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URAM-2 CRYOGENIC IRRADIATION FACILITY

1. Introduction

Nowadays there are a lot of different high-intensity neutron sources dedicated to study structural and dynamical characteristics of materials. The most promising investigations are given to "cold" neutrons, generated by cold moderators. The best materials for this kind of moderator are hydrogenous materials such as methane, water ice, etc. But there is a problem of using these materials because under radiation free radicals are produced in many chemical compounds and the candidates for the moderator material in question (methane hydrates, water ice, solid methane) are of no exception. At low temperatures radical accumulation gives rise to stored chemical energy in an irradiated sample, as recombination of radicals is an exothermic reaction. Usually, the process of fast recombination might be stimulated by heating of the sample but under specific condition the stored energy may be released spontaneously, with no perturbation in cooling condition. Such phenomenon is usually called "a burp" [2-12]. Naturally, the resulting sudden leap in temperature not only has an ill effect on the yield of cold neutrons from a moderator but may also lead to serious mechanical loads on the container.

For investigation of irradiation effects in materials-candidates for cold moderator material on channel N3 [1] at the IBR-2 reactor, JINR [13], an irradiation facility, called "URAM-2" is designed and constructed.

2. Description of the facility

The irradiation rig URAM-2 was designed to work with different types of materials such as methane, water ice, tetrohydrofuran, etc., in a spread range of low temperature of a sample. Owing to the cooling characteristics, it is possible to work on temperatures from 10 K up to the room temperature, but for the most part of experiments the range is $10 \div 60$ K.

The facility consists of four main parts (Fig. 1):

1. charging device;
2. irradiation rig head;

3. liquid helium cooling system;
4. measuring system.

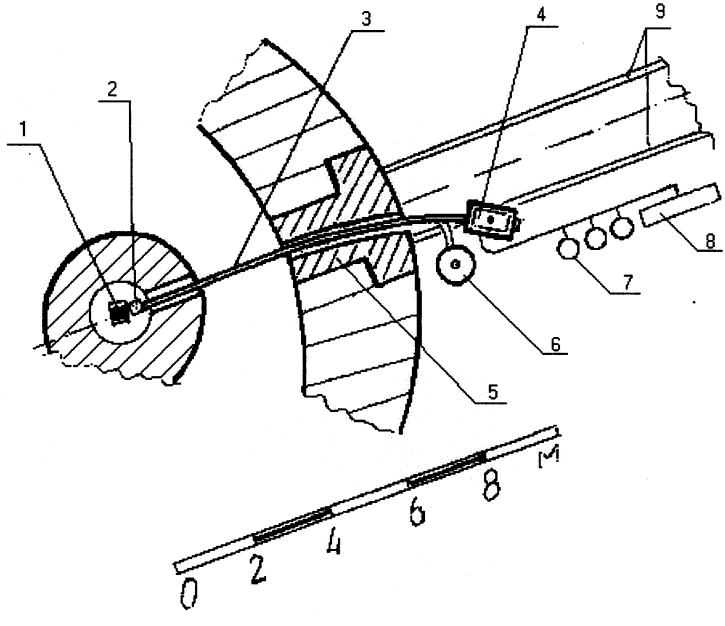

Fig. 1 General schematic view of irradiation facility "URAM-2":
(1) the IBR-2 reactor; (2) head part of the facility; (3) transport passage, helium pipelines and electric communications; (4) charging device; (5) shielding plug on rails; (6) liquid helium Dewar vessel; (7) gas containers; (8) PC with communications; (9) rails

The rig has been installed on a long movable platform which can brought the head part with communications to the irradiation position through the curved passage inside the outer biological shielding wall of the reactor. Due to construction properties of the rig only head part is subjected to radiation. That allows staff of the facility to load and unload samples and measure observable parameters without any trouble for the health.

As it shown in the Fig. 1, the head part is connected to charging device by a transport tube, which allows to load or to unload samples. Through the helium pipeline, helium is delivered from liquid helium Dewar vessel to the sample situated inside irradiation rig head and then it is collected in a gas accumulated vessel. Electric communication lines and PC intend for collecting data during an experiment.

The head part and terminal part of the helium pipelines are placed into an evaluated jacket with high vacuum. Keeping of high vacuum is utterly necessary to avoid wall heat load to the helium and to the samples.

There is a beryllium reflector set behind the head part of the rig. It was done to increase fast neutron flux with better isotropy.

2.1 Charging device

The charging device could be found in Fig 2.

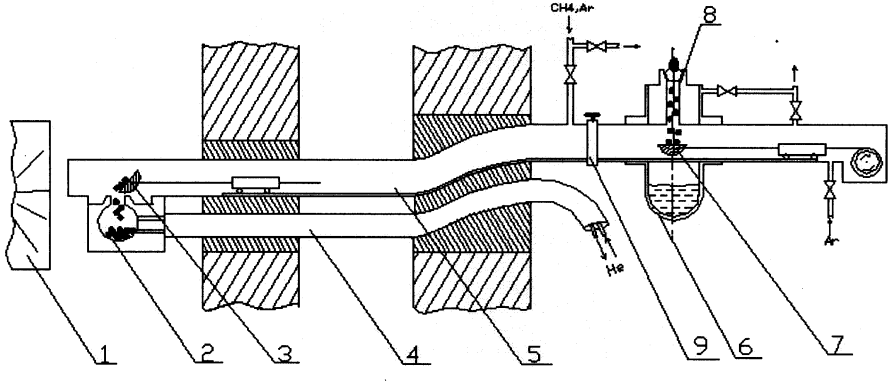

Fig 2. Conceptual design of the URAM-2 irradiation facility

(1) the IBR-2 reactor; (2) irradiation capsule; (3) carrying bowl with a cart in 'near of the reactor' position; (4) helium pipelines; (5) evacuated transport passage; (6) nitrogen cryostat; (7) carrying bowl with a cart in 'out of the reactor' position; (8) charging tube with plug; (9) vacuum lock

Before beginning of a sample preparation it is necessary to cool the irradiation cavity down to about 100 K. After it was done it is possible to charge materials have to be irradiated. They could be loaded by two ways: by charging beads of ice previously prepared or by condensing any gas into the irradiation cavity.

In the first case the volume behind the vacuum lock is filled with argon about 1.05 bar to avoid pollution of working volume by air. Then charging tube plug is opened and the ice beads are loaded through the charging tube, cooling by thermo-conducted buses connected to the nitrogen cryostat, into the carrying bowl with the cart, which had been set to the 'out of the reactor' position. Then the charging tube plug is closed, argon is pumped, the vacuum lock is opened and the charged bowl is moved along the transport passage to the 'near of the reactor' position, where it is unloaded into the irradiation cavity. (The carrying bowl with the cart could deliver safely into the irradiation capsule up to 150 beads of 5 mm diameter through 9 m length of a curved transport passage. Time of delivering is about 30 sec.) Depending on a type of preparing sample, the material could be melted and after that frozen as a hemisphere or saved as ice beads with freezing gaseous Ar into unfilled volume between beads. It enables to achieve better heat exchange between ice beads and cooling surface of the irradiation cavity. For the best quality of Ar ice, the crystallization should be done through the liquid phase.

In the second case the carrying bowl with the cart is set to the 'out of the reactor' position and the vacuum lock is closed. Then the needed amount of gas is filled into the evacuated transport passage and frozen onto the inner surface of the irradiation cavity. On this occasion the frozen gas will have a form of thin film. If a sample shape have to be a hemisphere, the film is melted and frozen again. There should be added some helium before melting to avoid evaporation of a material.

To remove a sample from the irradiation cavity, the helium supply is closed and the sample is melted and evaporated by reactor heat. Then the irradiation capsule and evacuated transport passage are pumped.

2.2 Head part of the facility

The head part of the facility is shown on Fig 3.

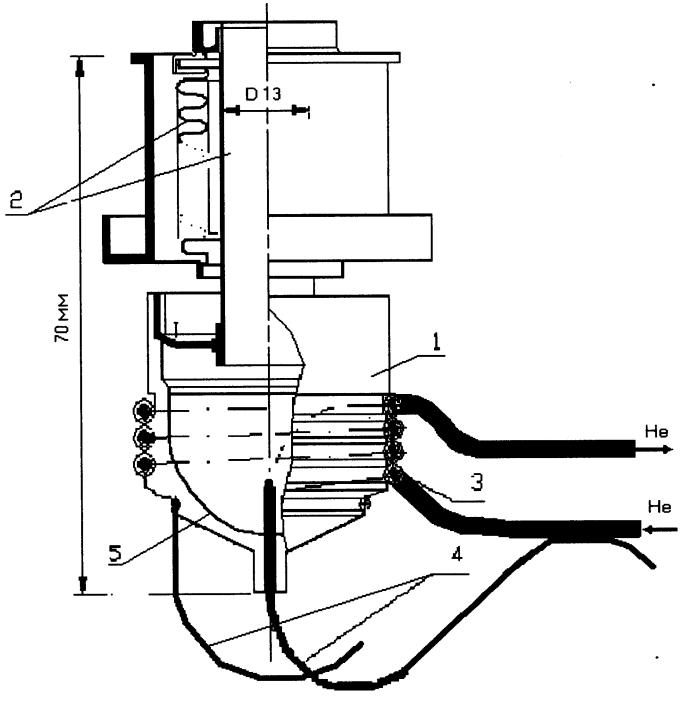

Fig 3. Head part of the facility: (1) capsule; (2) a suspender (thermal bridge); (3) cooling helium tubes; (4) thermocouples; (5) irradiation cavity.

Irradiation cavity inside the capsule has hemispherical shape with walls made of pure, oxigen-free copper (M1 purity of Russian standard, thermal conductivity 1000 W/m/K at 20K). The square of irradiation cavity is about 22 cm^2 and the inner volume is about 12 cm^3. The capsule is thermally insulated having a long neck of thin steel with a bellows. Total external heat (coming in through the neck and from the vacuum jacket walls) was estimated to be and really is ~1 W. A helium heat exchanger consists of four spires of a copper tube of 3 mm in diameter soldered onto a cylindrical wall of the

capsule. Nuclear internal heat in the wall is about 2 W. Together with heat in samples and heat loads on helium guide tubes, the total heat load is about 4.5 ÷ 5 W.

2.3 Cooling system

The cooling system of the facility is based on liquid helium, which is supplied from a Dewar vessel of 100 l capacity Fig. 1 (6), through the helium pipeline (3) to the irradiation capsule, Fig. 3. Then through another tube, helium is collected in gas accumulated vessel. With such cooling equipment it is easy to keep temperature of a sample constant during an irradiation. The variation of the temperature during an irradiation does not exceed 0.5 K.

There are three methods to change the temperature of the irradiation capsule.

- By handling the direct helium flow. This it the best precise regulation in all range of the helium consumption.
- By handling the back helium flow. This method is not so precise as the first one and because of flow rate oscillations it is possible to handle helium flow only up to 1 m^3/h.
- By changing power of a small heater, which is set inside the direct helium pipeline. This method is limited by changing the temperature up to 7 K. Nevertheless, it could be used for fast changing of the irradiation cavity wall temperature.

The liquid helium from a big Dewar is considered to be the most effective coolant from an economical point of view. 100 l Dewar is enough to work at temperature 20K for about 24 hour.

2.4 Measuring system

Temperature monitoring by thermocouples and PC with frequencies 1.25 and 5Hz are arranged at 5 points: one thermocouple is installed inside the capsule at a

distance of 8 mm from the bottom, two - at the outer surface of the capsule, and two of them measured temperature of input and output of cooling helium Fig. 4.

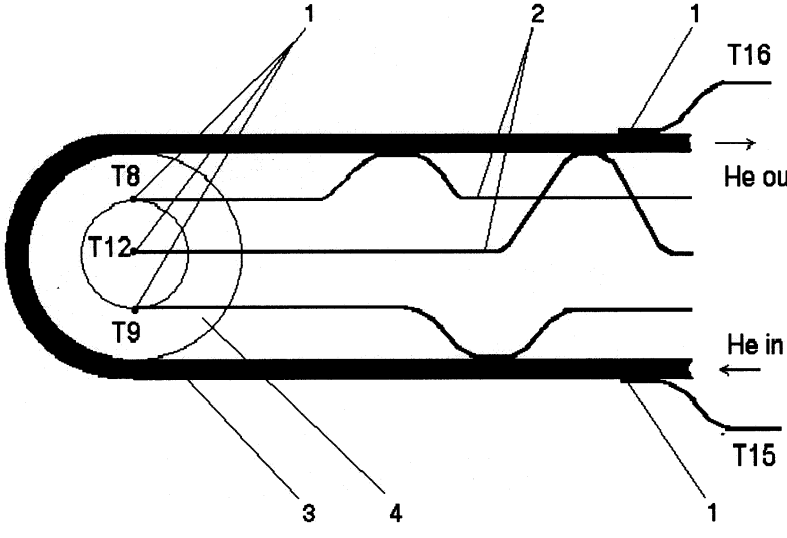

Fig 4. Arrangement of thermocouples: (1) thermocouples, (2) electric line, (3) helium pipeline, (4) irradiation capsule

Gas pressure monitoring is arranged with two gauges: 0-400 mbar, 0.1% accuracy at full scale, and 0-2 mbar, about 20% accuracy.

The helium consumption is measured by two flow meters 0-3 m^3/hour, 3% accuracy and 0.5-5 m^3/hour, with accuracy 0.2%.

The helium inside the Dewar vessel is controlled by superconductive indicator of helium level and a pressure control system.

3. Preliminary tests

Before the facility was constructed, some tests had been done.

1) The capsule was loaded with 20 cycles of rapid "cooling – heating" between 350 and 70 K. The tests showed no damage of the capsule.

2) Response of temperature of the walls of the capsule on a burp was measured by imitating a heat transfer from a sample with a flash of light

inside the capsule. It appeared to be 0.4 K/sec at 4W power of the light source, which is several times less than the expected power transfer during a burp. So that, a burp can be identified for sure by temperature jump at the walls of the capsule.

3) As long as condensed argon is heavier than ice, floating of ice balls would result in nonuniform distribution on them. Happily, temperature of condensation (87.6K at 1 bar) and freezing of argon (83.7 K, triple point) are very close. Therefore, fraction of liquid phase during condensation is expected to be minor. Nevertheless, it was decided to check the technology of ice balls in solid argon matrix preparation.

Installation was simple (see Fig. 5). A laboratory quartz glass flask, with a spherical volume of about 12 cm^3 simulated the URAM-2 sample chamber. It was connected to an ice balls loading site and to argon and vacuum supply through a long neck with a glass-metal junction. Liquid nitrogen in a transparent Dewar vessel with open surface served as a source of cool. The flask support was arranged so as move it easy up and down for a temperature control. Heat from a powerful electrical bulb was enough to simulate nuclear heat production in balls and argon. Ice balls were loaded through a funnel which was replacing a plug during a loading; gaseous argon prevented penetration of air into the flask volume.

In total, seven runs of freezing have been performed. Pressure of argon and time characteristics of argon supply were varied. When a pressure was kept constant during a condensation and freezing of argon, floating of beads occurred at pressure more than 1 bar. When argon was supplied by small portions, no floating was observed even at maximal pressure as high as 3 bar. As a result of these runs it was found that to avoid floating of ice balls during freezing argon at liquid nitrogen temperature, the rate of argon condensation should not exceed 0.5 cm^3 per minute.

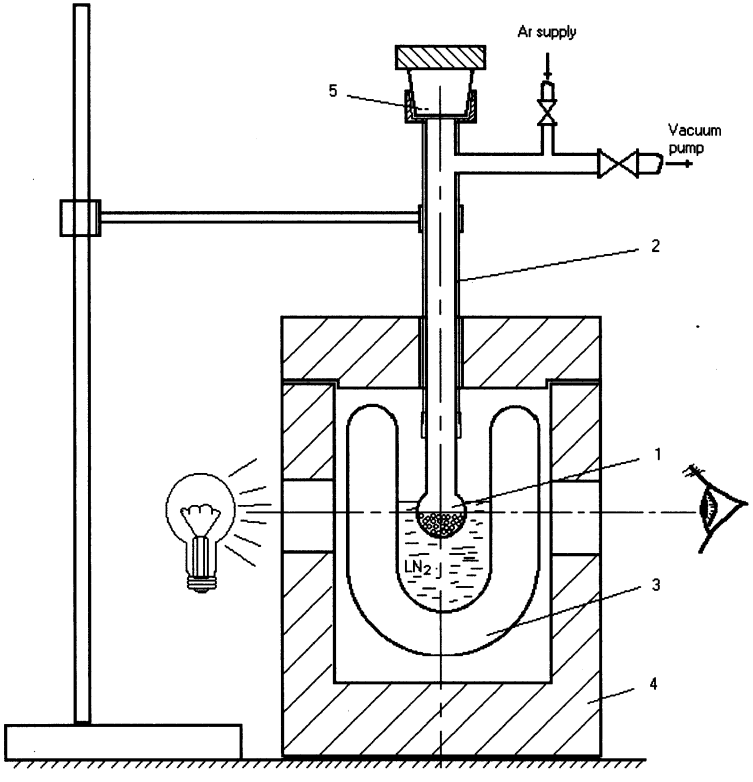

Fig. 5. Sketch of installation for "ice balls in solid argon matrix" preparation testing
1 – glass flak, 2 – metal tube, 3 – transparent glass Dewar vessel, 4 – foam plastic, 5 – a plug

4) The head part was modified to observe visually a process of methane condensation inside the capsule. The transport tube was replaced with a neck with an optical window. The device included also fiber, mirror and telescopic magnifying glass. Several runs of methane condensation into this modified capsule to make dense, transparent films of solid methane up to 2 mm thick or spherical segments were done. The best quality of a sample can be obtained at pressure-temperature condition being close to

the saturation curve. It is interesting to mention that phase transition of solid methane (at ~20K and ~14K) could be visually observed as waves inside methane sample caused by light reflection during the phase transition.

4. Main parameters of the URAM-2 facility

- fast neutron flux - 3 10^{12} n/cm^2/sec;
- absorbed dose rate: induced by γ - about 20 Gy/sec, by neutron in water - about 90 Gy/sec;
- maximal time of irradiation: about 24 hour at temperature 20 K;
- temperature range: 10 K ÷ 60 K
- possible sample material: any gases, low-boiling liquids.

5. Conclusion

Up to now some experiments at the URAM-2 facility has been done. Such materials as methane, water ice, clathrate of thetrohydrofuran, clathrate of methane were studied. Practically each experiment was successful. This means that at each experiment either spontaneous recombination of saved free radicals (see Fig. 6) or fast recombination induced by increasing a sample temperature (see Fig 7) was observed.

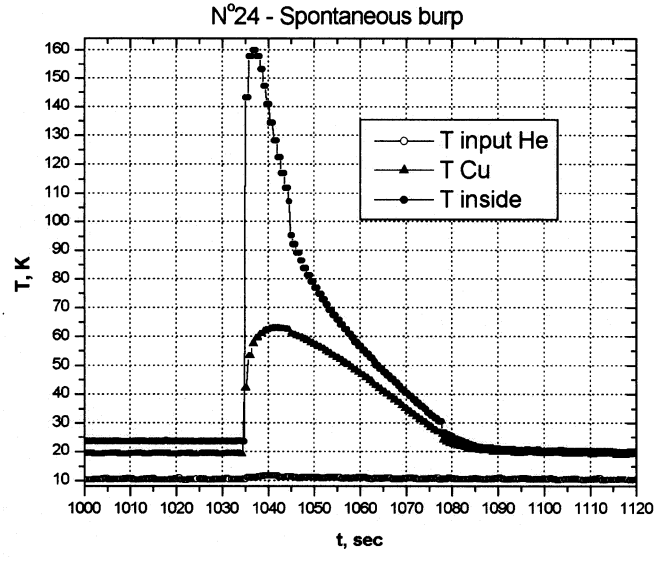

Fig. 6 Spontaneous recombination of free radicals ("burp") in water ice

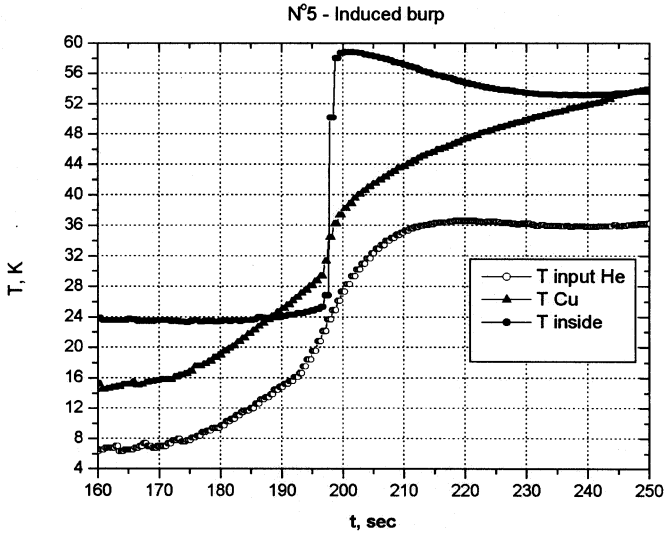

Fig. 7 Induced recombination of free radicals ("burp") in methane

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Криогенная облучательная установка УРАМ-2

На базе канала № 3 реактора ИБР-2 создана установка УРАМ-2 для исследования радиационных свойств материалов (таких как твердый метан, водяной лед и т. д.), перспективных для исследования холодных замедлителей нейтронов.

Система охлаждения установки основана на использовании жидкого гелия. Блок загрузки обеспечивает возможность помещения в облучательную камеру как предварительно подготовленных ледяных шариков, так и газа путем его конденсации внутри облучательной камеры.

После испытания узлов установки и монтажа ее на рабочем месте начаты эксперименты по изучению эффекта накопления химической энергии и ее спонтанного выделения при облучении материалов при низкой температуре.

Работа выполнена в Лаборатории нейтронной физики им. И. М. Франка ОИЯИ.

Сообщение Объединенного института ядерных исследований. Дубна, 2002

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URAM-2 Cryogenic Irradiation Facility

The URAM-2 irradiation facility has been built and mounted at the channel No. 3 of the IBR-2 reactor. It was constructed for study of radiolysis effects by fast neutron irradiation in some suitable for effective cold neutron production materials (namely: solid methane, methane hydrate, water ice, etc.).

The facility cooling system is based on using liquid helium as a coolant material. The original charging block of the rig allows the samples to be loaded by condensing gas into irradiation cavity or by charging beads of ice prepared before.

Preliminary tests for each facility block and assembling them at the working position were carried out. Use of the facility for study accumulation of chemical energy under irradiation at low temperature in materials mentioned above and its spontaneous release was started.

The investigation has been performed at the Frank Laboratory of Neutron Physics, JINR.

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