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Investigation of the energy accumulation rate in solid deuterium irradiated with fast electrons

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ABSTRACT

The feasibility studies for a new ultra cold neutron (UCN) source to be installed at the Institut Laue-Langevin (ILL) are underway. One of these studies concerns a source using solid deuterium (SD_2) as a neutron moderator. During source operation the SD_2 would be exposed to intense γ-ray and neutron fluxes. These may result in radiolysis in SD_2. Our work has shown that this effect—if it is produced—is not incompatible with a normal operation of the source.

1. Introduction

The so-called "burps" in solid-ice samples could occur under irradiation with γ-quanta, electrons, and fast neutrons [1–4]. They have been observed in a number of hydrogen-containing materials (for instance, in ice and solid methane). In these processes, molecules of the ice material are destroyed and the energy of the resulting free radicals accumulates. This energy can be released suddenly as the radicals recombine. The result can be a significant and violent increase in the temperature of the sample. This possibility has to be taken into account when considering the feasibility of SD_2 UCN sources [5–23]. If such a phenomenon occurs in SD_2, it might limit the time where the converter can be kept at low temperature. This could necessitate a periodical regeneration of the converter. In the worst case, this might even cause "burps" which could destroy the converter and/or even the source.

In fact, such stored energy effects had already been investigated for SD_2 and SH_2 samples irradiated by electrons, protons, and γ-quanta [24–31]. The energy from incoming 3 keV electron and 10 keV ion beams, partially absorbed in SH_2 and SD_2 samples at temperatures of 2.5–3 K, was studied in [24]. A phenomenological rate process theory was developed in [25] to explain the storage and rapid recombination of atomic-hydrogen radicals in crystalline molecular hydrogen solid at temperatures in the range $0.1\,K \leq T \leq 4\,K$. It quantitatively described the observed storage time as a function of temperature in many cases. This model can also be applied to SD_2. The infrared absorption spectrum of SD_2 at 4.2 K was studied with a sample irradiated by a 15 MeV proton beam [26]. Comparison of the beam-induced spectral features with those previously seen in a tritium-doped sample conclusively established the presence of a radiation-induced absorption process. After turning off the proton beam, these features persisted for the order of hours. They could be quenched by heating the sample to ~11 K. Charge-induced absorption spectra in the fundamental band of SD_2—under proton-beam irradiation—were measured in [27]. The timing experiments were presented there, in which the growth and decay of these spectral features are monitored following the onset and termination of beam irradiation. Both spectra and timing as a function of temperature were examined. In [28], electron-spin-resonance (ESR) spectra of H (D) atoms, produced in γ-irradiated SH_2 were studied at 4.2 K. The ESR linewidth of the trapped hydrogen atoms depends significantly upon the matrices of hydrogen isotopes. The ESR linewidth has been calculated with the orthogonalized wavefunction on the basis of the model presented. It is concluded from the comparison of the experimental ESR linewidth with theoretical ones that H atoms in solid H_2 are trapped in substitution sites, while H (D) atoms in solid HD and SD_2 are trapped in interstitial octahedral sites. In [29], the authors measured the isothermal build-up of the atomic-hydrogen (T and D) concentrations inside ST_2 and D–T from 1.3 to 10 K. The atoms are produced by the β decay of tritium. The measurements were made using adiabatic slow-passage ESR techniques. The atom concentration increases with time and with decreasing temperature; it reaches 0.2% below 3 K. In [30],

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samples of SD_2, cooled below 4.2 K, and irradiated by 15 MeV protons, spontaneously flashed. The flash spectrum occurred in the near infrared with a maximum close to 920 nm. The flash intensity was two to three orders of magnitude greater than a steady-state emission observed in the same spectral region. The flash frequency was 1 every 1 to 3 min after several hours of irradiation of 10 nA/cm^2. The heat pulses were detected accompanying many of the flashes and both can be stimulated by the application of external heat triggers. Following the optical flash, the steady-state emission intensity decreased by an amount that is wavelength dependent. While it is thought that these flashes are caused by atomic association in the solid, the species responsible for the optical emission and the transition are uncertain. Finally, optical emission from proton-beam-irradiated SD_2 was studied in [31] after termination of the proton beam. The continuous red emission showed a residual intensity that persisted over 30 min from termination of the beam. Optical flashes can also be thermally triggered over 10 min after termination of irradiation. Such triggered flashes were shown to quench the infrared absorption of Stark-shifted charge-induced features. Ultraviolet photons can stimulate this red emission after termination of irradiation with no measurable decrease in intensity for 40 min.

Although much data is available on comparable systems and some predictions for a SD_2 moderator can be deduced, the straight experimental test on a pure D_2 sample under representative irradiation has been missing. The closest full-scale test to the realistic Institut Laue-Langevin (ILL) conditions was probably carried out in Ref. [9] with 6 l of SD_2 installed in the PNPI reactor in Gatchina. However, the temperature was too high: 8–10 K instead of 5–6 K required; the heat load was too low; 0.03–0.05 W/g instead of a few tenths W/g required. No "burps" were observed under these conditions but this does not yet prove that this phenomenon—if it is produced—is compatible with a normal operation of the source.

In order to achieve maximum UCN density, we have to keep the source temperature low, at 5–6 K. Under the intense radiation inside the reactor the D_2 molecules decompose into D-atoms. At low temperatures the diffusion of D-atoms is very slow; the recombination might therefore also be slow. This would result in the accumulation of radicals. In this case, as the SD_2 heats, there is the possibility of a fast chain reaction leading to the recombination of D-atoms and a subsequent sudden increase in temperature. If the energy released exceeds 55 J/mole, any SD_2 sample would start to melt. The effect of such a rapid recombination of free radicals in SD_2 has never been directly measured. This is the reason underlying the present work. An adequate understanding of this process is essential for the construction of any SD_2 source in the world. It is particularly important for the ILL, given the powerful fluxes of cold neutrons and γ-quanta at the future source position (see Fig. 1).

Such a UCN source would be in line with the Munich project [10]. There, the UCN density is accumulated in a neutron guide or in a storage volume with low wall losses. UCN are produced in small volume solid-deuterium source placed in maximum flux of cold neutrons. In contrast to that, the ILL project would differ by new technology available, by additional constraints and advantages of its reactor, as well as by specific criteria for the optimization of our source: We would take advantage of specular neutron guides with nearly ideal transmission of neutrons at specular trajectories [32–34]. Long specular neutron guides of limited cross-section would be useful, in particular keeping in mind many geometrical constraints at the existing ILL reactor. In case of an installation of a third cold neutron source at the ILL, we would profit from higher initial density of cold neutrons in its vicinity. Otherwise a dedicated cold neutron converter around a

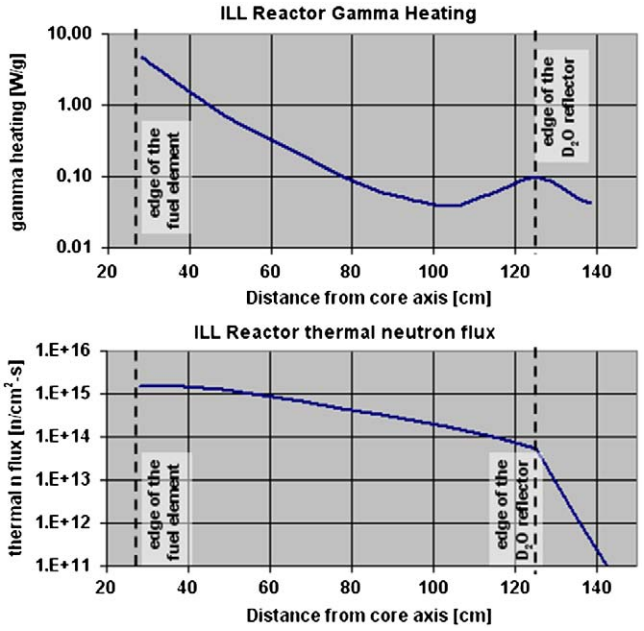

Fig. 1. γ-heating and thermal neutron flux in the ILL reactor. A possible distance of a SD_2 source from the core is 50–80 cm.

SD_2 UCN source would be needed. Anyway a SD_2 UCN source has to be placed in maximum flux of cold neutrons. We would also optimize our source for maximum brightness/density against the total flux of produced neutrons, keeping in mind such brightness-demanding projects as, for instance, the resonance transitions of neutrons between the gravitationally bound quantum states [35,36].

2. Tentative source at the ILL

No decision has been taken so far concerning the construction of a SD_2 UCN source at the ILL. Therefore, we will present only approximate parameters, leaving any detailed study for the future. The deuterium converter is installed in the vicinity of the reactor core, in a very high γ-flux (see Fig. 1). It has to be cooled down to a few Kelvin temperatures using liquid helium. In order to minimize the required helium flow, the heating load to be evacuated (i.e. the total mass to be cooled) has to be as small as possible. We studied various exchanger geometries and materials and favoured the configuration as shown in Fig. 2.

The heat exchanger shape is shown in Fig. 2 on the right. The heat exchanger is cooled by helium flow between the two plates. Deuterium is frozen on its cold surface from deuterium gas around the converter. Deuterium gas is injected directly into the neutron guide.

The choice of the exchanger material is constrained by the following criteria: (1) high critical velocity in order to allow storage of UCN, (2) high thermal conductivity in order to minimize the temperature gradients and thus the absolute temperature at the external skin, (3) low density in order to minimize the heating load, (4) mechanical strength compatible with standard operation load, also with extreme loads for safety requirements, (5) machinability in order to produce the exchanger. Beryllium is the best candidate from these points of view.

For the chosen geometry and material, we estimated the exchanger skin temperature and assumed growing of SD_2 along equal-temperature surface (see Fig. 3). Thus, we calculated the shape of the frozen deuterium and the temperature distribution in the bulk of the SD_2 converter under the known γ-heating level.