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On the initiation of DD reactions in the zirconium–deuterium system

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The possibility of initiating DD reactions on vibro-milling of zirconium together with heavy water and deuterated polypropylene has been investigated. Weak emission of neutrons has been detected both during the vibro-milling process and some time after its completion.

1. Introduction

At present, an active investigation of the possibilities of carrying out nuclear synthesis reactions (of the DD type) is going on in metal–deuterium systems [1–3]. The results obtained prove to be extremely conflicting and require thorough analysis. It is clear, however, that the possibility of observing DD reactions and the corresponding neutrons depends both on the conditions of sample preparation and on the experimental conditions. In this connection, metals of group IV of the periodic system are of the greatest interest, since these are able to be strongly saturated by deuterium (the ratio of deuterium to metal is 2:1), and possess very low equilibrium phase pressures of deuterium, say, 10^{-4}–10^{-3} mm Hg [4].

Now, to increase the dissolution rate of deuterium in the lattice of such metals requires a high dispersity of the powder particles used. Moreover, to attain a high deuterium dissolution rate the surface of the metal samples should be saturated by dislocations and point defects that serve as sinks of deuterium and as catalysts of the decomposition of D_2 molecules into atoms [4]. In our opinion, the latter circumstance is one of the causes of the poor reproducibility of the results of the initiation of DD reactions in the titanium–deuterium system whether in the course of saturation of titanium from the gas phase or during electrolysis in D_2O [1,5]. The thermal vacuum treatment of titanium in the deuterium atmosphere also does not lead to an increase in the reactivity of the samples, since this does not cause the formation of a fresh surface.

The suggested method of carrying out DD reactions of synthesis is free from such a substantial drawback. The method is based on the injection of deuterium through the freshly formed titanium surface which possesses a high reactivity and has not had sufficient time to undergo oxidation during the characteristic time of failure of the samples, amounting to less than 10^{-6} s (the characteristic time of the developing of a crack through a particle having a transverse dimension smaller than 1 mm).

We have already shown earlier the essential possibility of generation of neutrons in the titanium–deuterium system in the course of the dispersion of titanium in a vibro-mill together with deuterated substances [3,6].

2. Experimental

In the present work, we have used the possibility of initiating DD reactions in the course of deuteration of another metal of group IV, i.e., zirconium during its joint dispersion with deuterated substances. D_2O, purity 99.8%, and polypropylene PP(D_6), with a degree of deuteration of 98.5%, were

used as deuterium carriers. Zirconium, purity 99.9%, was used in the present research work, zirconium being not subjected to a preliminary thermal treatment. The Zr chips having dimensions $2\times1\times0.2$ mm^3 were mixed with 4% PP(D_6) and 10% D_2O, and placed into a steel sealed drum filled for two-thirds by steel balls.

As has been demonstrated in refs. [3,6], just such a ratio of deuterated components in the course of vibro-milling (deuterating) of titanium brings about the maximum effect of generation of neutrons. For comparison, individual milling of Zr was carried out, as well as vibro-milling of a Zr + 4% PP(H_6) + 10% H_2O mixture – that is, under the conditions that deuterium is absent from the system (blank experiments). After a total milling time of 1800 s, the specific surface of the obtained ZrD_x powder was measured by the BET method in accordance with the low-temperature adsorption of nitrogen, and amounted to about 6.0 m^2/g for the Zr + 4% PP(D_6) + 10% D_2O systems. After vibro-milling, part of the ZrD_x powder was dried and then rinsed in boiling benzene for the purpose of removing polypropylene. Subsequently the deuterium content was evaluated by the high-temperature extraction method. As was experimentally shown, in 30 min of vibro-milling the value of x amounted to about 1.8. However, it will have to be noted that this value cannot be regarded as being very accurate, since no total washing off of polymer occurs, whose pyrolysis products may to a sufficient degree distort the results of measuring the content of deuterium in zirconium.

All the experiments of the present Letter were carried out in an eccentric ball vibro-mill having an applied power of 10 W/g, as described in refs. [3,6].

The recording of the neutrons was effected by the use of a unit made of seven proportional counters immersed into a cadmium covered tank filled with vacuum oil. The dimensions of the tank were $250\times250\times700$ mm^3. The recording circuit is described in refs. [3,6]. The detector efficiency was determined by the use of a calibrated Po–α–Be neutron source having an intensity of 200 neutrons/s, which was placed into a cell of the vibro-mill instead of the working drum. During the whole working period, the natural neutron background was also taken into account as measured with the working drum not in the cell. The cosmic background was fairly permanent, and amounted to about 0.030 neutrons/s.

During the experiments, the shape and duration of the pulses being recorded were continuously controlled by the use of a memory oscillograph, and compared with similar parameters of the pulses of neutrons yielded by the Po–α–Be source. This neutron source has a maximum neutron energy of about 3 MeV, which is close to the maximum energy of neutrons resulting from the $d+d\rightarrow{}^3He+n$ reactions. Measurements carried out with the use of a neutron source have demonstrated that most of the pulses have an energy corresponding to that of channels 2–32 of the AI-264 style amplitude analyzer used by us (fig. 1a). Therefore, we identify the pulses getting into channels 1–11 of the analyzer with electromagnetic disturbances. As has been shown by the experiments, both in the vibro-milling process (fig. 1b, 1c) and after switching it off, the number of pulses in the low-amplitude channels is insignificant as compared with that of the "neutron" pulses that are recorded in channels 12–32 of the analyzer. Therefore, we concluded that there was a minimum influence of the electromagnetic and acoustic microphone on the reliability of the experimental results.

Measurements carried out with the use of a neutron source have also demonstrated that the efficiency of a detector at a distance of 15 cm (the working distance) from the neutron source amounted to $E=1.1\times10^{-3}$. Knowing the intensity of the natural background allows a determination of the minimum number of neutrons, P_D, that could be registered by our detector at a level 2σ higher than the background [5]. In other words, P_D is the minimum number of counts per unit time, which may be considered as an effect, as was accepted, for example, in ref. [5]:

$$P_D=2\sqrt{R_b/E^2t}\,, \qquad (1)$$

where R_b is the natural background, E is the detector efficiency, and t is the exposure time. Substituting $R_b=0.03$ neutrons/s, $E=1.1\times10^{-3}$ into eq. (1) and assuming, for example, a recording time interval $t=3600$ s, the minimum number of neutrons, which may be considered as an effect, amounts to about 5 neutrons/s during one hour.

Control experiments carried out on pure zirconium without deuterium have shown that no excess over the natural neutron background is observed both

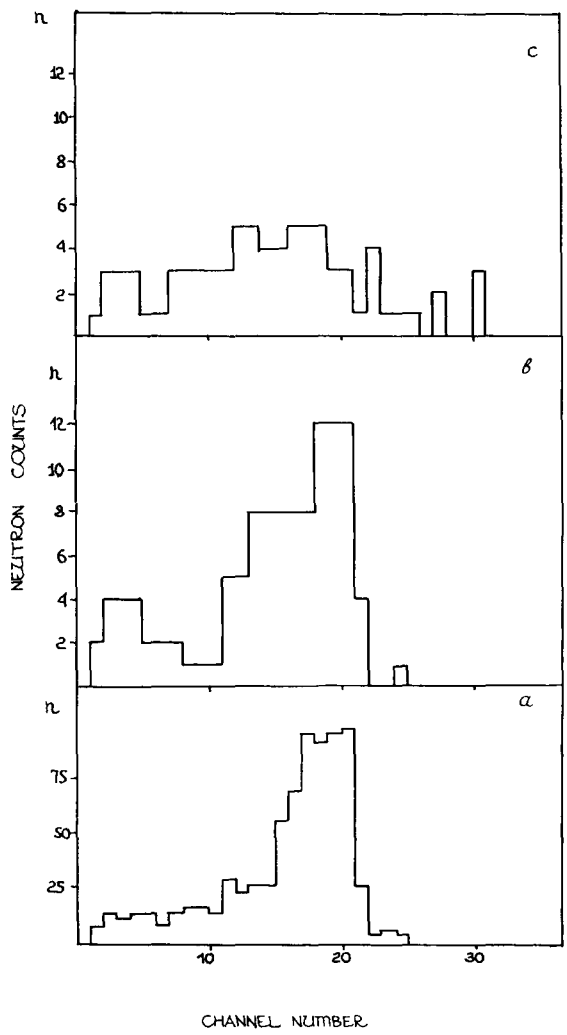

Fig. 1. Histogram of the distribution of pulses over the channels of the amplitude analyzer: (a) Po–Be neutron source; (b) vibro-milling of the Zr+4% PP(D_6)+10% D_2O system during 1800 s; (c) vibro-milling of the Zr+4% PP(H_6)+10% H_2O system during 1800 s (blank experiments).

in the course of milling Zr (as frozen in liquid nitrogen) and on its completion.

The subsequent experiments on the pure Zr+4% PP(H_6)+10% H_2O system have also led to a value that does not exceed the value of the natural neutron background within the limits of the statistical error (fig. 1c). Similar results were also obtained on the completion of vibro-milling, as well as in the process of freezing of the working drum in liquid nitrogen.

3. Results and discussions

The experiments on the deuteration of zirconium were carried out under conditions similar to those described above for the Zr–H system. The algorithm of the experiments was as follows:

Process 1. The working drum containing a Zr+4% PP(D_6)+10% D_2O (per 10 g Zr) mixture was frozen in liquid nitrogen to a temperature of -160°C. In this case, we note that no changes in the temperature of the counter block did occur in the drum freezing process. After the freezing of the drum, the vibro-milling of the system was effected during 3 min. (The technical conditions of the operation of the vibro-mill do not allow any prolongation of that time.)

Process 2. On switching off the vibro-effect, the drum remained in the cell, and the neutron background was registered during a period of 10 to 15 min (the so-called "post-effect"). In this time, the temperature of the "outer wall" of the working drum would rise from 0°C (on the completion of the vibro-effect) to 25–30°C.

Process 3. The repeated freezing of the drum was effected directly in the vibro-mill cell during 6 to 9 min down to a temperature of -160°C with the simultaneous recording of the neutron background. Then, the 1–2–3 cycle was again repeated, etc.

In fig. 1b we present a histogram of the distribution of pulses over the channels of the energy analyzer, which has been obtained during a total vibro-milling time of 30 min (e.g. process 1). As appears from fig. 1, the shape of the histogram and the position of the maximum agree well with the norm of the source (fig. 1a). Similar amplitude spectra were obtained for processes 2 and 3.

In table 1 we present the data thus obtained. In table 1, τ is the total exposure (or recording) time for each type of work, P_D is the 2σ level for each exposure time, calculated with formula (1); $(N-R_b)/E$ is the effect having subtracted the cosmic neutron background, $R_b=0.03$ neutrons/s (while taking into account the detector efficiency, E);

$$L=\frac{N-R_b}{(R_b/\tau)^{1/2}}$$

is the level of trustworthiness of the effect observed (σ is the standard deviation).

Table 1
Parameters of neutron emission for the Zr–D system.

Parameter	Process 1 vibro-milling	Process 2 post-effect	Process 3 freezing
τ (s)	1860	3240	2160
P_D (neutrons/s)	7.4	5.5	7.1
$(N-R_b)/E$ (neutrons/s)	17.3	18.2	24.5
L (σ)	4	6	7

As appears from table 1, a substantial increment over the natural background level is observed. The most essential effect, exceeding 7σ, is observed in the drum freezing process with the object under investigation being in liquid nitrogen. A similar maximum of the effect was also observed for the titanium–deuterium system in the course of freezing [3,6]. The post-effect has also a considerable value, and exceeds 6σ.

We note that in separate experiments the duration of the after-emission of neutrons attained 30 to 60 min. However, the cause of the longer duration of the effect is so far unclear.

The emission of neutrons observed is of a strongly unsteady character.

Hence, if we admit the existence of the pulse neutron processes associated, for example, with the formation of cracks in zirconium [7], whose duration is very short (a few μs), then the efficiency of our detector can be reduced by many orders of magnitude, while the real effect may be much higher [5]. Therefore, in the present case, it is desirable to employ detectors having a short "dead" time.

We have concluded that when the zirconium–deuterium system undergoes mechanical stress, emission of neutrons is observed, which are produced by single or separate DD reactions occurring in the course of the deuteration of its surface.

We also suppose that generation of neutrons is also possible when other transition metals that are likely to form deuterides in the presence of deuterated substances, are subjected to a mechanical treatment.

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