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Research paper

Solid methane in neutron radiation: Cryogenic moderators and cometary cryo volcanism

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ABSTRACT

The effect of ionizing radiation on solid methane has previously been an area of interest in the astrophysics community. In the late 1980s this interest was further boosted by the possibility of using solid methane as a moderating medium in spallation neutron sources. Here we present test results of solid methane moderators commissioned at the ISIS neutron source, and compare them with a model based on the theory of thermal explosion. Good agreement between the moderator test data and our model suggests that the process of radiolysis defect recombination happens at two different temperature ranges: the "lower temperature" recombination process occurs at around 20 K, with the "higher temperature" process taking place between 50 and 60 K. We discuss consequences of this mechanism for the designing and operation of solid methane moderators used in advanced neutron sources. We also discuss the possible role of radiolysis defect recombination processes in cryovolcanism on comets, and suggest an application based on this phenomenon.

1. Introduction

The properties of frozen methane, when exposed to ionising radiation, have attracted significant interest in the planetary and interstellar astrophysics community since the early 1980s [1,2]. In the late 1980s this interest was further boosted by the possibility of using solid methane as a moderating medium in spallation neutron sources [3]. Solid methane possesses unique neutronic properties that enable the conversion of hot, energetic neutrons into cold neutrons with an efficiency approximately 3.5 times that of liquid hydrogen based moderators [4]. However, practical applications of solid methane as a neutron moderator material turned out to be much more challenging than initially expected.

Early developments in methane moderators were carried out by the IPNS neutron source based at Argonne National Laboratory. Here solid methane at around 10 K was exposed to neutron radiation for few days, leading to a build-up of radiolysis products in the solid methane matrix. It was then shown that at some critical number of defects a spontaneous self-accelerated recombination process would take place, described by J. Carpenter as the 'burp' phenomenon [3,4]. This effect, in combination with the expansion of hydrogen built up in bulk solid methane during irradiation, was believed to be responsible for the moderator's breakdowns.

In the late 1990s the effects of neutron radiation on solid methane were systematically studied at the IBR neutron source in Dubna [4,5]. The results of this research confirmed the assumptions of Carpenter's model and gave some estimations of the scale of the process. After these experiments the consensus in the neutron scattering community was that all of the radiolysis products accumulated in solid methane (irradiated at temperatures around 10 K) start to recombine either spontaneously, if the density of the defects exceeds critical value, or by thermal activation mechanism during warming up if temperature exceeds 20 K. In either case, all defect recombination processes should be completed at temperatures below 40 K [3–6] and the design of the Target Station 2 Solid Methane Moderator (TS2 SMM), commissioned by the ISIS neutron source in 2009, was based on this assumption. However, the very first test of this moderator operated at 38 K ended in a similar burp-like event, which damaged the moderator.

This observation reignited interest in the results of the IPNS neutron source and suggested that the defect recombination process might be more complex that initially thought, with multiple defect types recombining at different temperature ranges.

Further support for this concept came from results obtained by researchers from the Verkin Institute in Kharkiv, Ukraine [7], who studied the radiation defect relaxation mechanisms in solid methane films, pre-irradiated by an electron beam. This experiment was able to measure both mass ejection and electromagnetic luminescence events. An expected peak of particle ejection (post-desorption) was found in solid methane at temperatures as low as 15 K, but another maximum was also observed at higher temperatures around 50 K.

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Here we present and discuss the results of commissioning tests of solid methane moderators used in Target Station 2 at the ISIS neutron source, and compare them with a model based on the theory of thermal explosion [8]. We also compare the results of our model simulation with the IPNS solid methane moderator test data published in [3]. The temperature range of the TS2 SMM commissioning tests was 37–100 K. The strong agreement between measured moderator temperature data and our simulation, combined with methane irradiation data from [7], suggests that the process of radiolysis defect recombination happens in two stages, at two different temperature ranges. The "lower temperature" recombination process occurs at around 20 K, with the "higher temperature" process taking place between 50 and 60 K.

We then use our model to simulate the pressure experienced by the walls of the TS2 SMM vessel produced by the rapid expansion of solid methane during self-accelerating heating, driven by the recombination process. We argue that this pressure spike can be intense enough to damage the vessel, which applies certain limitations on solid methane moderator design and operational procedure. We also discuss the possible role of radiolysis defect recombination processes in cryo-volcanism on comets, and suggest an application based on this phenomenon.

2. Design and commissioning of the ISIS Target Station 2 solid methane moderator

An illustrative sketch of the decoupled TS2 SMM is presented in Fig. 1. The moderator cryostat consists of an outer vacuum can (3), infrared radiation shield (4) and the solid methane moderator vessel (5). The "mark one" version of the moderator vessel was made of aluminium alloy 5083–0 with internal dimensions 124 × 140 × 70 mm and wall thickness 3 mm. The aluminium foam block (6) from aluminium alloy 1199-F and 8% density has been pressure fitted into the moderator vessel (5). The moderator comprises a heat exchanger coil (7) made of Ø 10 mm aluminium tube welded into the vessel's body. The fully loaded TS2 SMM (mark one) contained 405 g of CH_4. The high purity methane gas is condensed into the moderator through the inlet pipe (1) and exhausted through the outlet pipe (2) at the end of the operation cycle.

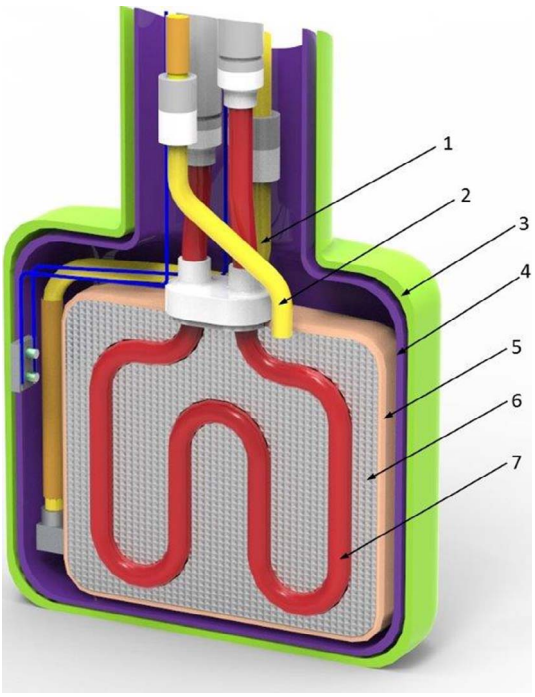

Fig. 1. Illustrative sketch of the decoupled TS2 Solid Methane Moderator. The description of the components is given in the text.

During testing, the moderator was irradiated by a neutron flux of $\sim 10^{16}$ neutron $cm^{-2}s^{-1}$, at a temperature of 38 K, for intervals of between 8 to 24 h. In three instances the self-accelerated heating process was triggered at ~50 K during an intentional warmup of the moderator (this is a standard procedure, carried out with the aim of annealing the solid methane) leading to moderator damage. In all three cases the damage occurred at temperatures around 65 K.

In the current moderator design (mark two), the thickness of the aluminium walls of the vessel has been increased from 3 mm to 10 mm. However this reduces the methane capacity of the moderator by almost half of the mark one prototype – 225 g of CH_4. Such radical design change has completely eliminated the possibility of moderator damage, but at the same time significantly reduced the cold neutron flux.

3. Model of solid CH_4 in neutron radiation

Our model utilises an approach similar to that used by Carpenter in [3] based on a simplified version of the theory of thermal explosion [8]. In order to reflect the presence of radiation defect recombination occurring at higher temperatures (50–70 K) we have introduced secondary binary collisions, with a rate coefficient K_2 that also has Arrhenius-form temperature dependence. Then the process of accumulation and recombination of radiolysis defects can be described by the equations:

$$\frac{dN_{1,2}(t)}{dt} = R_{1,2}(t) - K_{1,2}(T)N_{1,2}^2(t) \tag{1}$$

$$MC(T)\frac{dT}{dt} = P(t) + \varepsilon_1 K_1(T)N_1^2(t) + \varepsilon_2 K_2(T)N_2^2(t) - AH(T)(T(t) - T_{cool}(t)) \tag{2}$$

$$K_{1,2}(T) = K_{o1,2}e^{-E_{1,2}/k_B T} \tag{3}$$

Here, T is temperature, N_1 is the number of defects recombining around 20 K and N_2 is the number of defects recombining at around 50 K. Equation (1) is the same for both species, so for simplicity we use $N_{1,2}$ where index 1 and index 2 are for lower and higher temperatures of recombination respectively. $R_{1,2}$ is the defect production rate for each species. In our case of solid methane irradiated by fast neutrons, we assume that production rate is the same $R_1 \approx R_2$ for both species. M is the mass of the system, C is heat capacity of solid methane, H is the thermal conductivity, A is the area available for heat-exchange between the system and coolant, T_{cool} is the coolant temperature and P is the heating power provided by the external source.

Eq. (3) gives radiation defect recombination rate coefficients $K_{1,2}$ for each species, where $K_{0\ 1,2}$ are the recombination rates at infinite temperature for each species. $\Delta E_{1,2}$ are the activation energies for defect diffusion. We have used the activation energy of dislocation motion in solid methane at different temperatures [9] as an estimation for choosing $\Delta E_1/k_B = 108.6$ K and $\Delta E_2/k_B = 235$ K. In Eq. (2) ε_1 and ε_2 represent the heat released per defect annihilation. In our calculations we have used 218 kJ $mole^{-1}$ for $H + H \rightarrow H_2$ reaction (which, we assume, is the main reaction at lower temperatures) and 368 kJ $mole^{-1}$ for $CH_3 + CH_3 \rightarrow C_2H_6$ [3] (which is expected to happen around 50 K) ignoring all other reactions. Our model also makes use of linear interpolations from the solid methane thermal conductivity data in [10] and heat capacity in [11].

The differential Eqs. (1)–(3) are inherently non-linear and so we solve them numerically in order to produce a simulation of the system's behaviour under different parameters. Starting at some known temperature, with a fixed defect production rate $R_{1,2}$, we calculate the new number of defects $N_{1,2}$ from Eq. (1) over our suitable chosen time step dt. Eq. (2) shows the balance of power into, and out from, the system, from which calculating the step increase in T over dt is simple. The process then loops over the chosen length of time.

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