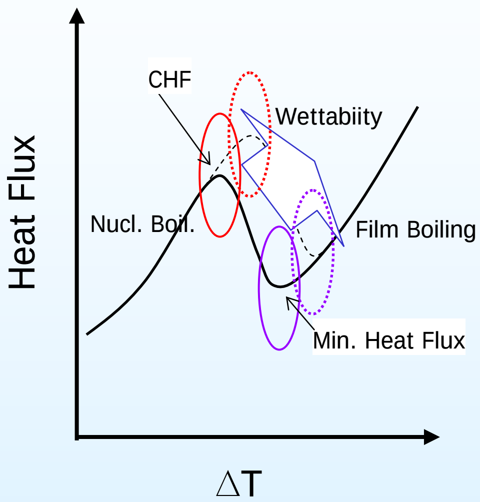
TUMSAT
Power and Energy Lab.

3) CHF in pool

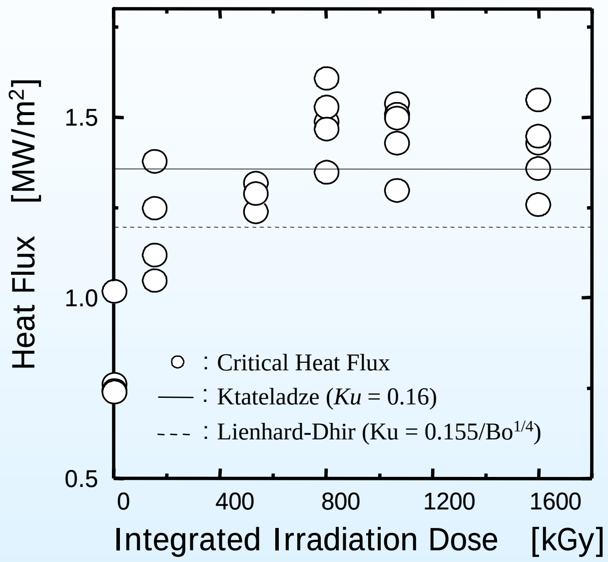


CHF experiment: the pool boiling condition
The pressure: atmospheric pressure
(resulting in the boiling point to be 100 degree C)
The heating: conducted using the Joule heating by DC supply
The test piece: hold horizontally on the electrode
To generate the oxidized surface, the test pieces were oxidized using plasma jetting for 40 seconds.

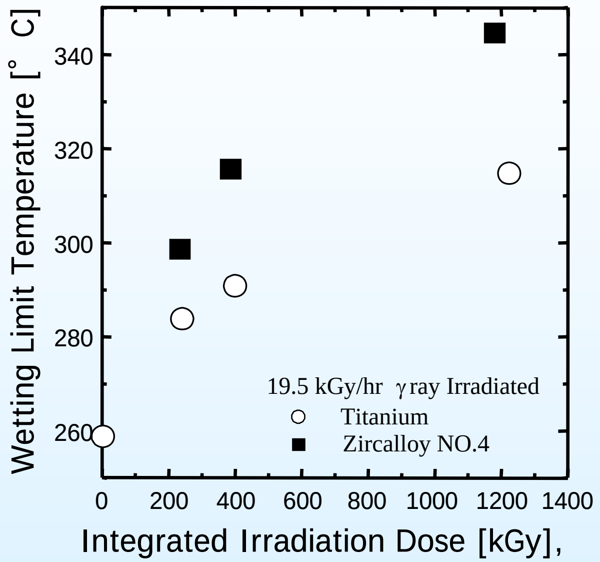
Boiling heat transfer improved with surface wettability
(Takmasa et.al. 2003)



Change in Boiling Curve



CHF in Pool Boiling



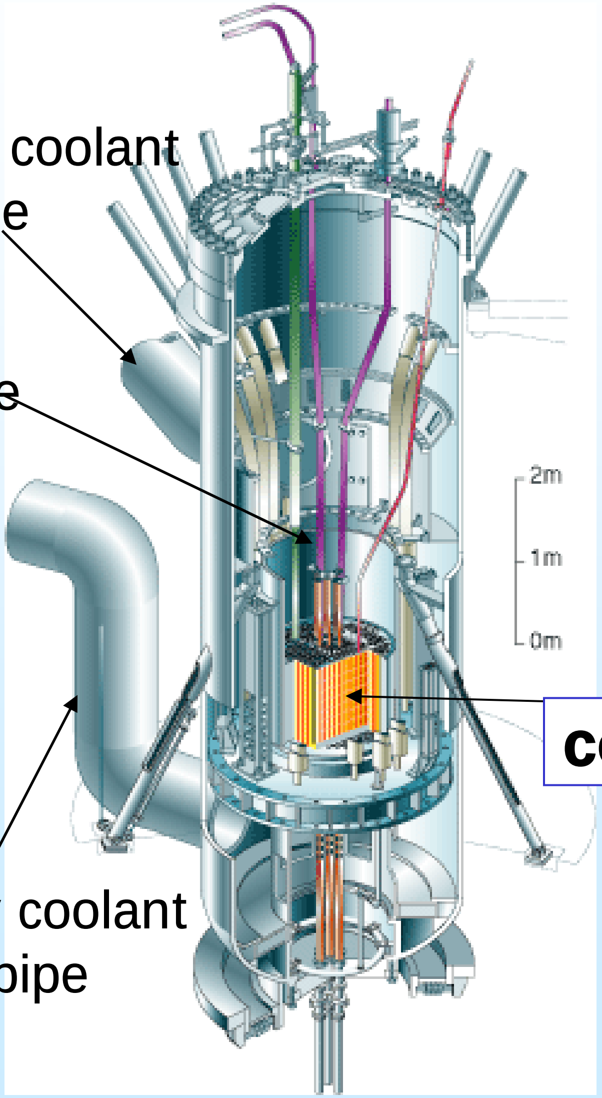
Increase in WLT

- ✓ Critical Power Improved
- ✓ Quench (front) speed increased
- ✓ Past experiments in NSRR & Halden suggest the possibility

Increase in Safety Margin
Natural characteristics to confirm

TUMSAT Power and Energy Lab.

4) Reactor experiments for CHF/RISA 2005



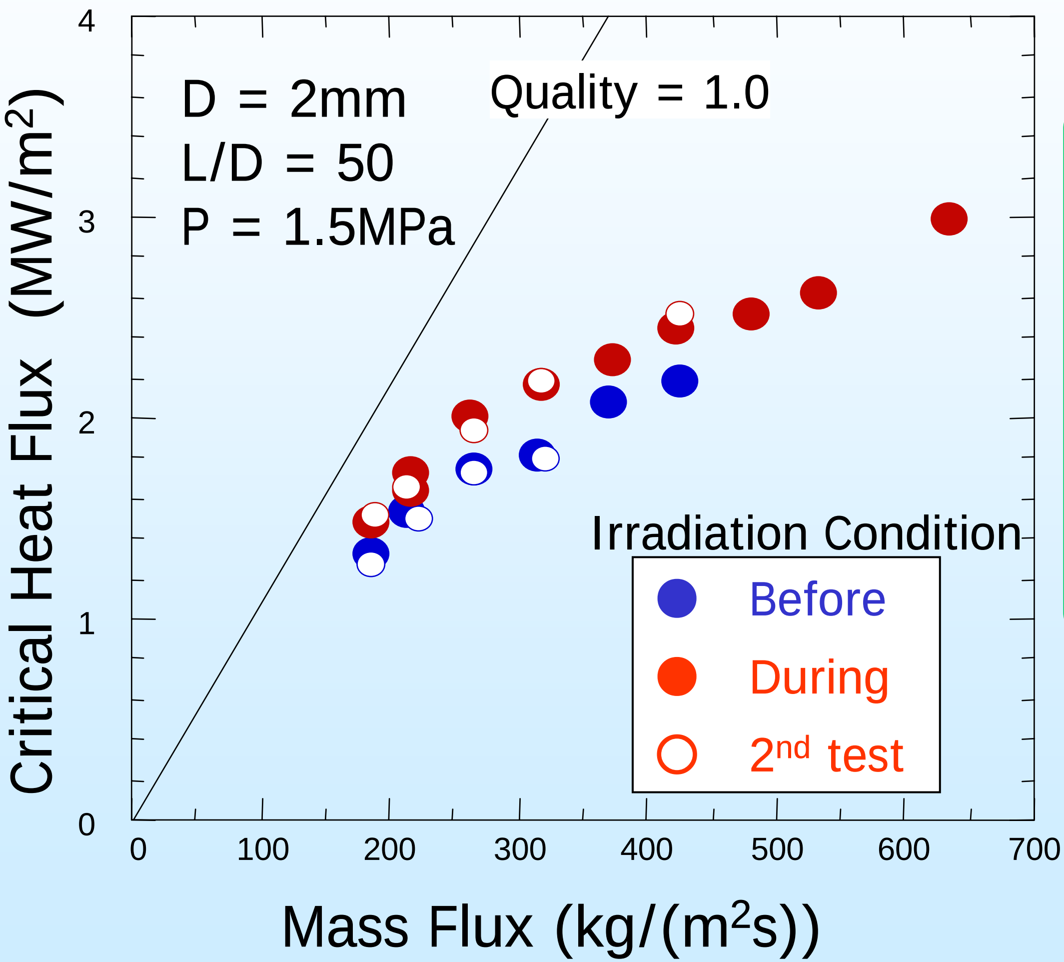
Japan Materials Testing Reactor (JMTR) JAEA

Reactor type : Light water moderation
Thermal power : 50 MW
Primary coolant
Inlet temp. : 322 K
Outlet temp. : 329 K
Flow rate : 6000 m^3/h
Pressure : 1.5 MPa
Neutron flux : max.; $4.0 \times 10^{18}/m^2/s$
RISA exp.; $1.0 \times 10^{17}/m^2/s$

- An outermost irradiation hole will be used in the core to minimize gamma-ray heating ratio.
- Absorbed dose is 100 times larger than that in the previous experiments.

TUMSAT
Power and Energy Lab.

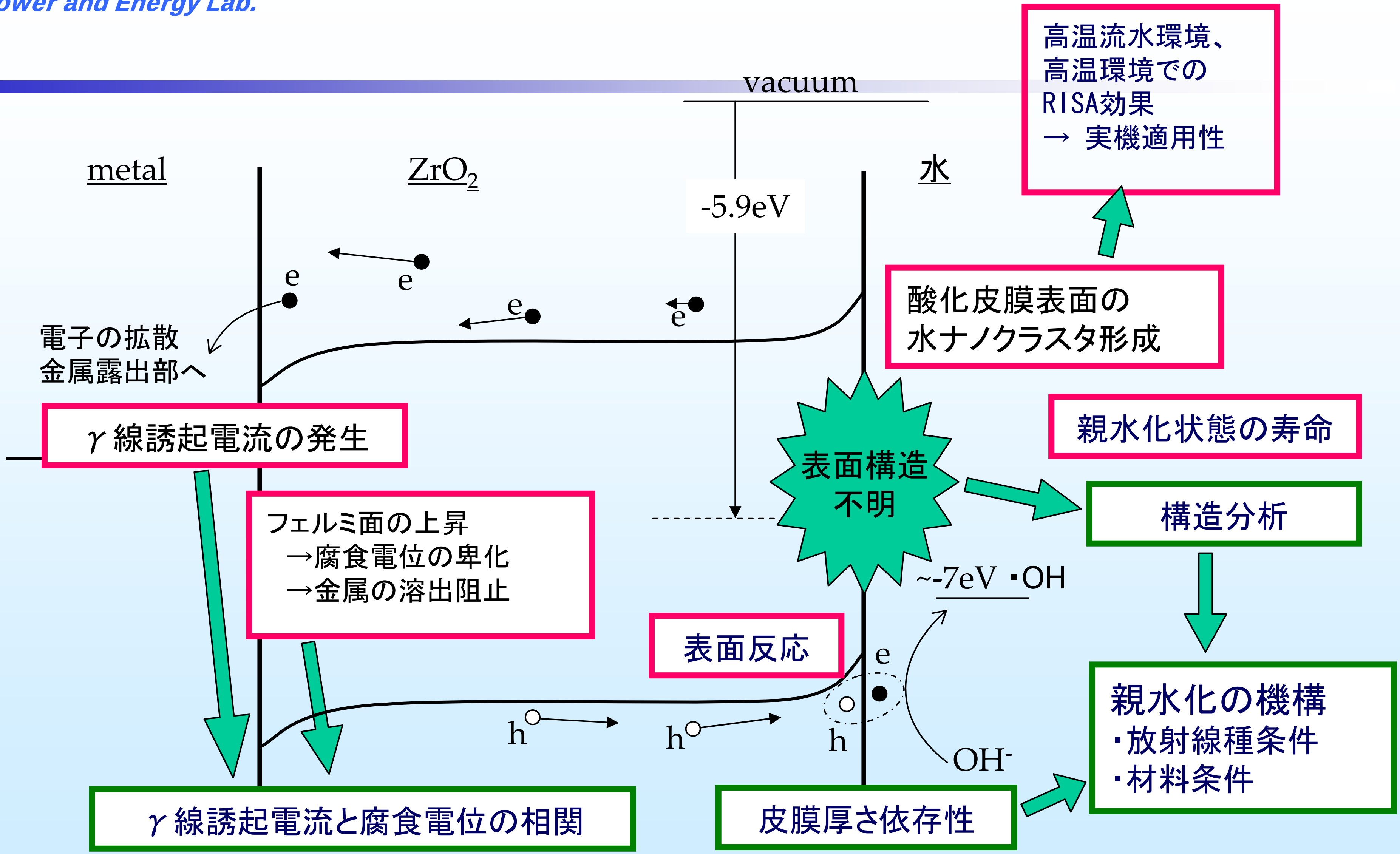
CHF results



- ✓CHF increased by 17% due to irradiation in JMTR
- ✓CHF appeared at high quality conditions
- ✓CHF mechanism would be dryout-type similar to that in BWR core

TUMSAT Power and Energy Lab.

3. Mechanism behind RISA



TUMSAT
Power and Energy Lab.

RISAメカニズムの不明点（東大　阿部）

(A)放射線照射による励起過程に及ぼす皮膜・基板及び水の効果
(B)放射線種の効果
(C)表面反応電荷
（D）放射線照射によって励起し親水化に寄与する物質とその形態
（E）高温におけるRISA

Analysis by AFM (Atomic Force Microscope) and FFM (Friction FM)

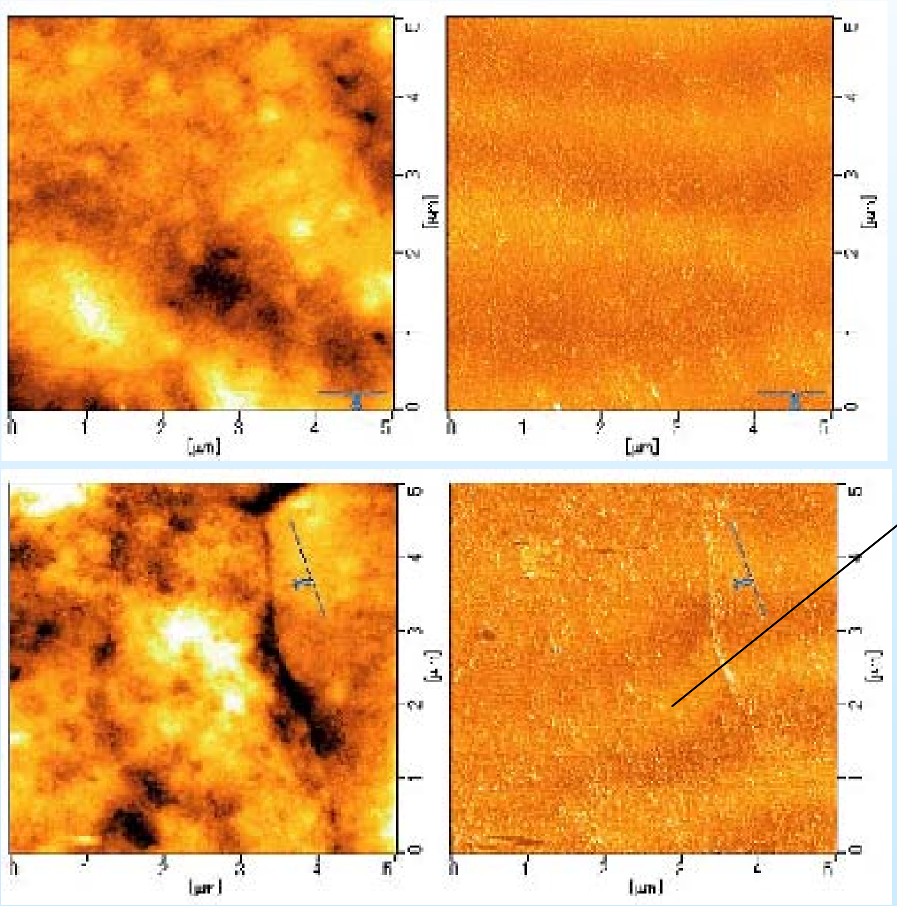
(D)放射線照射によって励起し親水化に寄与する物質とその形態

Substrate; Zircaloy-4
Oxide layer; by autoclave
Irradiation; No irradiation
Measurement; at r.t. in air
Contact angle; 77 deg.

↓

Substrate; Zircaloy-4
Oxide layer; by autoclave
Irradiation; 340 kGy
Measurement; at r.t. in air
Contact angle; 12 deg.

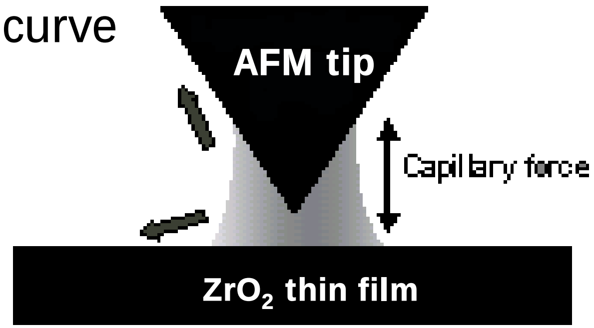
AFM image | FFM image

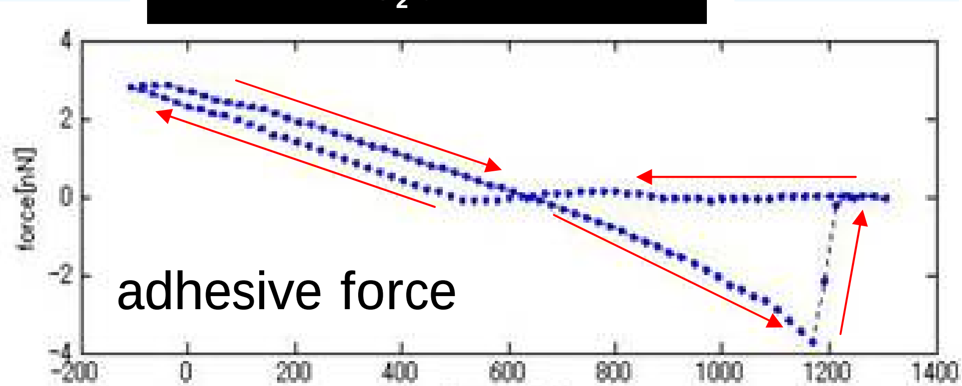


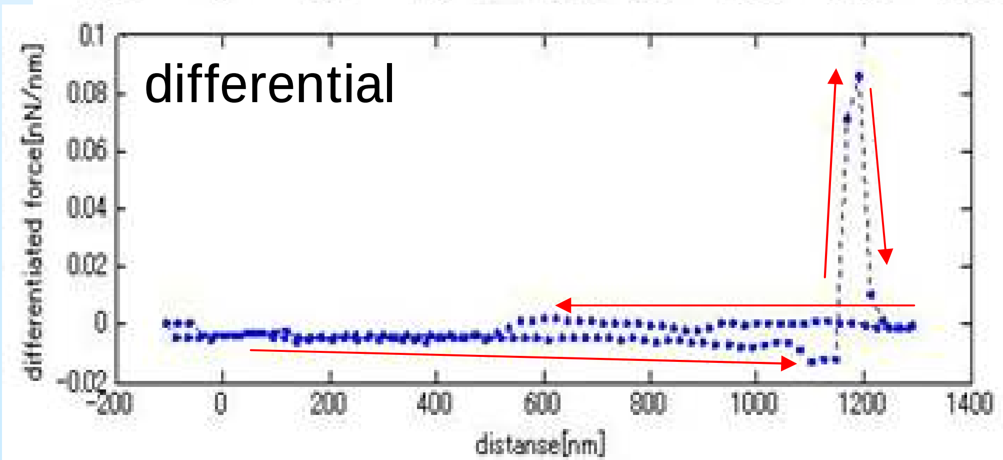
White spots are observed

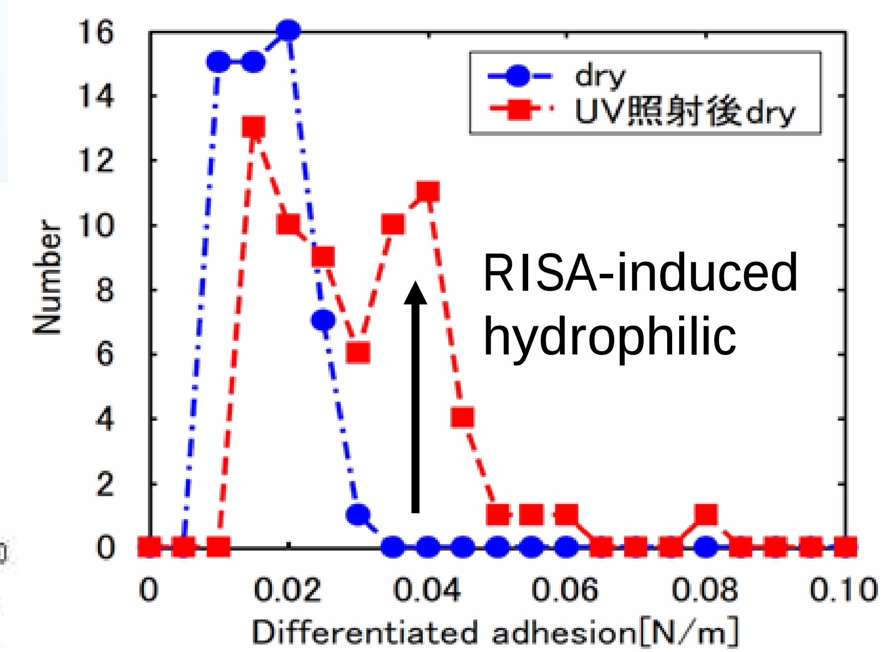
Wettability of ZrO_2 surface at AFM scale

Force curve









matrix water?

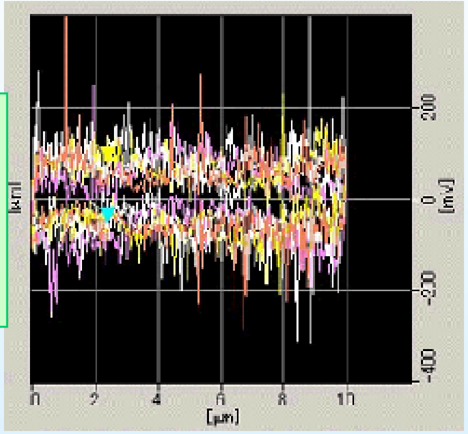
hydrophilic clusters

TUMSAT
Power and Energy Lab.

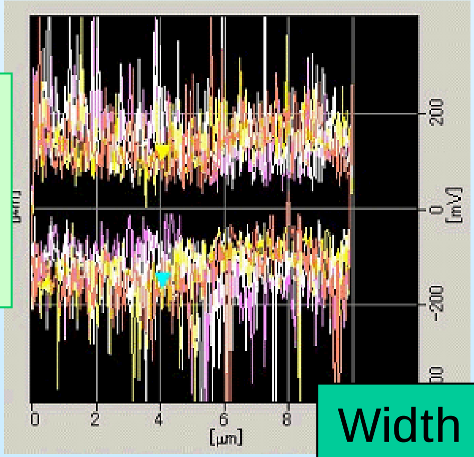
Analysis by AFM

- Surface friction force of the Zircaloy samples increases by gamma ray irradiation.

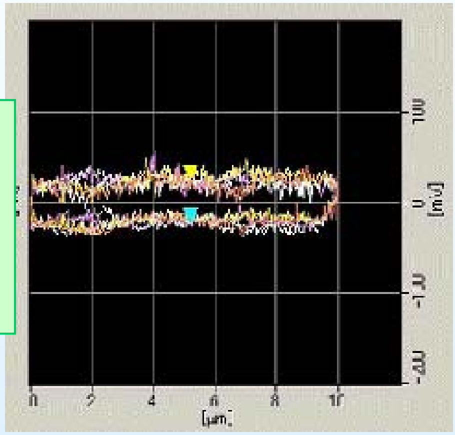
Substrate; Zircaloy-4
Oxide layer; by autoclave
Irradiation; No irradiation
Measurement; at r.t. in air
Contact angle; 77 deg.



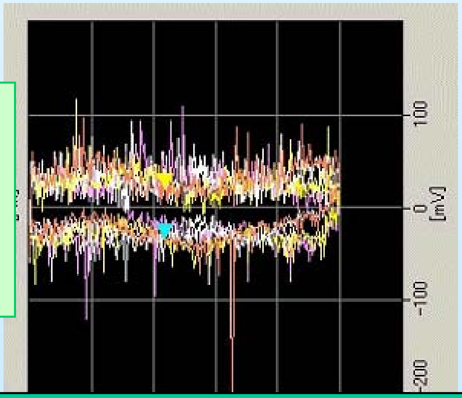
Substrate; Zircaloy-4
Oxide layer; by autoclave
Irradiation; 340 kGy
Measurement; at r.t. in air
Contact angle; 12 deg.



Substrate; SUS304
Oxide layer; heated in air
Irradiation; No irradiation
Measurement; at r.t. in air
Contact angle; 95 deg.



Substrate; SUS304
Oxide layer; heated in air
Irradiation; No irradiation
Measurement; at r.t. in air
Contact angle; 39 deg.



Width between upper line and lower line corresponds to magnitude of surface friction force.

TUMSAT
Power and Energy Lab.

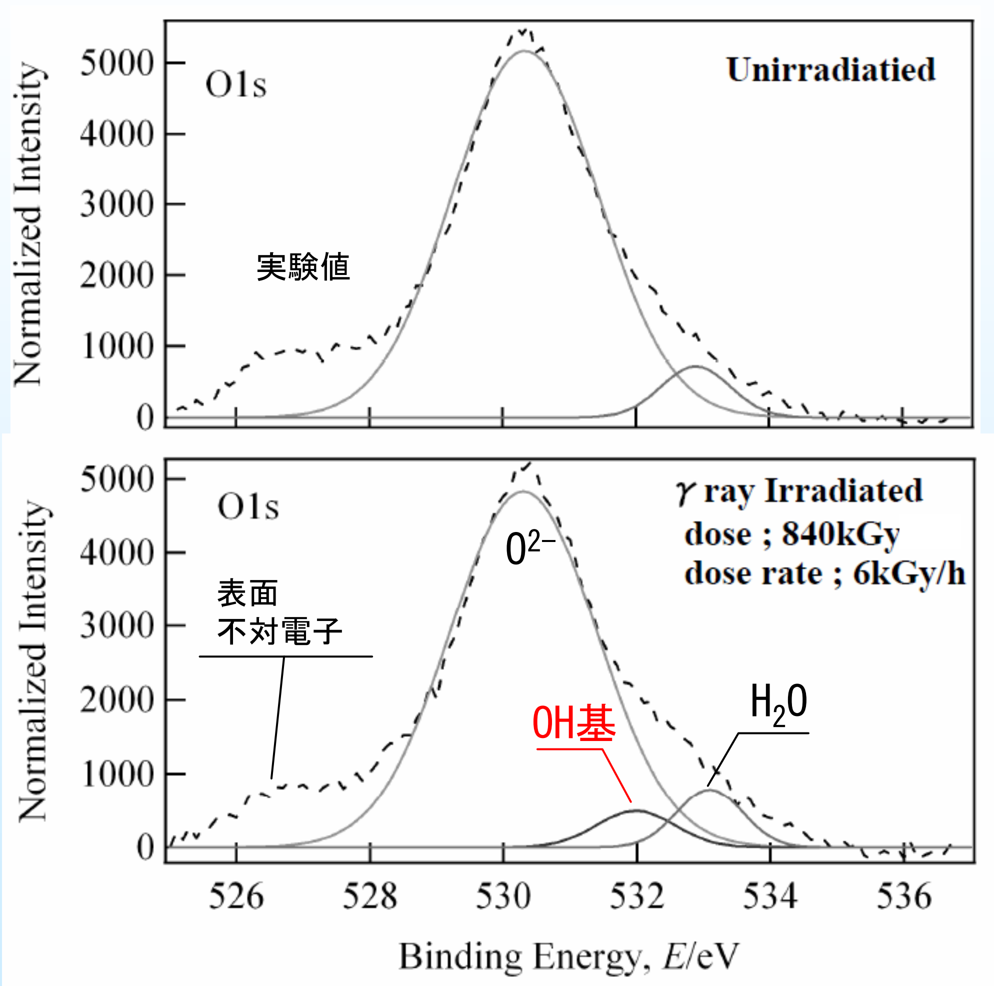
Surface Wettability

放射線照射によって形成された正孔は酸化被膜表面に拡散し溶液中の電子と再結合消滅し、適切な不純物準位や欠陥準位が存在すると励起効率は向上する。この時の電気化学反応は酸化ジルコニウムの荷電子帯上端の水素基準電位（**Standard Hydrogen Electrode (SHE)**）に対する電位（**-3.9eV vs. SHE**）と表面におけるバンドの褶曲を考えると

$$OH^{-} + h^{+} \rightarrow \cdot OH \quad (-2.5eV\ vs.\ SHE) \qquad (1)$$

であると考えられ、形成された**OH**基によって表面の水ナノクラスタが励起され、これが親水化に寄与する。

表面構造分析結果（その１）　最表面にOH基が形成される

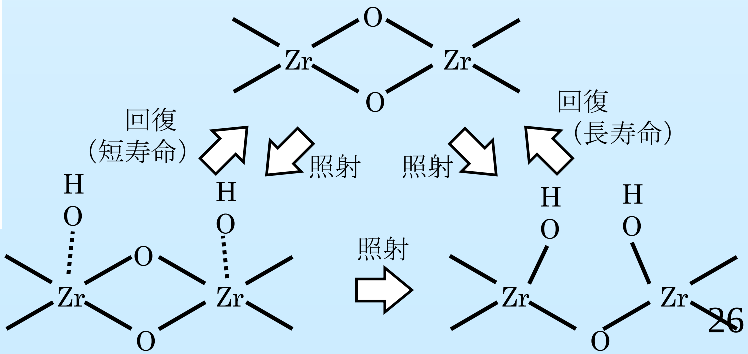


実験方法

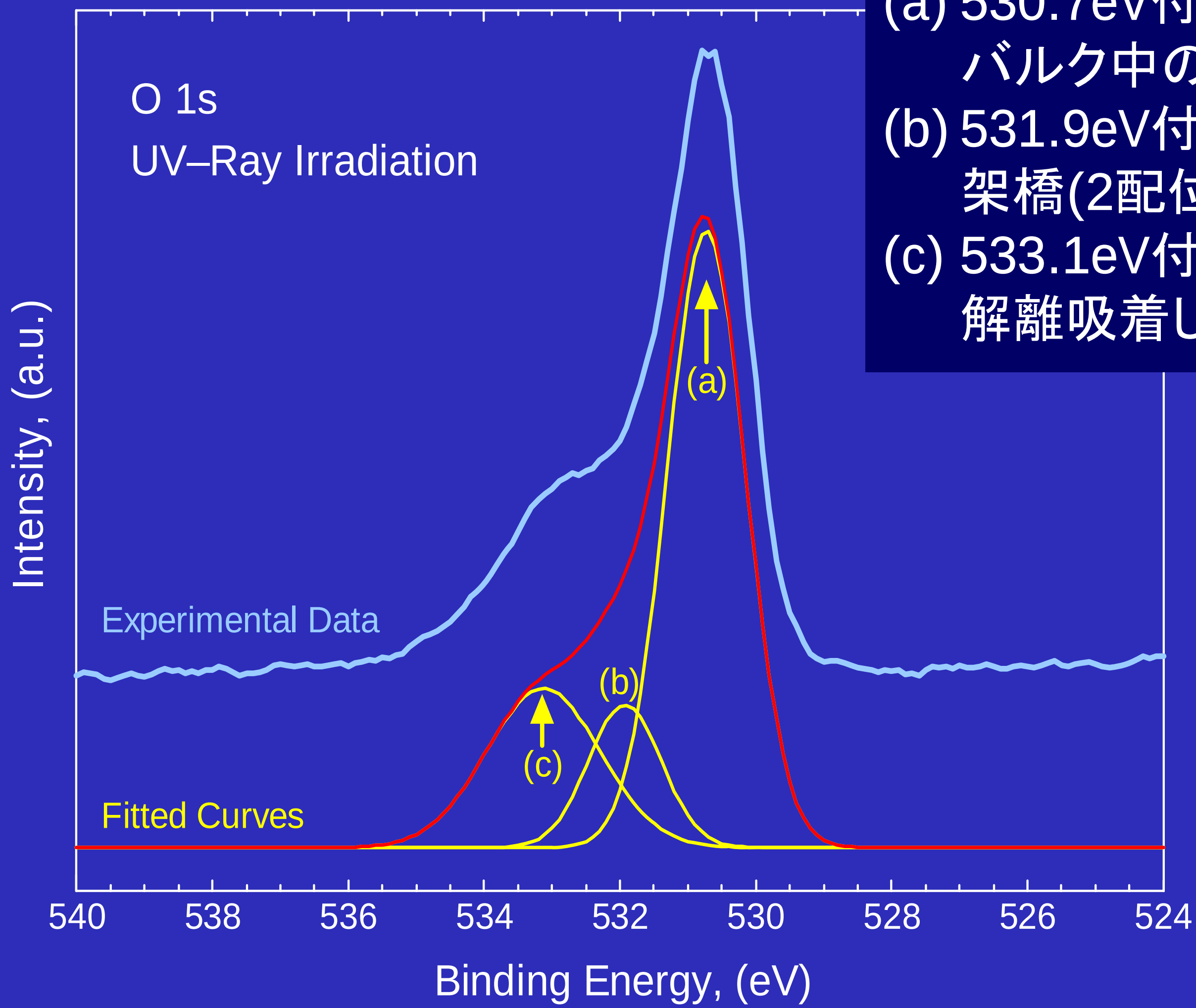
材料：m-ZrO_2粉末予焼鈍材
照射：γ線840kGy、室温、大気中
分析：XPS

考察

　γ線照射によりZrO_2表面にOH基が形成されることがわかった。またH_2Oの割合も相対的に増加した。
　親水化因子として表面OH基（Zr-OH）に由来した水クラスタを提唱してきたが、これを支持する結果である。



XPS分析結果　紫外線照射

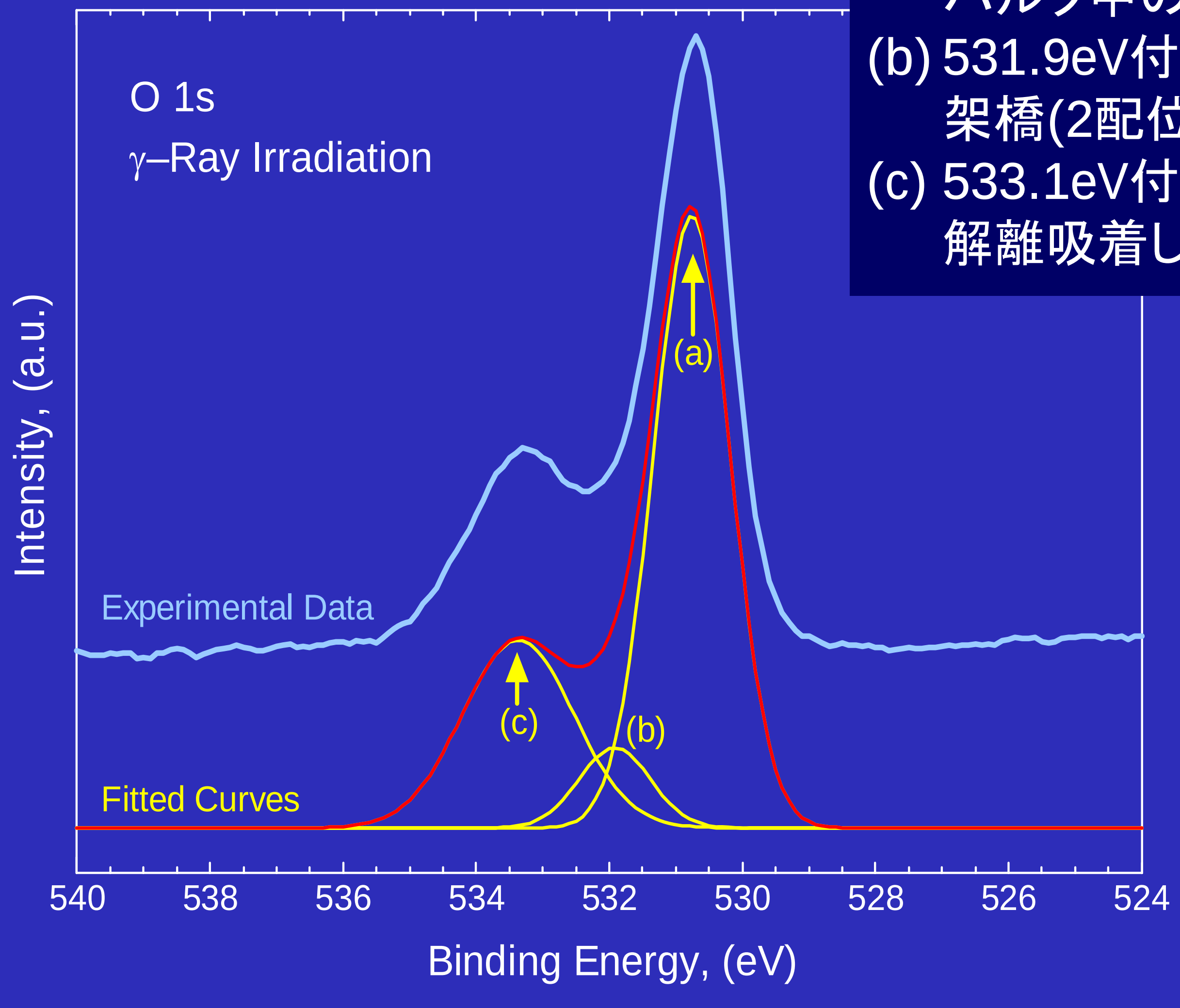


(a) 530.7eV付近
　バルク中の(3配位酸素)Ti-O結合
(b) 531.9eV付近
　架橋(2配位)酸素のTi-O結合
(c) 533.1eV付近
　解離吸着した水酸基(OD)

2 mW/cm^2
60 h

Furuya(2008)

XPS分析結果　γ線照射



(a) 530.7eV付近
バルク中の(3配位酸素)Ti-O結合
(b) 531.9eV付近
架橋(2配位)酸素のTi-O結合
(c) 533.1eV付近
解離吸着した水酸基(OD)

1.5 kGy/h
85 h

Furuya(2008)

冷却材喪失事故に関する安全解析
V&V (Verification and Validation) 越塚

RISAの影響は考えなくてOK?

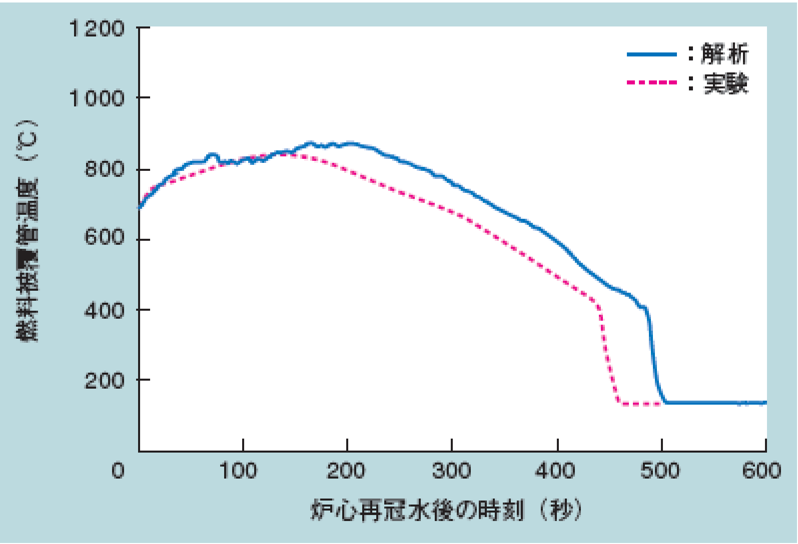


図３　MCOBRA/TRAC コードによる CCTF 試験解析例
炉心の最高出力領域において燃料被覆管最高温度は，解析結果が試験結果を上回っている．

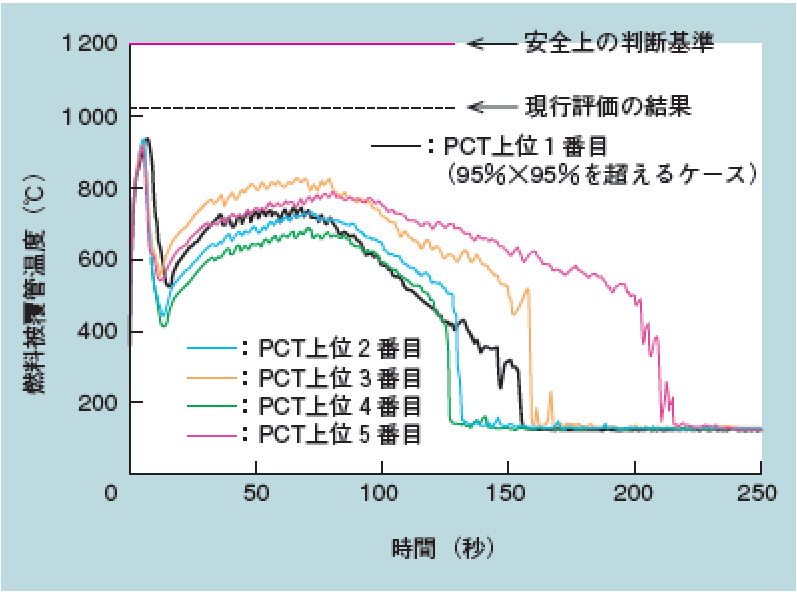


図４　順序統計法による計算結果例
燃料被覆管温度（最高値が上位１番目から５番目までのケース）の時間変化である．

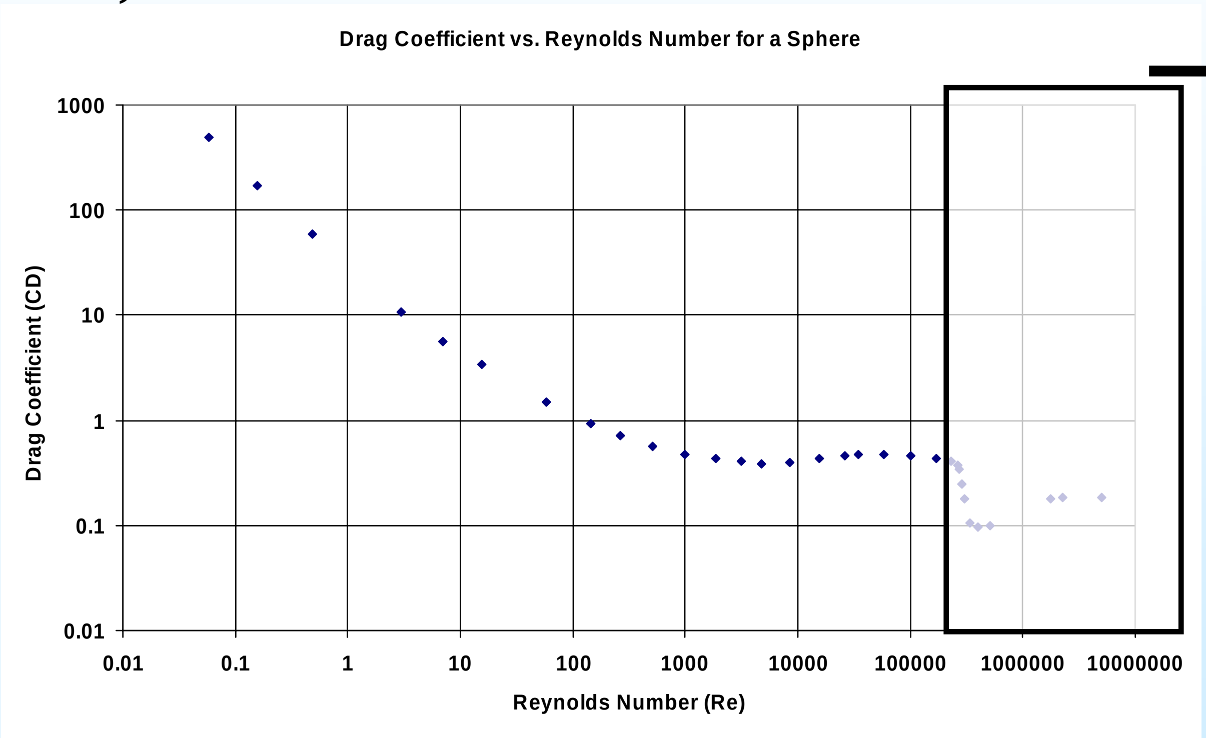
モデル実験との比較によるvalidation

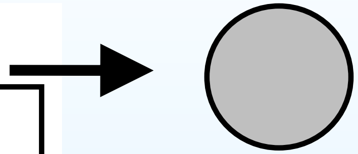
実炉の解析

寺前ら, 三菱重工技報 43(4), 25-31 (2006)

外挿に対する懐疑　越塚

- 球の抗力係数は、1,000<Re<100,000でほぼ一定。
- Re=200,000付近で急激に低下。





Re<100,000の妥当性確認では、Re>100,000の値を予測できない。
→　弱い放射線環境(1kGy/h-)下の実験結果では、強放射線環境下(1MGy/h-)の熱流動現象を予測することができない？

熱水力開発・安全関連技術　ロードマップ

大分類	新型炉、現行炉の共通課題
個別項目	**炉心健全性**
対応すべき技術テーマ	炉心の限界出力評価、BWR異常過渡変化

役割分担：産　官　学　学協会

	2008	09	10	11	12	13	14	15	16	17	18	19	20	25	30

次世代炉：FS → 概念設計検討 → 概念設計 → 基本設計 → 詳細設計 → 許認可 → 建設 → 運開

BWR異常過渡変化時の炉心健全性（Post-BT 熱伝達）

- 解析モデルやコードの高度化と検証に活用可能な実機条件データベースの拡充
- 最適解析手法の予測性能評価と精度向上

放射線照射表面活性（RISA）による沸騰熱伝達の向上

- 伝熱面表面性状とドライアウト熱流束の系統的調査
- 照射下濡れの計測 など
- 実機炉心条件（照射下、高圧、高流量）でのCHF試験
- 最適解析手法の予測性能評価

技術の検証、規格規準・規制への反映

- 学会標準の策定・改訂
- 炉心の限界出力評価と燃料設計への反映
- 最適評価手法、解析手法の精度向上
- 安全余裕の正確な評価
- 最適評価手法、解析手法の精度向上
- 安全余裕の正確な評価（クロスチェック解析）

基盤の確保・整備（特記事項のみ）

- 施設基盤：研究基盤施設（JMTR、THYNCなど）の維持・活用
- 制度基盤：学会標準や指針の策定・改訂に関する検討