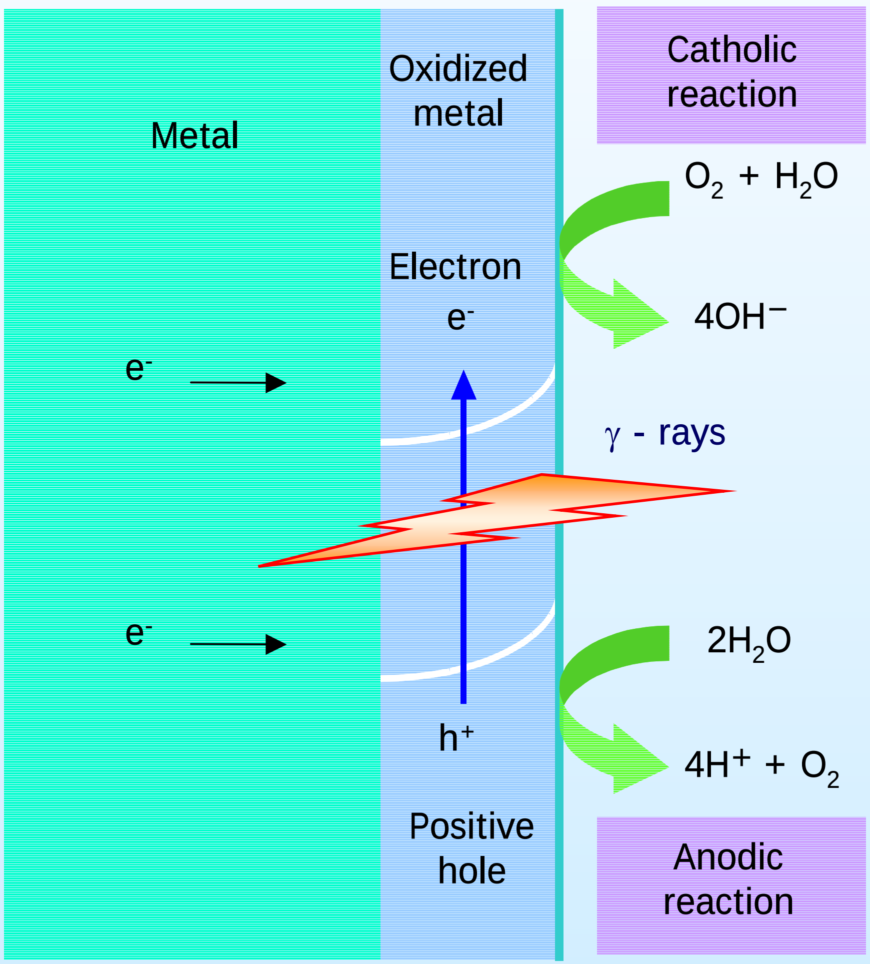
T.U.M.S.A.T. Power and Energy Lab.

Radiation-induced surface activation (RISA)



Assumed mechanism behind RISA

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

Activating the surface and increasing surface wettability

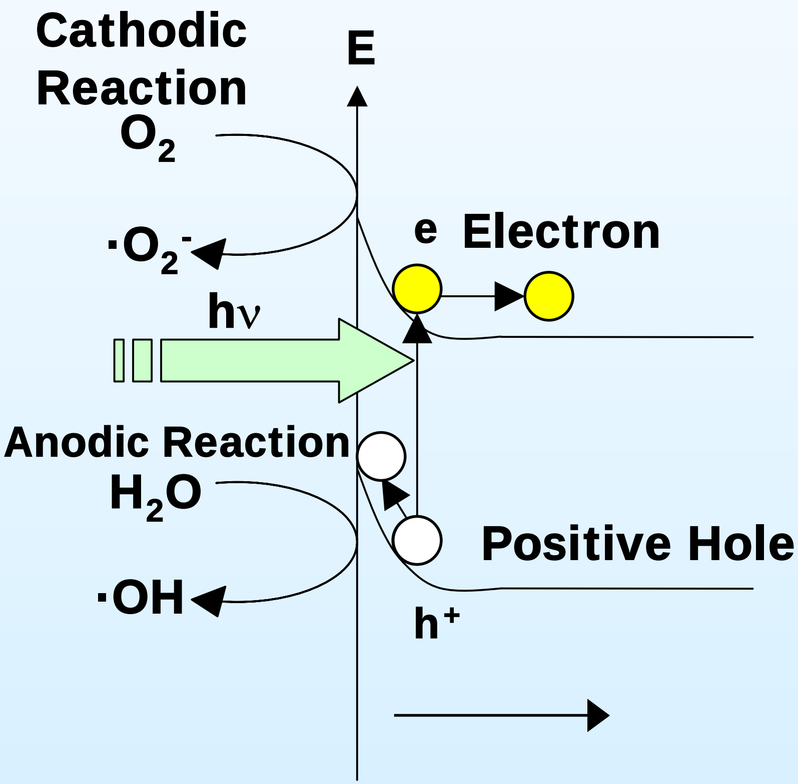
Improving heat transfer

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

RISA reaction
→Corrosion control
Radiation measurement

T.U.M.S.A.T.
Power and Energy Lab.

UV Light Catalyst (Fujishima, 1980)



Electrochemical reaction on the material surface with TiO_2 under ultraviolet (UV) irradiation.

TiO_2 representative n-type semiconductor, is excited by photon irradiation and generates anodic current.

Photon energies which can excite the electron in the valence electron band :

- Rutile type TiO_2: <420nm (Eg=3.0eV)
- Anatase type TiO_2: <390nm (Eg=3.2eV)

This reaction decomposes organic substance by its strong oxidizing power and commonly used in environmental purification as anti bacterial, antipollution and deodorant material.

Superhydrophilicity

When the surface coated with TiO_2 is irradiated by UV light, the contact angle for water decreases with the irradiation and finally reaches almost *zero*. (Fujishima, A., et al., Nature, (1997))

TiO_2-coated glass : exposed to water steam, the droplets on the surface form very thin film and the glass becomes transparent.

The self-cleaning effect : remove oil materials from the surface because the TiO_2-coated surface has greater affinity for water than oil.

the anti-fogging side-mirror film, coating of automobiles and various materials that can be self-cleaned by rainfall.

Thermohydoulic Application

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.

Boiling and quenching with a superhydrophilic surface

Takata, Y., et al., Thermal Science & Engineering, (2000)

(1) The critical heat flux (CHF) of TiO_2-coated surface is about two times larger than that of a non-coated one.

(2) The minimum heat flux (MHF) temperature for a TiO_2-coated surface is much higher than that of non-coated one in a quenching experiment.

Possibility of application of UV catalysis to improve heat transfer incidents

Difficulties of UV Light Catalyst in Actual Heat transfer Applications

1. Need a light source and a transparent wall
2. Only TiO_2 –can be used
3. Unstable surface – cannot be used over a long period

Conventional oxide matels, Zircaloy, SUS ?

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

> Very low efficiency for surface activity

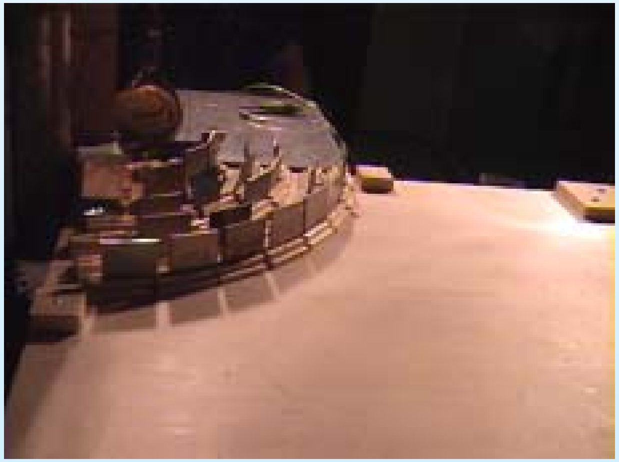
> discrepancy between its wave energy and the valence electron band for TiO_2 and other metal oxides

First RISA study (1999-2002)

Takamasa, Hazuku, Mishima, Okamoto

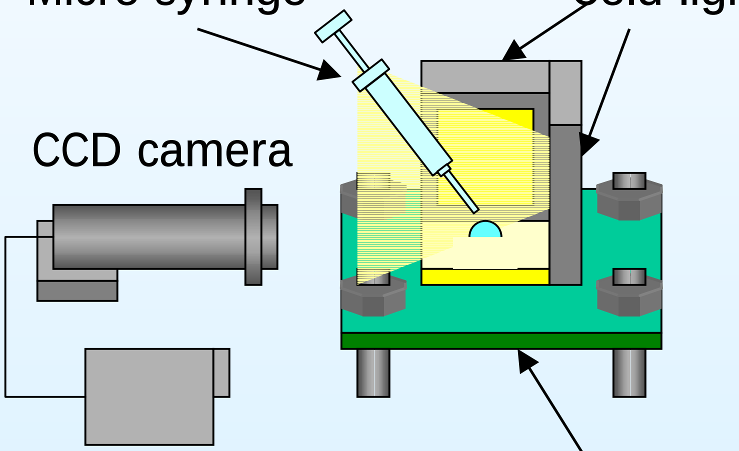
1. Improvement of surface wettability by use of an oxide-coated material under a radiation environment.
2. Improvement of thermodynamic properties.

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)

Apparatus



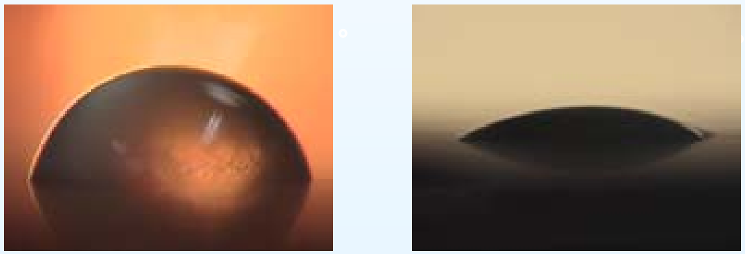
Experimental apparatus for contact angle measurement

(30×30×3 mm)

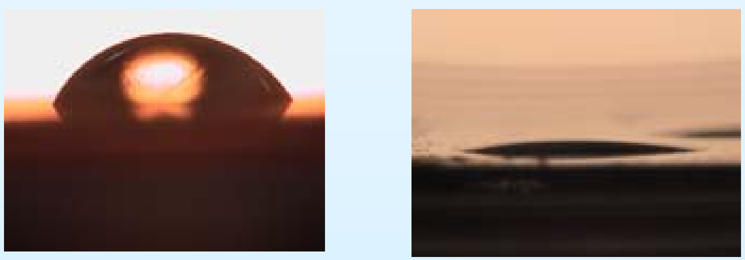
Test pieces
Titanium, Stainless, Zircaloy, and Copper
(Oxidized by plasma jet)

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

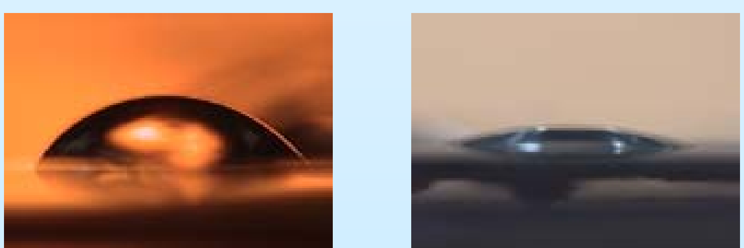
Before irradiation ⇨ After irradiation



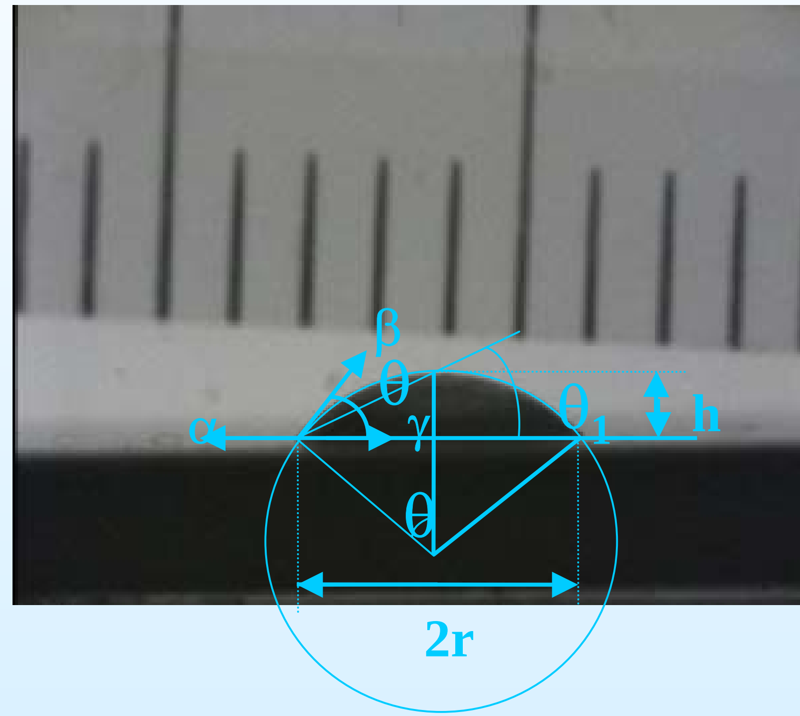
Titanium



Zircaloy No. 4

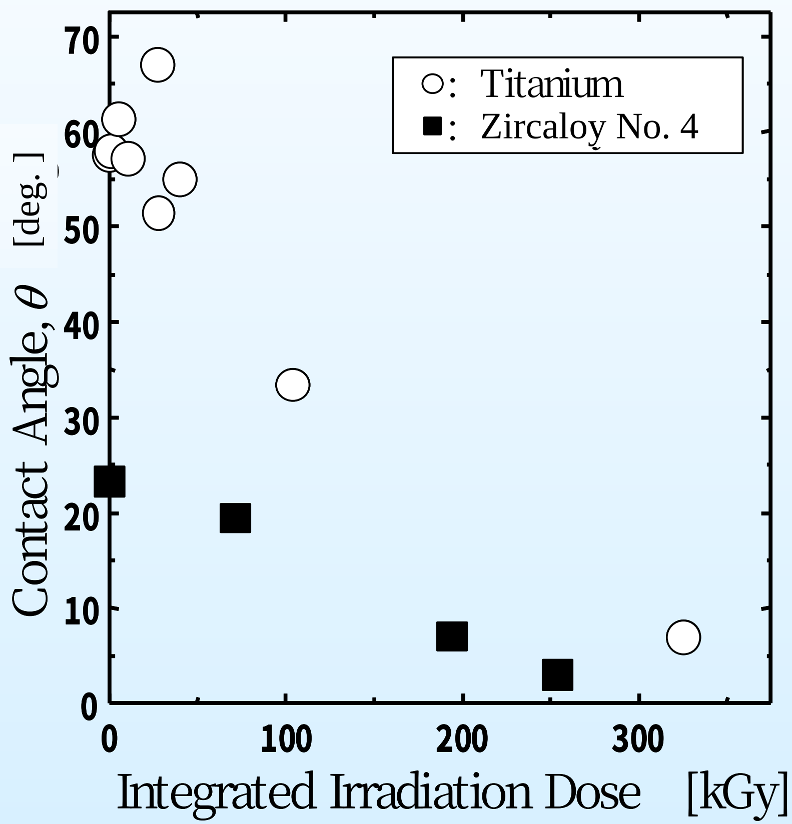


Aluminum

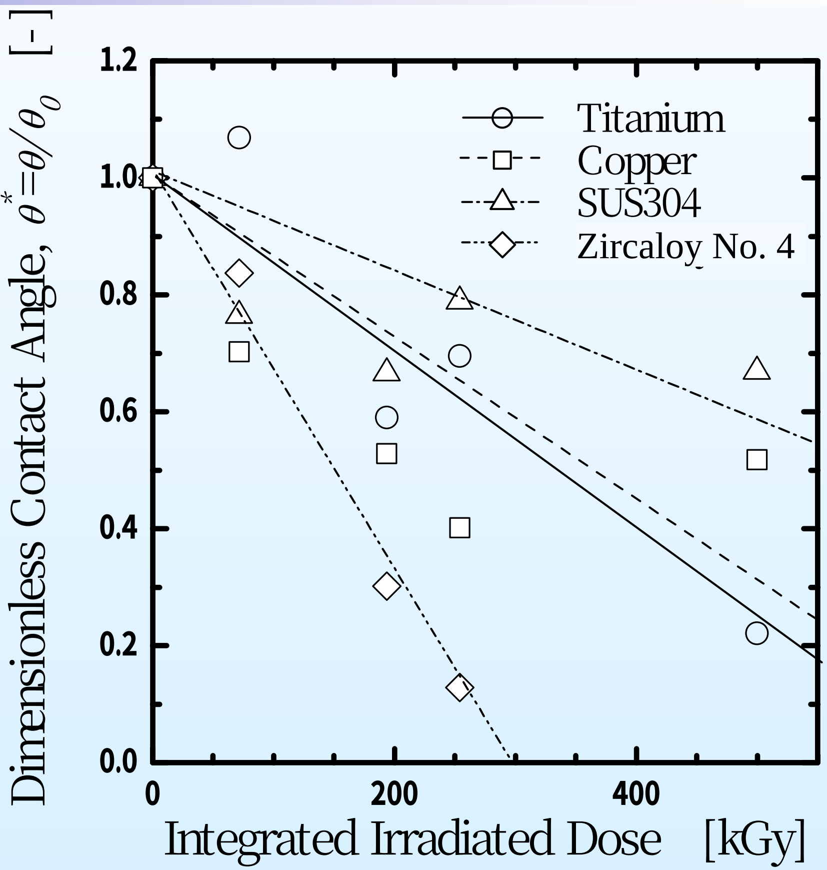


Hydrophilic condition changes resulting from γ-ray irradiation (after 250kGy integrated irradiation)

Changes of contact angle by γ-ray irradiation



Change of contact angle by γ-ray irradiation

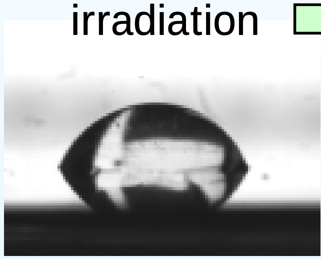


Dimensionless contact angle of test pieces

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

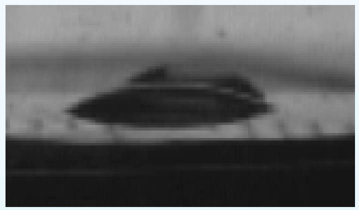
Cyclic change of contact angle

Before irradiation



Contact Angle : 71.4 °

After irradiation (160 kGy)



Contact Angle 23.7 °

8hr non irradiation

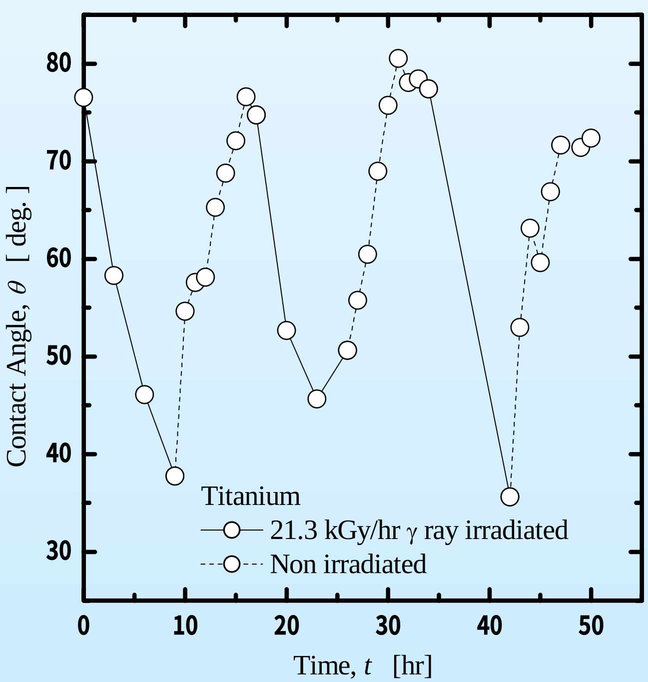
Contact Angle : 90.6 °

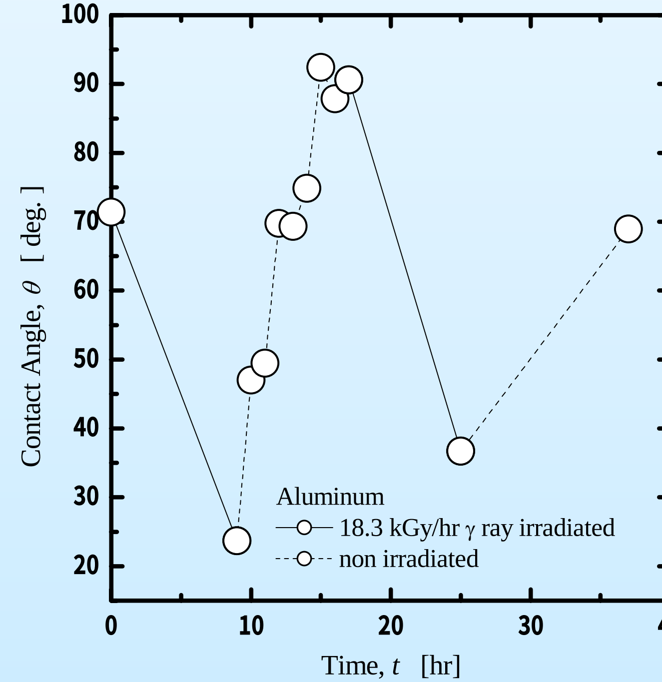
After irradiation (140 kGy)

Contact Angle : 36.7 °

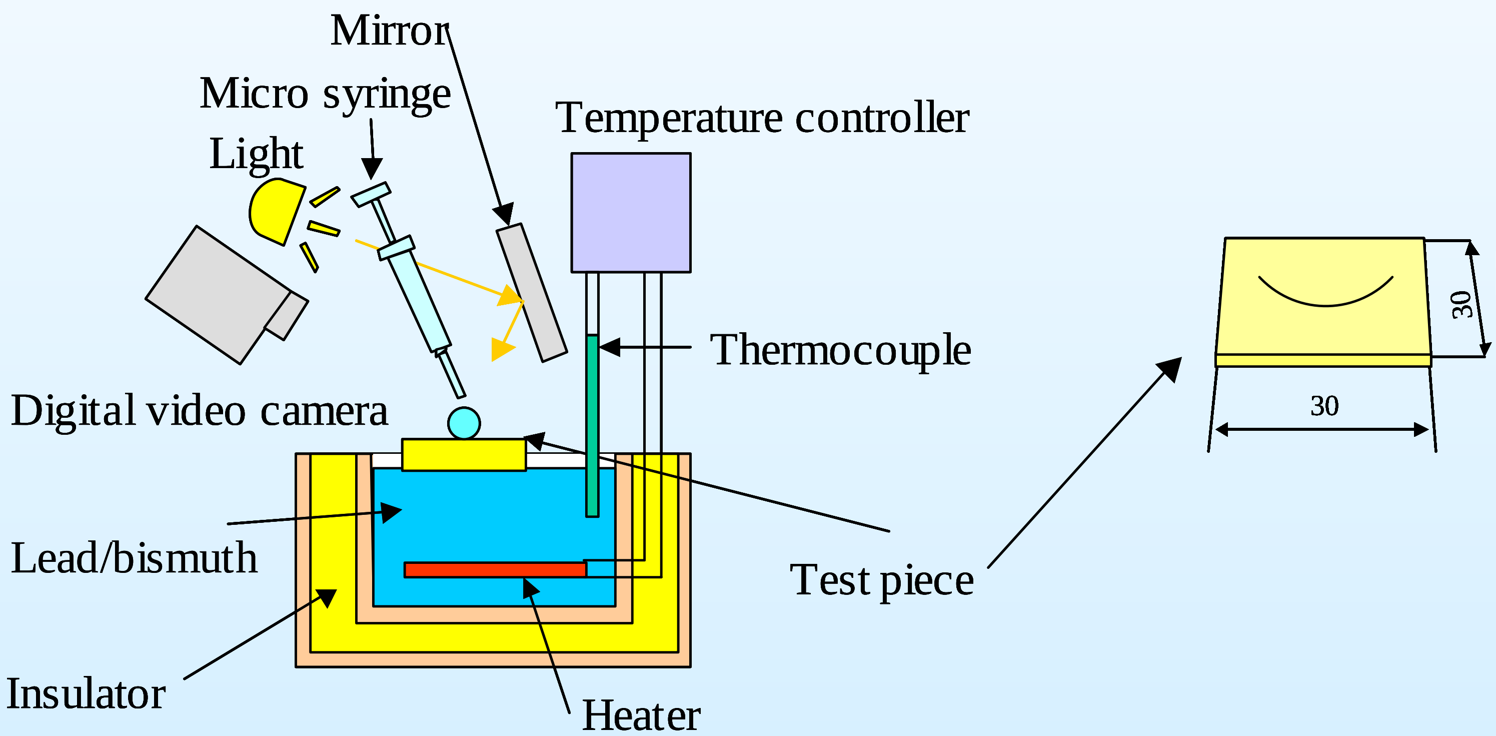
12hr non irradiation

Contact Angle : 68.9 °





2) Heat Transfer experiment
a. Experiment to Investigate Leidenfrost Temperature (2002)



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.

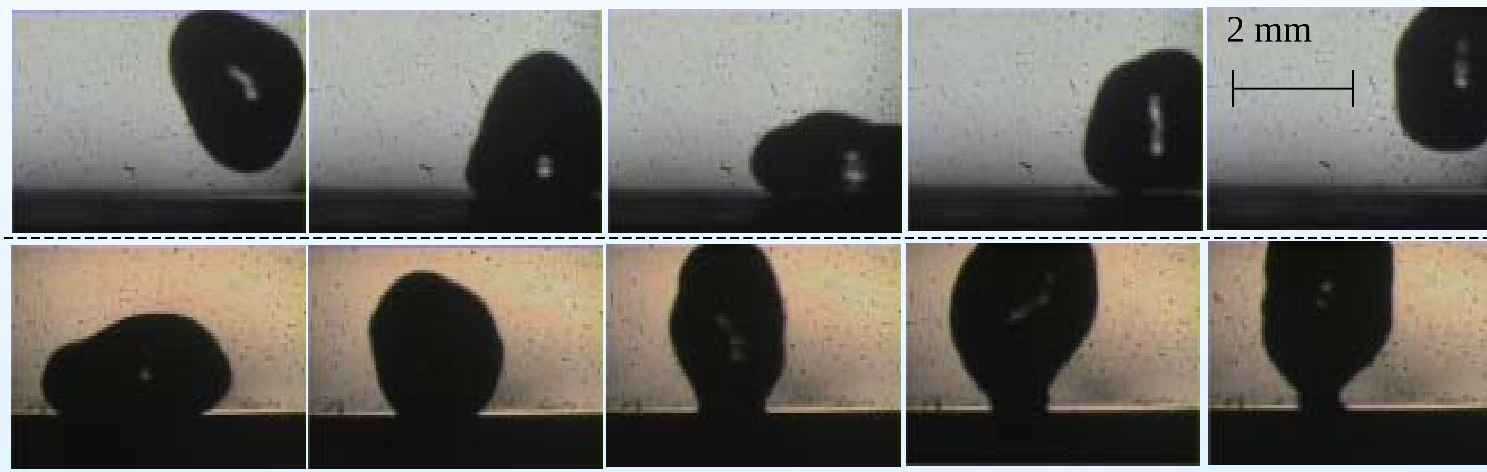
Takata et al., (1999)

Droplet Behavior near Leidenfrost Condition

59 ms | 63 ms | 67 ms | 71 ms | 75 ms

Before irradiation Over wetting limit temperature

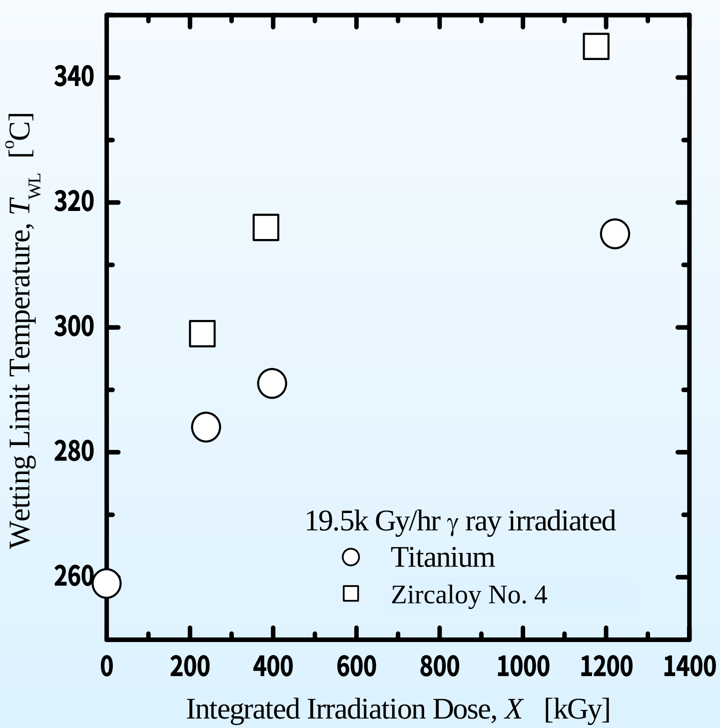
After 260kGy irradiation Under wetting limit temperature



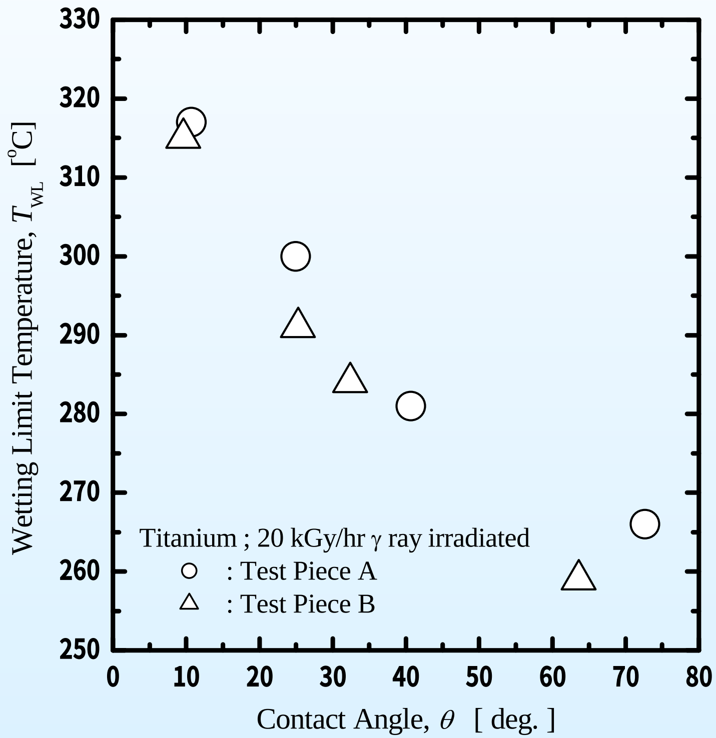
Behavior of droplet on heated TiO_2 plate
(droplet diameter: 2.7 mm, temperature of Pb-Bi pot: 320 ℃)

T.U.M.S.A.T. Power and Energy Lab.

Wetting Limit Temperature



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

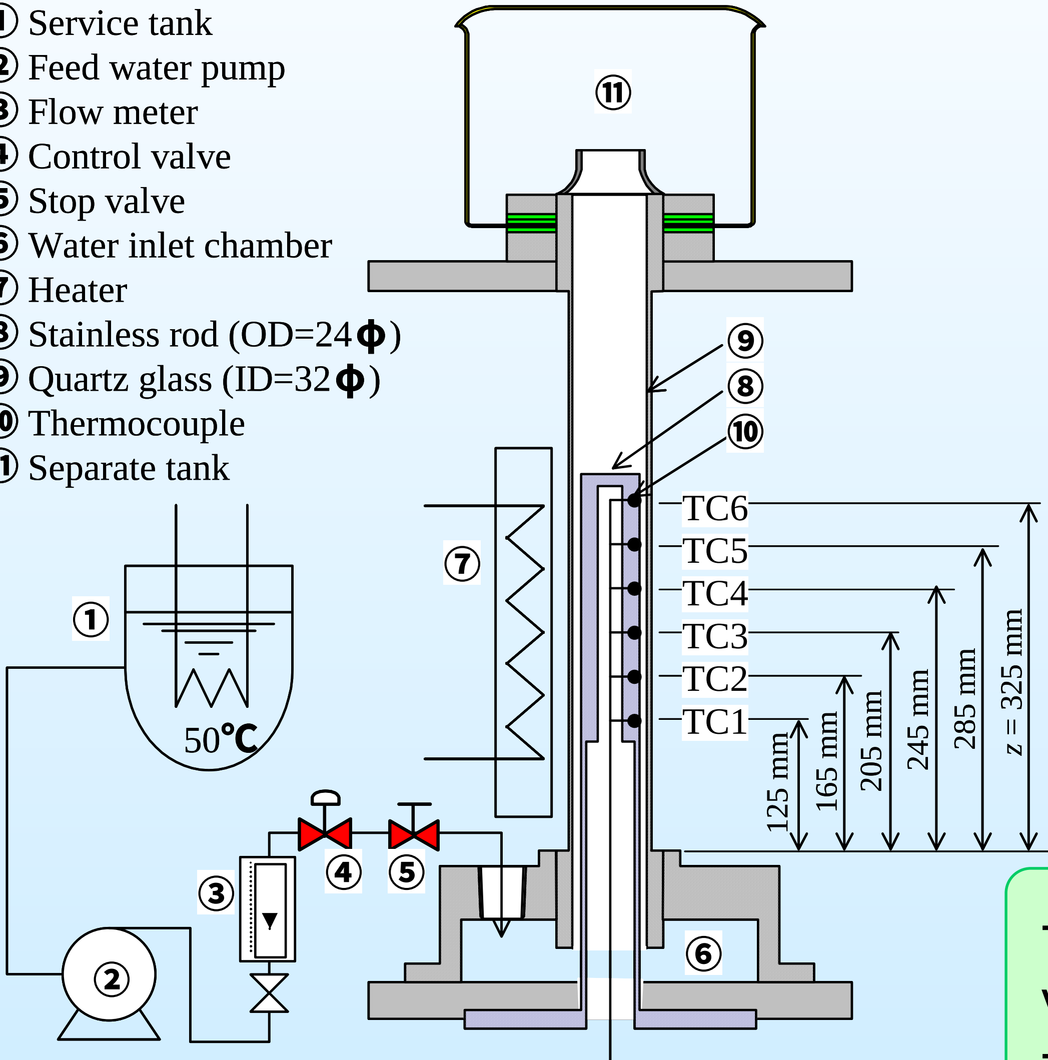


The wetting limit temperature against contact angle

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

b. Quenching Experiment

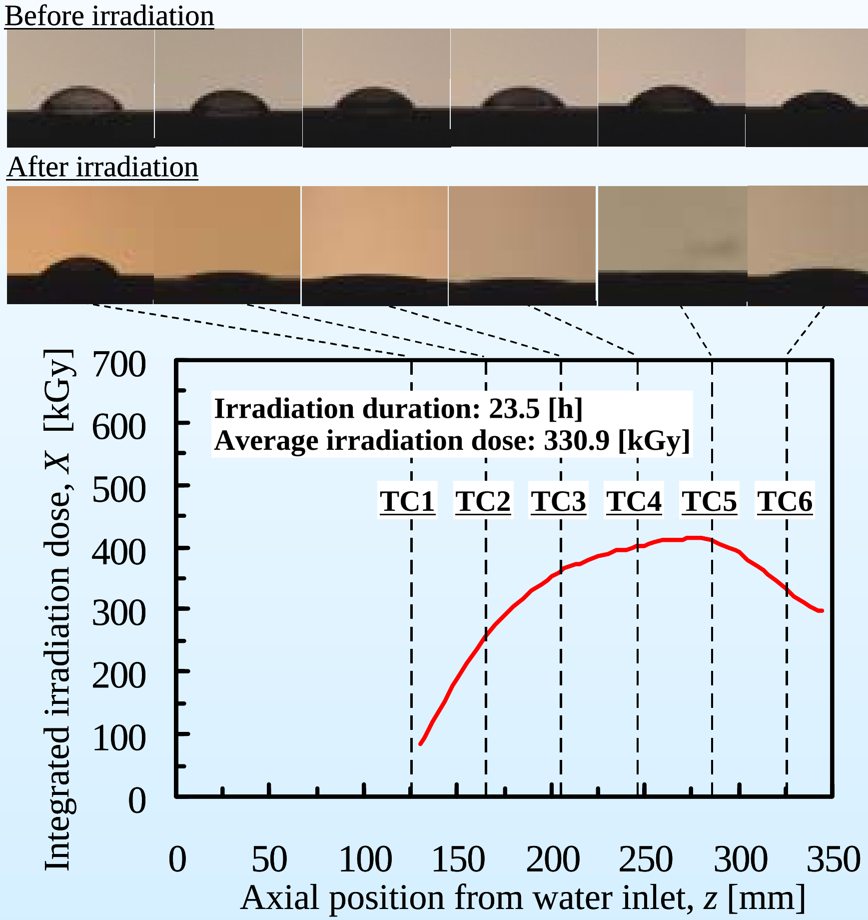
① Service tank
② Feed water pump
③ Flow meter
④ Control valve
⑤ Stop valve
⑥ Water inlet chamber
⑦ Heater
⑧ Stainless rod (OD=24ϕ)
⑨ Quartz glass (ID=32ϕ)
⑩ Thermocouple
⑪ Separate tank



Apparatus for reflooding experiment

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

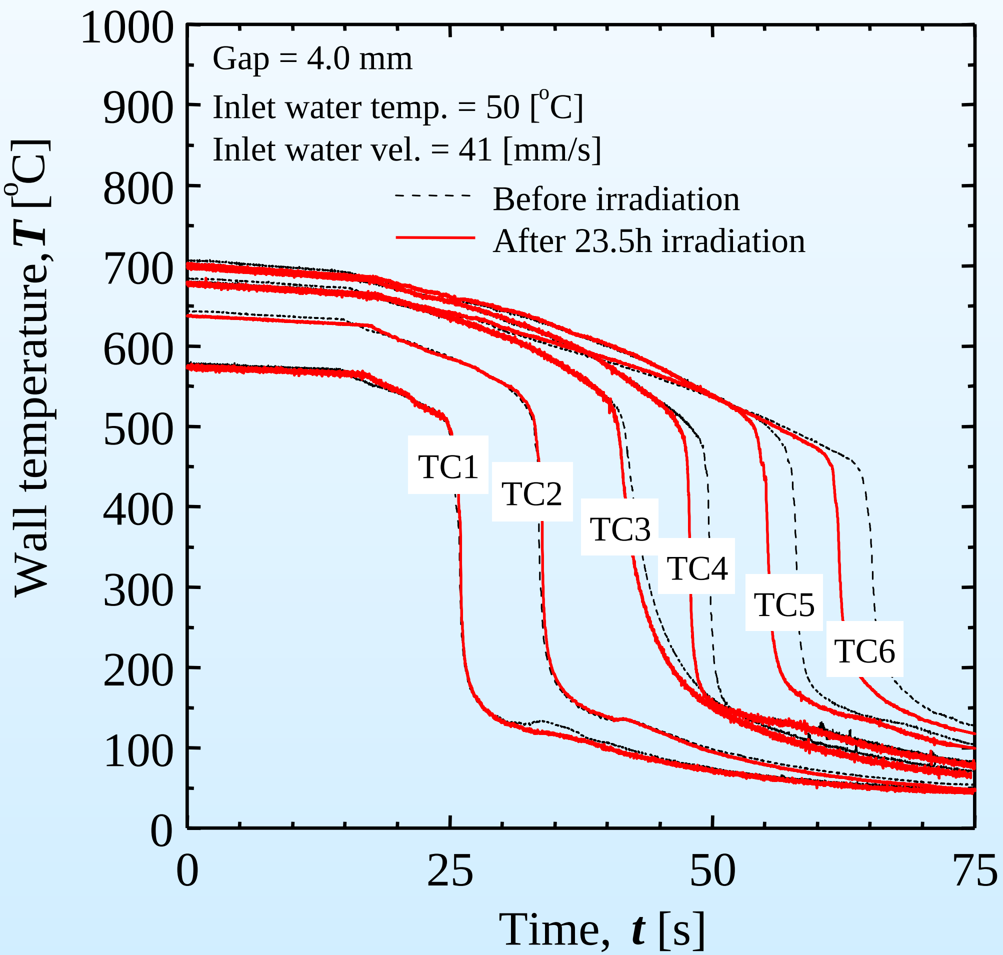
Rod Surface Wettability Change due to γ-ray Irradiation



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.

Typical Result of the Surface Temperature Record



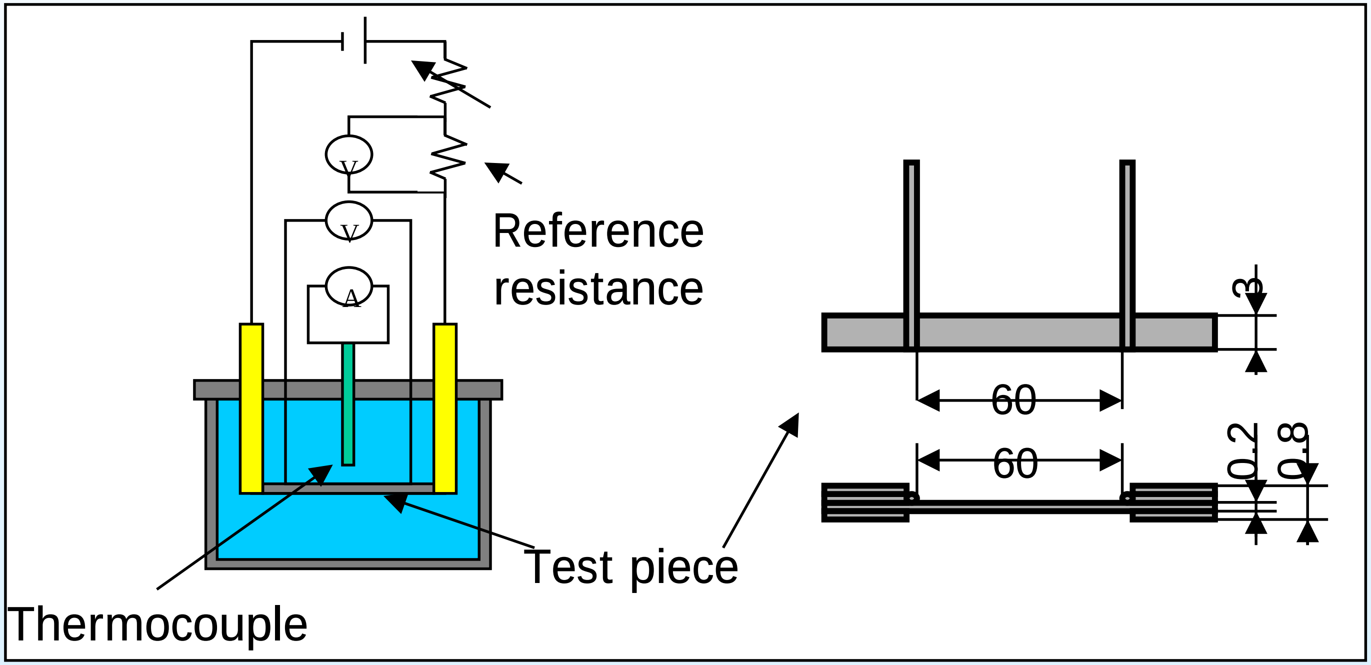
- No discrepancy exists in temperature records between before and after irradiation at TC1 and TC2 where no changed wettability was observed.

  ⇒ 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.

c. CHF Measurement



CHF experiment: the pool boiling condition

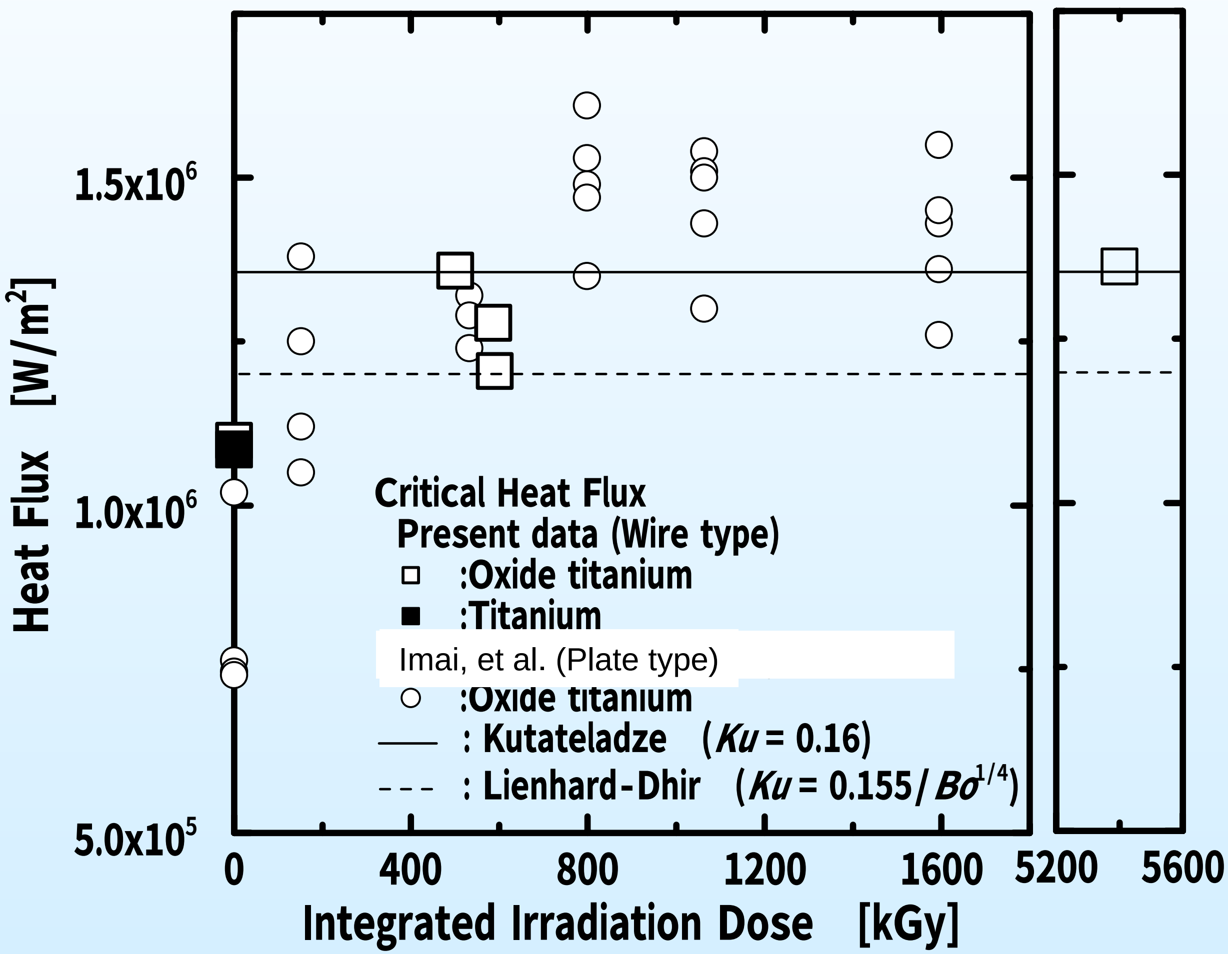
The pressure: atmospheric pressure
(resulting in the boiling point to be 100 degree C)

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.

Radiation Induced Boiling Enhancement

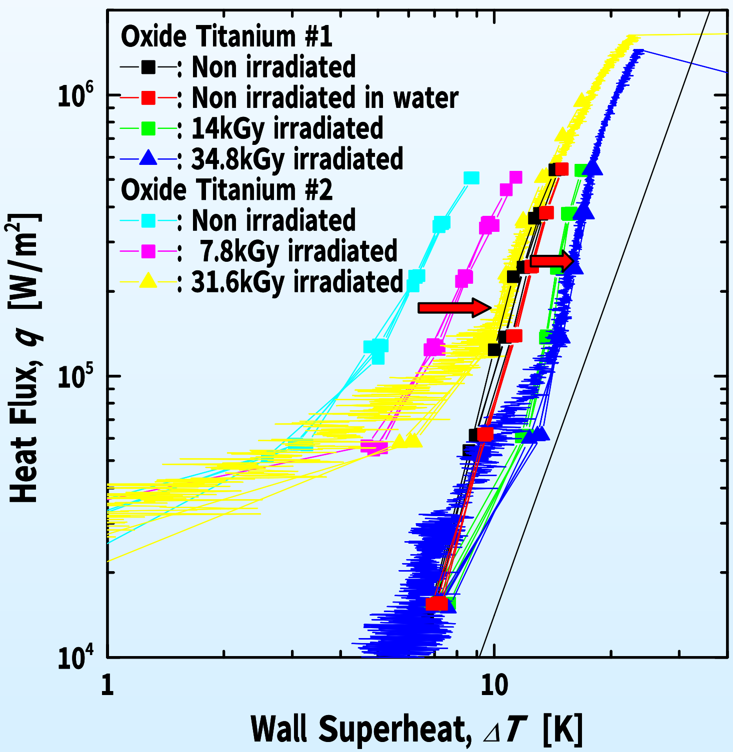


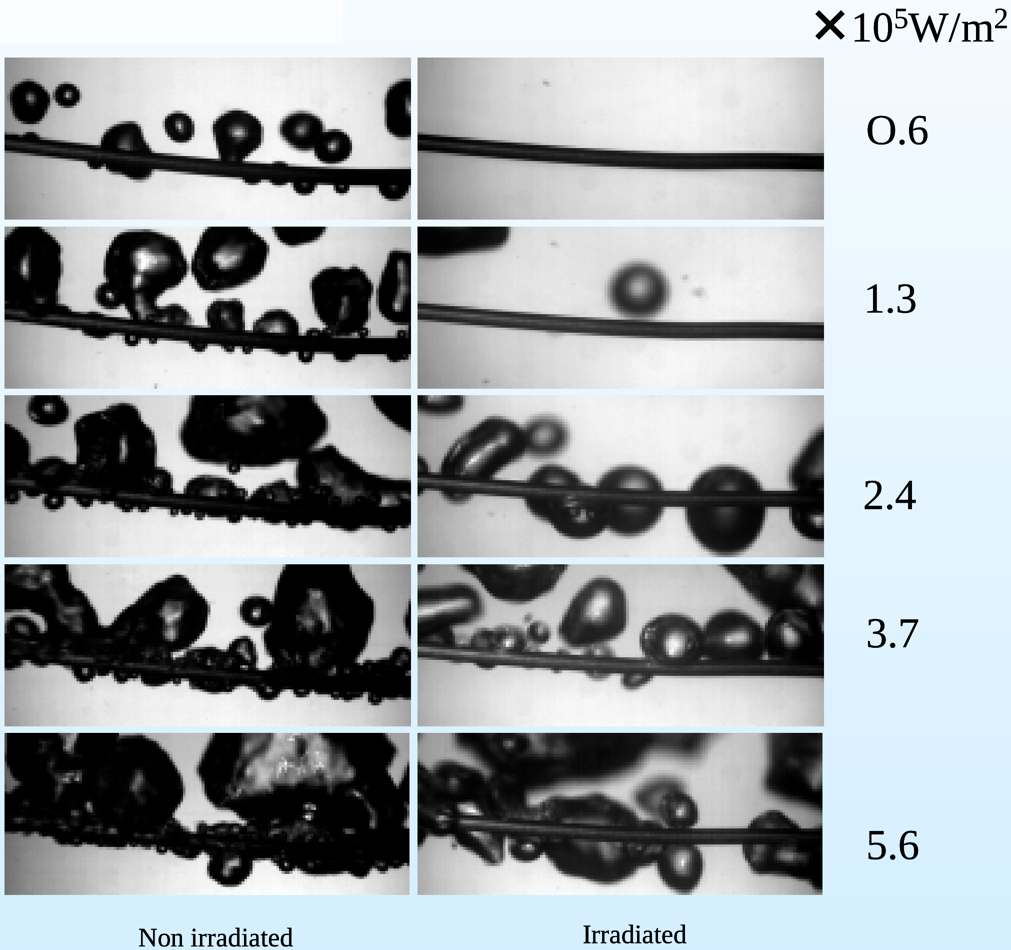
Improvement of CHF by γ-ray irradiation (TiO_2)

CHF:

improved as much as 100 % after 800-kGy ^{60}Co gamma-ray irradiation.

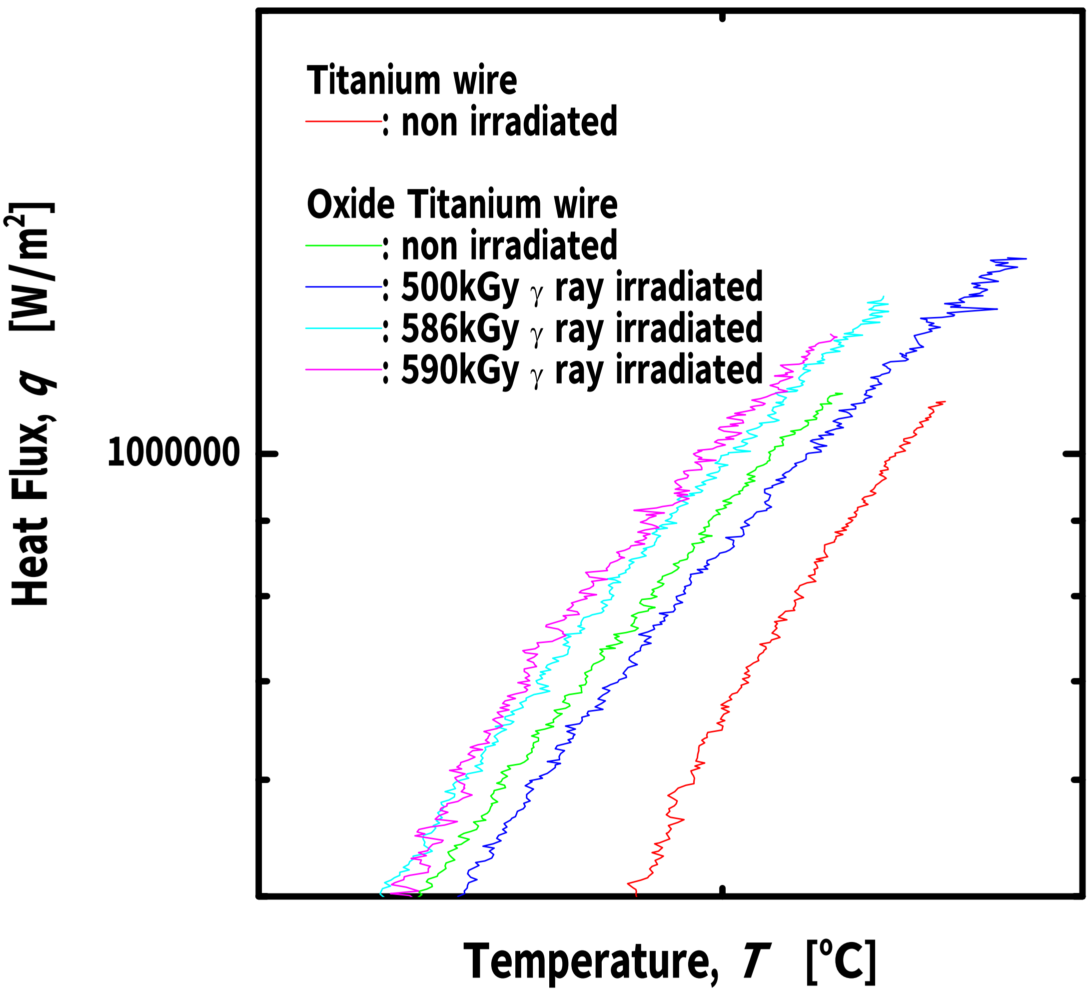
Nucleate boiling under irradiation





Boiling curve shifts to high-wall-temperature with increasing gamma-ray dose.

Boiling Curve of Off-site experiment
The gamma ray irradiation effects



Boiling curve of oxide titanium wire against temperature

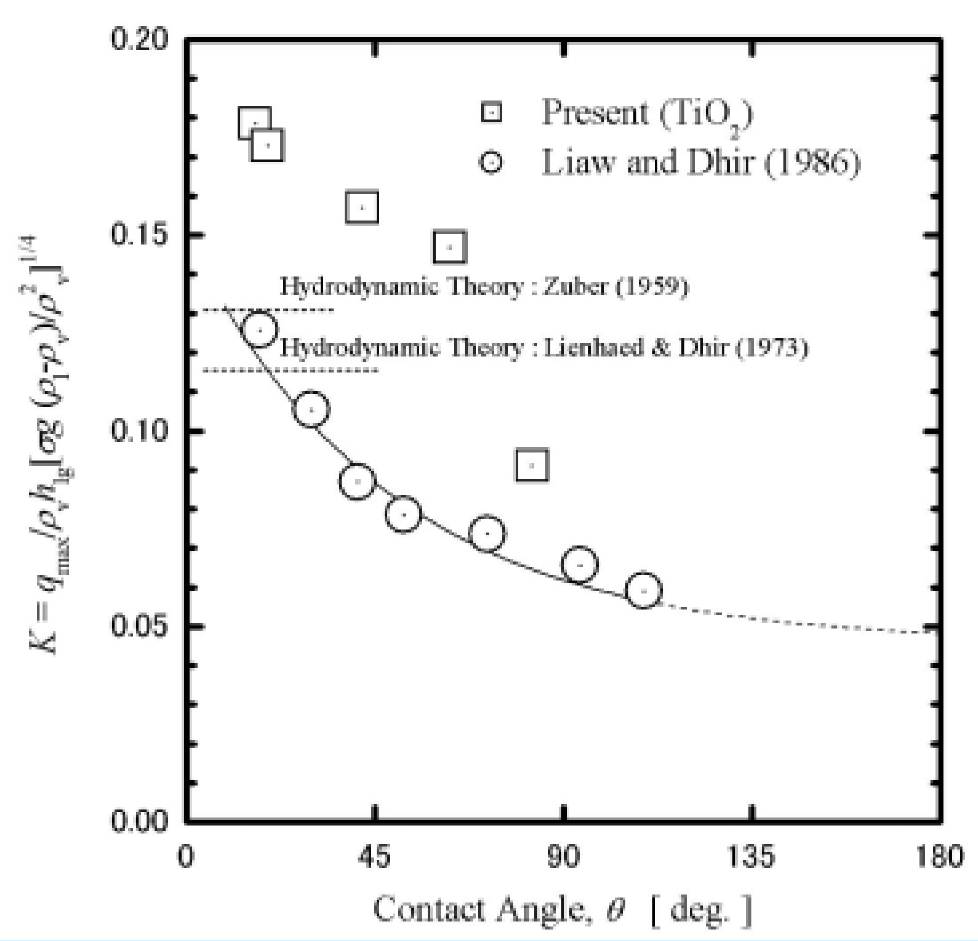
The CHF of the oxidized titanium wire is higher than that of non-oxidized one.

The boiling curves also move to lower temperature side. The oxidized layer does not play the thermal resistance.

The CHF of irradiated wire is higher than that of non-irradiated wire.

The irradiated titanium wire can reach higher temperature at the boiling transition.

Relationship between wettability and CHF



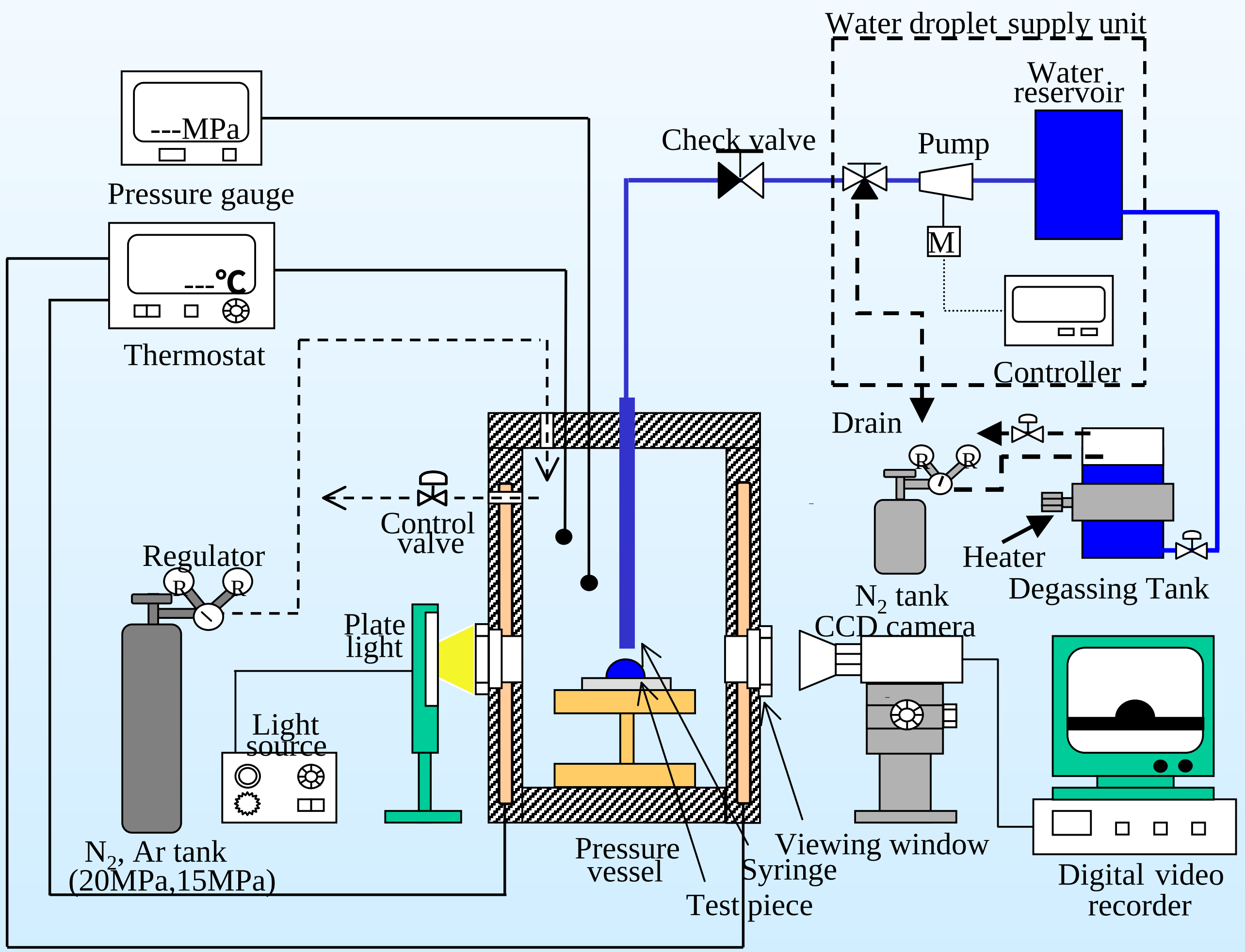
Contact angle and CHF

K is defined by the following equation from the interfacial stability analysis conducted by Zuber.

$$K = q_{\max} \big/ \rho_v h_{fg} \left[\sigma g \left(\rho_l - \rho_v \right) \big/ \rho_v^2 \right]^{1/4}$$

CHF in the present experiment increases with surface wettability in the same manner as shown by Liaw and Dhir's results.

Surface wettability in high temperature and high pressure condition



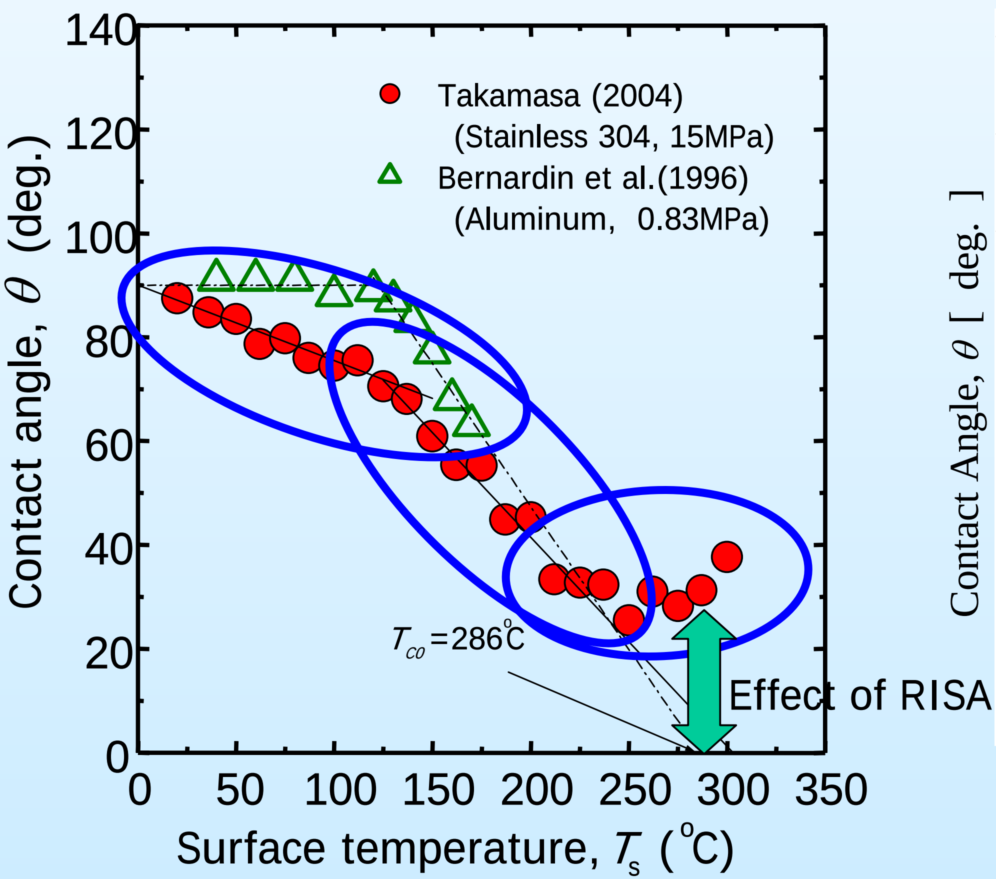
(ID=50 mm, h=150 mm, t =40 mm)

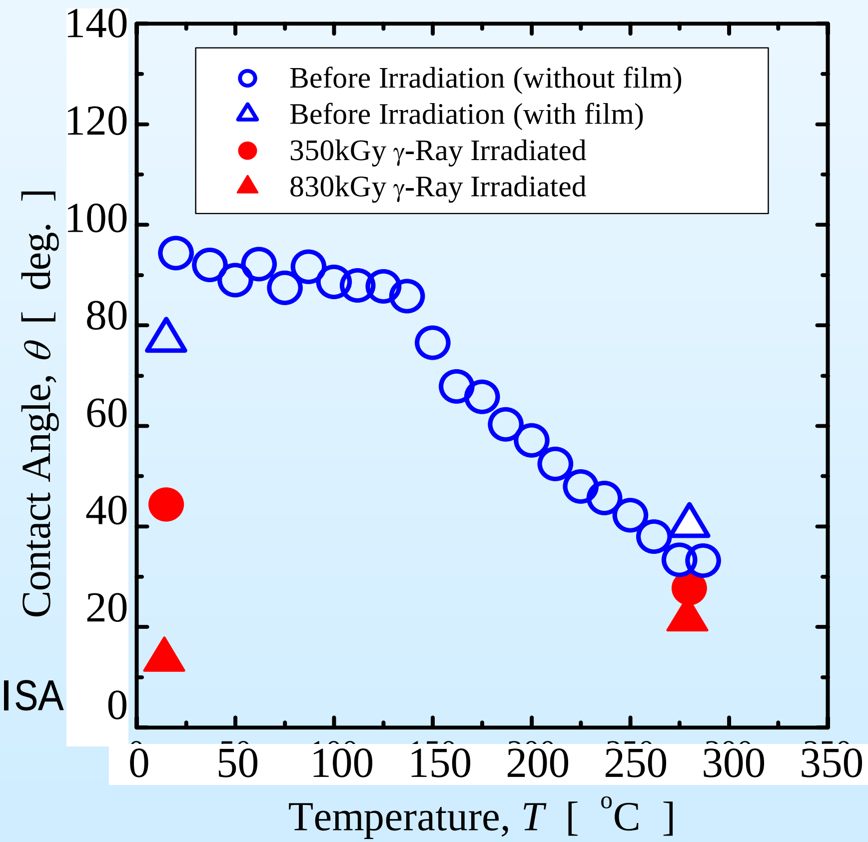
Contact angles of water droplets using a pressure vessel at temperatures from 20 to 300°C and at a constant pressure of 15 MPa

Takamasa. et al. (2004-2006) corroborated with Prof. K. Vierow, Mr. A. Pollman

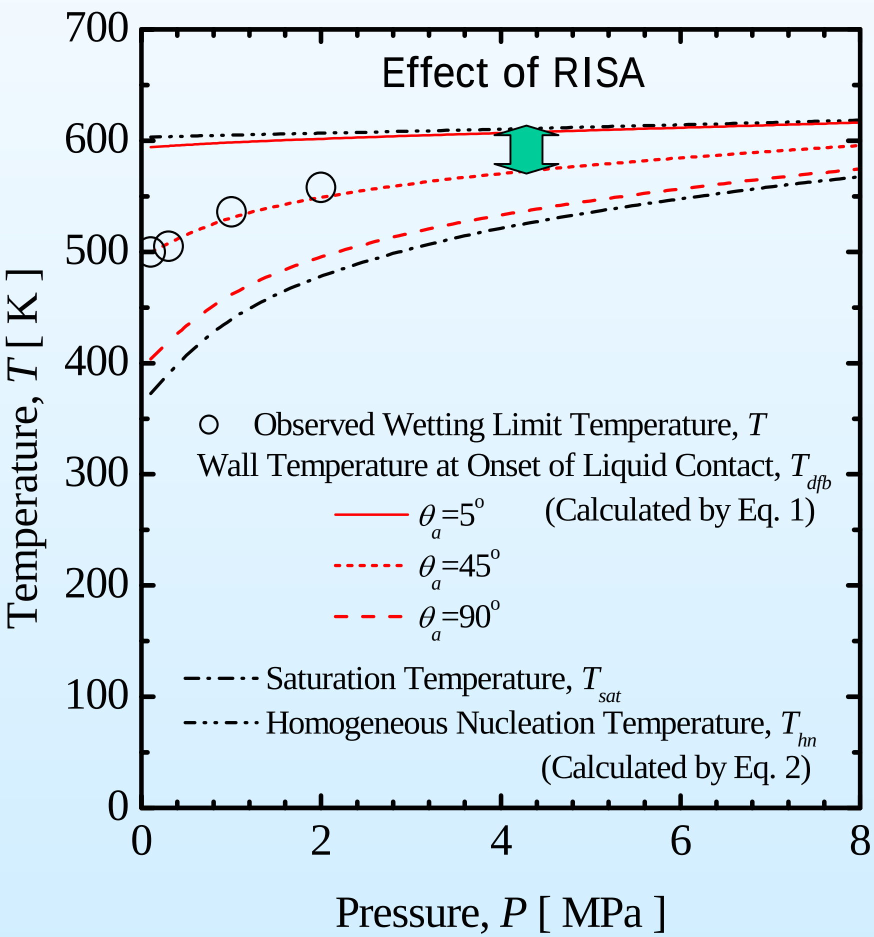
Contact angle in high temperature condition

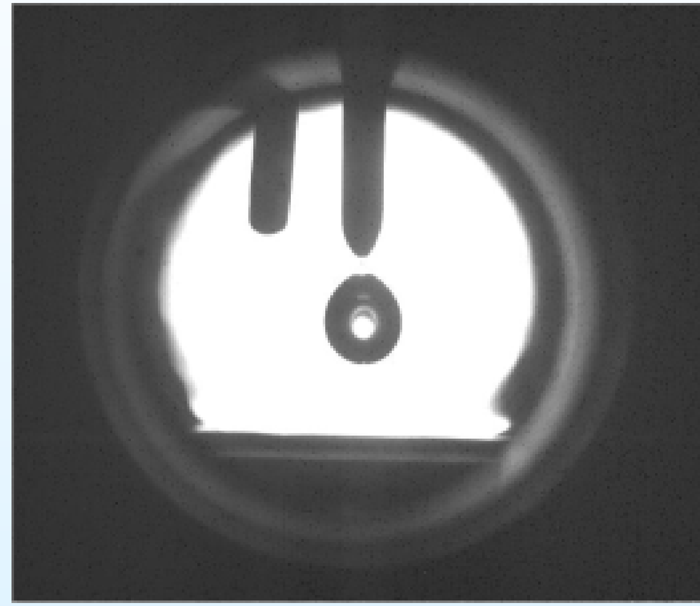
At temperatures above 250°C, contact angles remained constant independent of temperature, and contrary to the existing theoretical results, no highly hydrophilic condition or null contact angle condition was achieved.





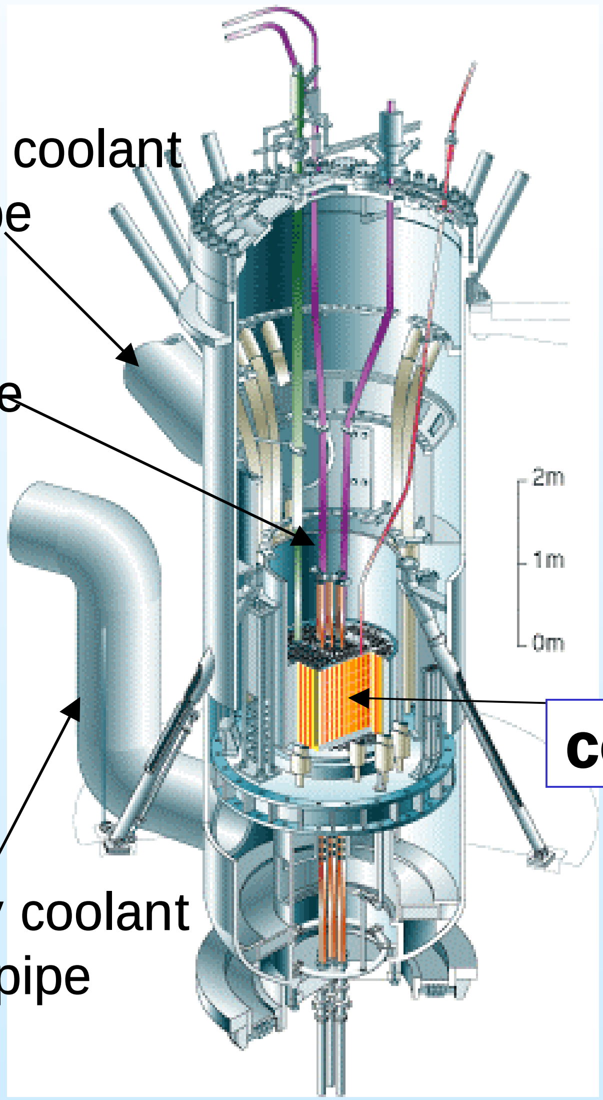
Leidenfrost condition in high temperature condition





By utilizing a pressure vessel, Leidenfrost conditions of water droplets were measured up to 2.1 MPa of ambient pressure. The results revealed that the wetting limit temperature increased with the ambient pressure. The theoretical equations regarding to the wall temperature at onset of liquid contact in pool boiling predicted well the present results.

d. JMTR Experiments for CHF/RISA

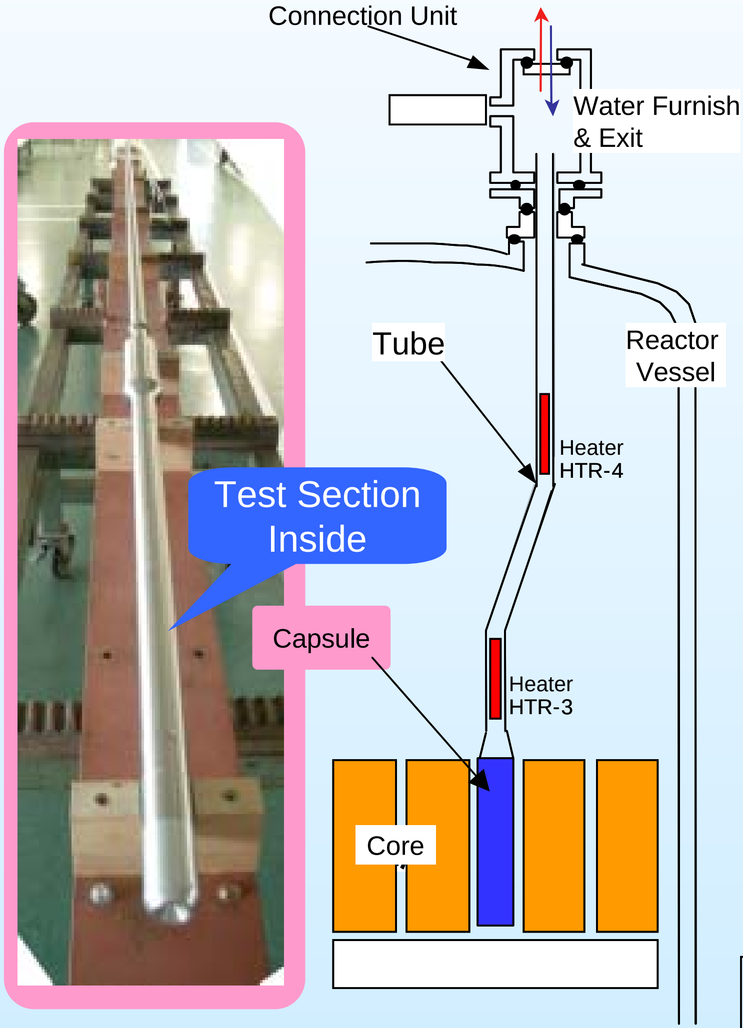


Japan Materials Testing Reactor (JMTR)
JAEA

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.; $4.0x10^{18}/m^2/s$
RISA exp.; $1.0x10^{17}/m^2/s$

- 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.

Experimental Conditions



Capsule and Irradiation Setups

Forced-convection boiling heat transfer (upward flow)

Test Channel : tube (i.d. 2 mm, heated length 100 mm, in SUS cylinder)

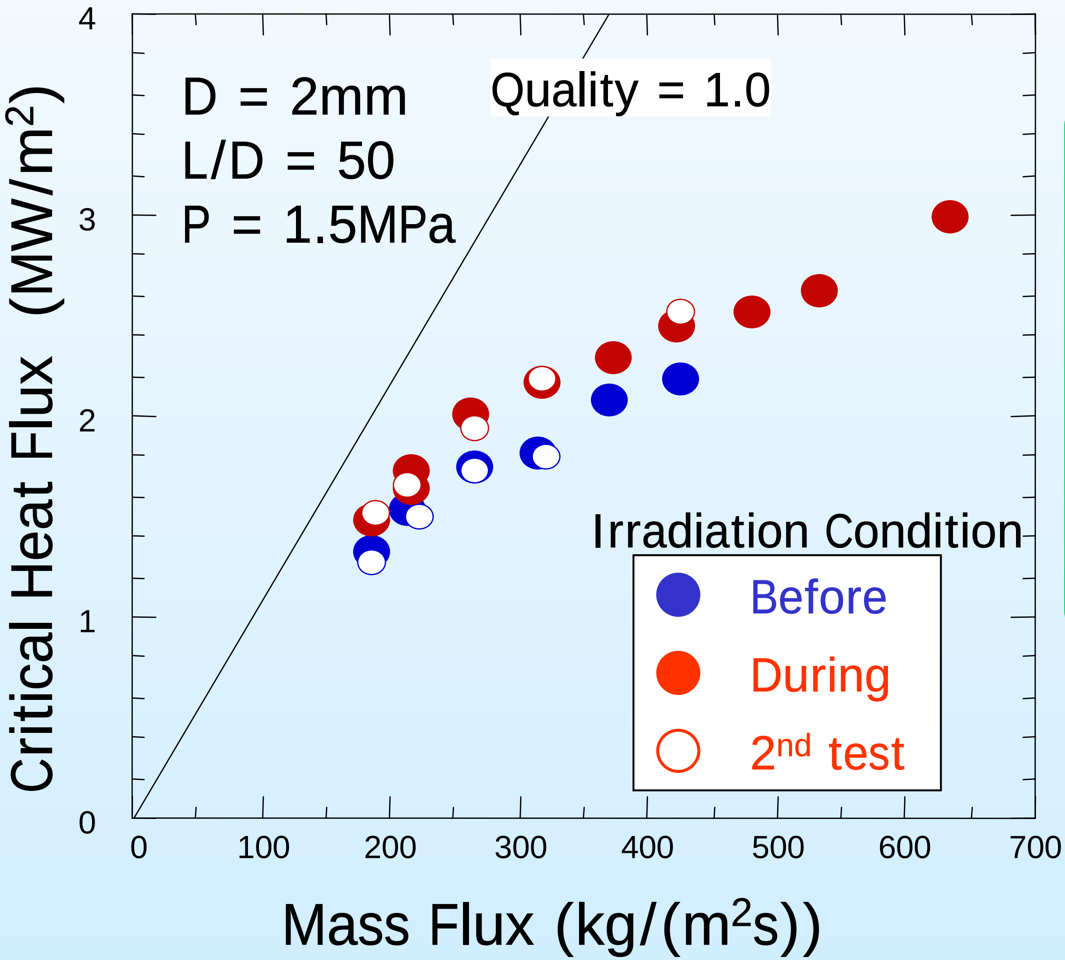
Pressure : typically 1.5MPa (BWR -- 7 MPa)

Flow Rate : 180 to 630 kg/(m^2s) (BWR -- 1,500 kg/(m^2s))

Inlet Subcooling : 35 to 120 K

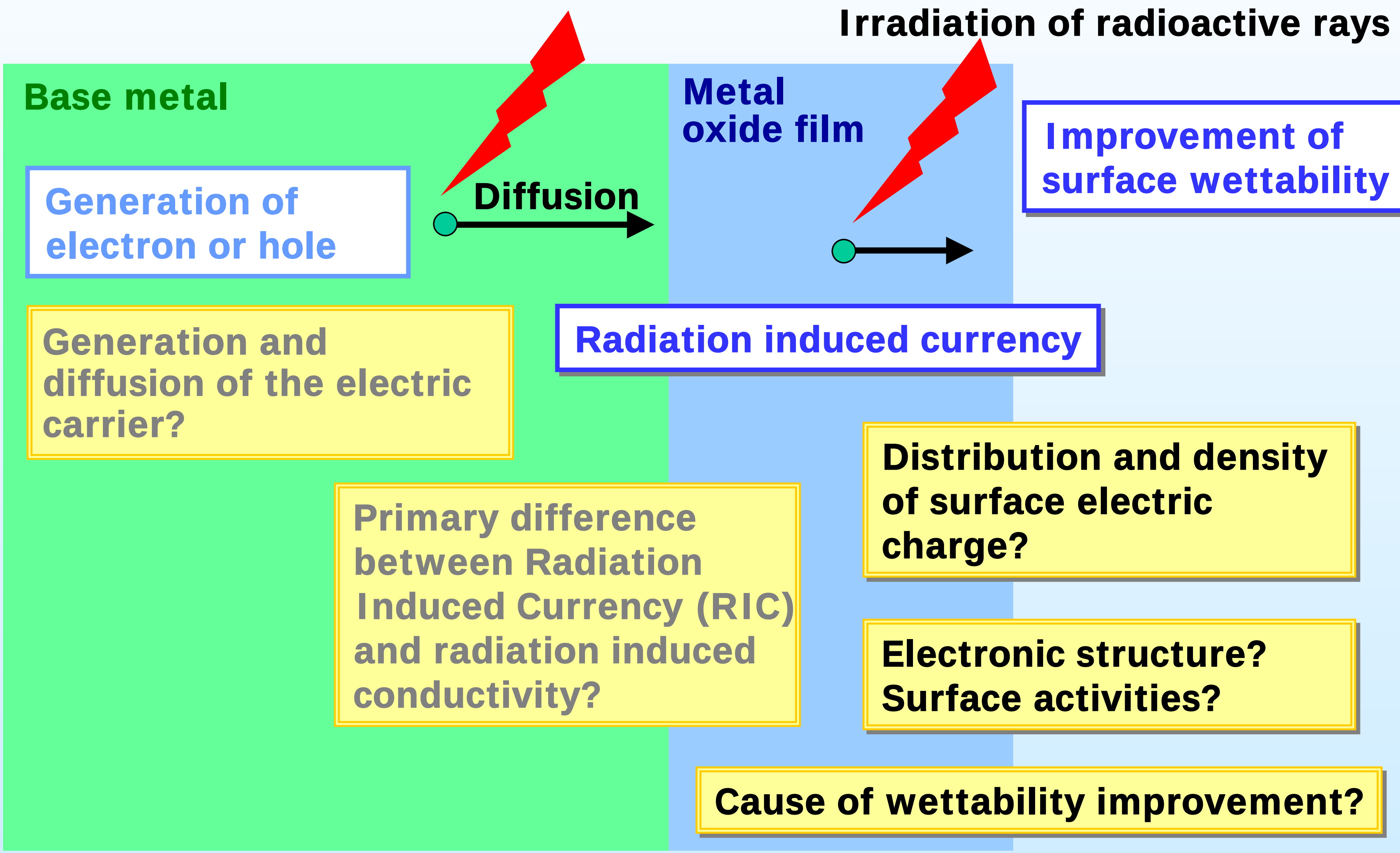
- Gamma ray dose rate -- 540kGy/h
- Integrated gamma irradiation -- 100MGy

CHF results



- ✓CHF increased by 13% due to irradiation in JMTR
- ✓CHF appeared at high quality conditions
- ✓CHF mechanism would be dryout-type similar to that in BWR core

RISA mechanism



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

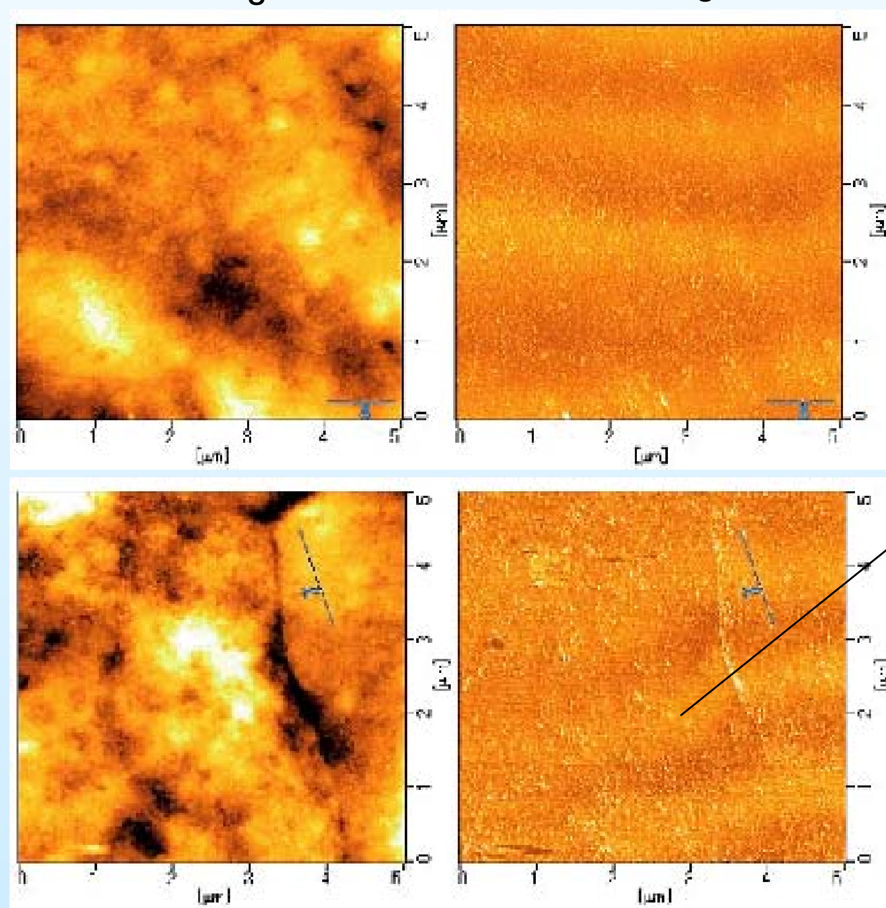
●White spots corresponds to high friction force are observed in the sample after gamma ray irradiation.

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

↓

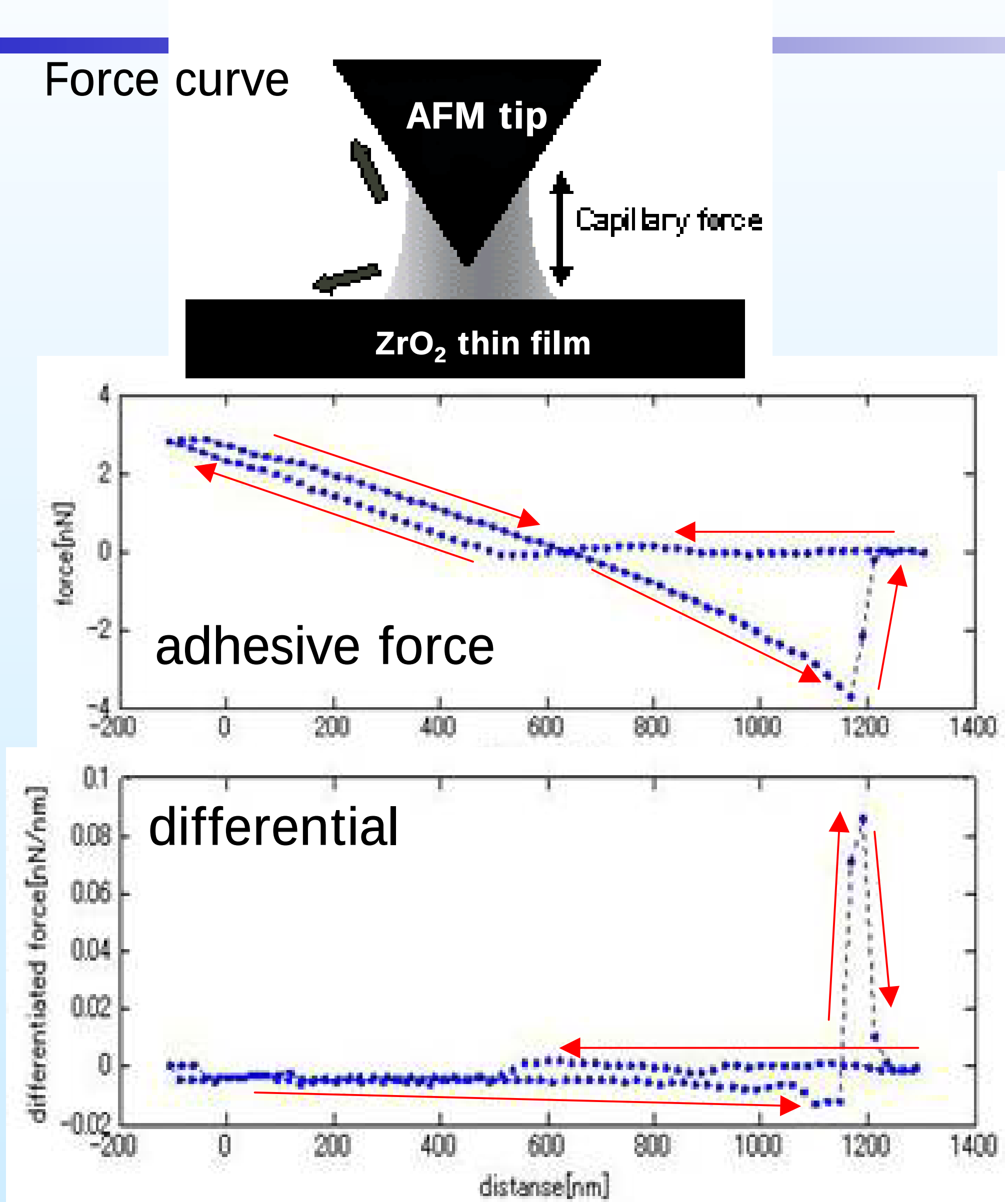
Substrate; Zircaloy-4
Oxide layer; by autoclave
Irradiation; 340 kGy
Measurement; at r.t. in air
Contact angle; 12 deg.

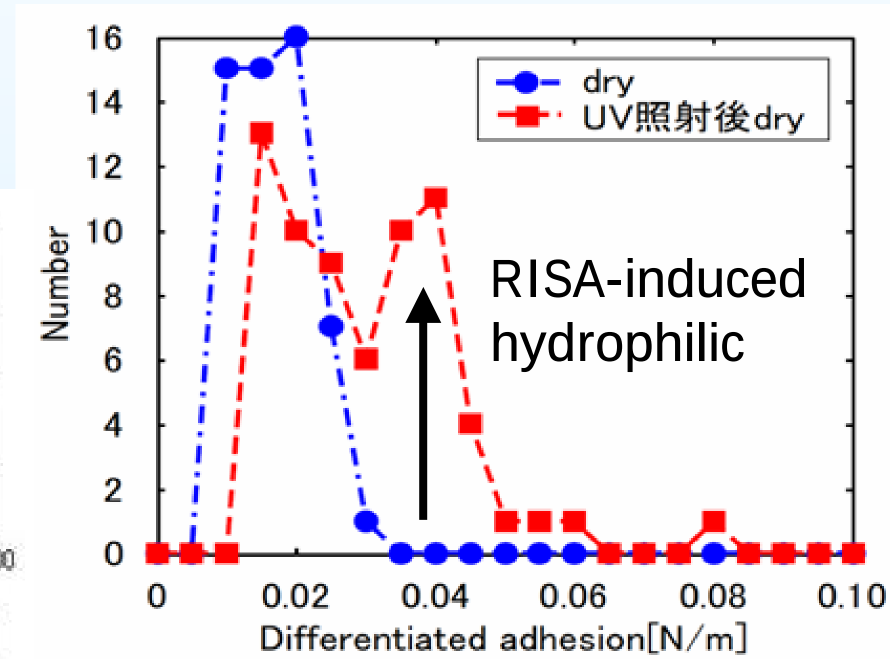
AFM image FFM image



White spots are observed

Wettability of ZrO_2 surface at AFM scale





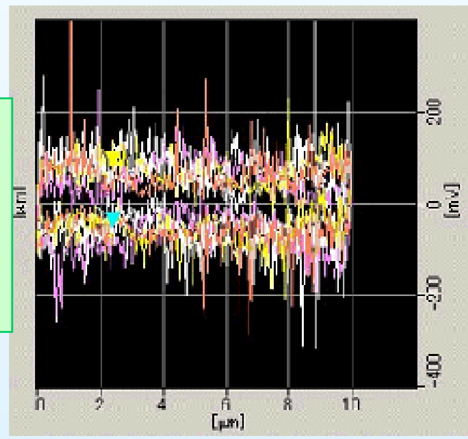
matrix　water?

hydrophilic domains?

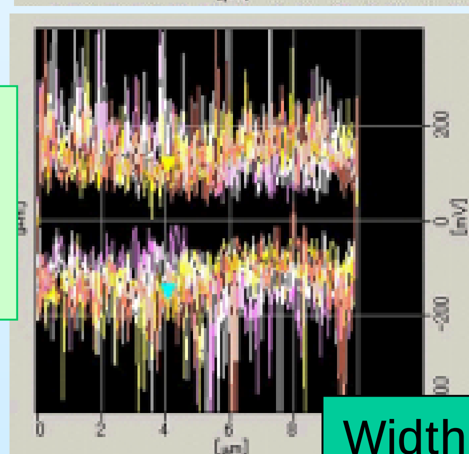
Analysis by AFM

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

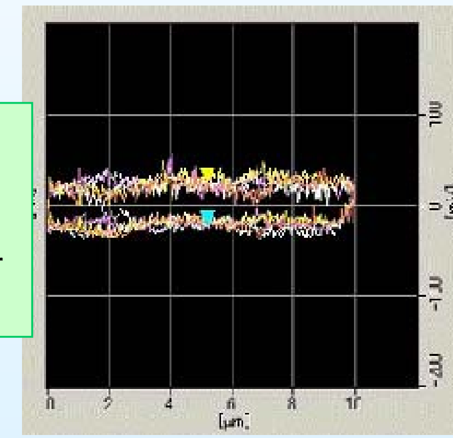
Substrate; Zircaloy-4
Oxide layer; by autoclave
Irradiation; No irradiation
Measurement; at r.t. in air
Contact angle; 77 deg.



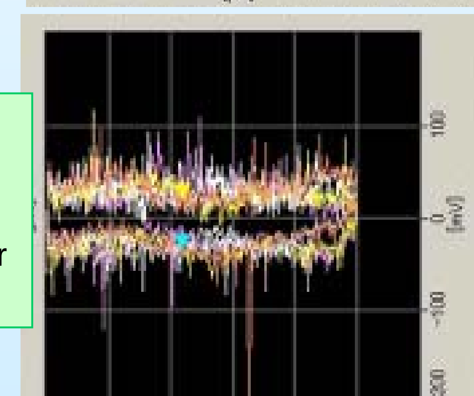
Substrate; Zircaloy-4
Oxide layer; by autoclave
Irradiation; 340 kGy
Measurement; at r.t. in air
Contact angle; 12 deg.



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.

Surface Wettability

- Hydrophilicity rate depends heavily on the integrated irradiation dose, and effect of irradiation intensity is small.
- It is assumed that the existence of water content both in air and on the surface of the oxide layer plays an important role for the hydrophilicity phenomenon.
- The contact angle for water increases gradually after the irradiation when sample is preserved in air, whereas the contact angle of the same sample preserved in water stays unchanged, and high hydrophilicity is maintained at least for 2,040 hours.
- It is assumed that the appearance of hydrophilicity by the dosage of gamma ray irradiation is related to the increase of the surface friction force.

Expected Applications of RISA

Improvement of heat transfer properties of nuclear reactors:

Nuclear reactors are designed and assessed by thermohydraulic data obtained from electric heater experiments. Nobody has paid attention the relationship between radiation in nuclear reactor and thermohydraulic incidents, boiling, critical heat flux, Leidenfrost temperature, rewetting and so on.

・High heat flux can be achieved by application of the oxide material (Zircaloy, Stainless) to fuel clad in a reactor, which makes possible reductions in both weight and size of the reactor vessel.

・In the event of an accident, rapid reactor cooling can be achieved through use of the material for the internal structure of the reactor.

Expected Applications of RISA

Use of the electrical and chemical reactions caused by RISA is expected to offer many applications in nuclear reactor.

The RISA current induced in Oxide metal film: Radiation Detection, Corrosion Protection, Hydrogen Gas Production, Self Cleaning, etc.

Hopefully research on this new technology may be conducted more widely in the near future, shedding light on the radiation effect in nuclear reactor.

Thank you.