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DEPARTEMENT DE PHYSIQUE DES PARTICULES ELEMENTAIRES

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Abstract

In "cold fusion" experiments with heavy water a source of neutrons is the dissociation of deuterium induced by alpha particles emitted by natural occurring radioisotopes. We evaluate the rate of fast neutron emission as a function of the concentration of U, Th, Rn in contact with deuterium and discuss the possibility that the neutrons claimed to have been observed in "cold fusion" experiments could be due to this conventional source.

Alpha particles of ^{212}Po (8.8 MeV) and of ^{214}Po (7.8 MeV) can dissociate deuterium through the reaction :

$$\alpha + d \rightarrow \alpha + n + p \qquad (1)$$

The α energy for this reaction at threshold is 6.6 MeV in the laboratory system or 2.2 MeV in the center of mass. The neutrons have a spectrum extending up to 3.1 MeV, with a large fraction above 2 MeV. The cross section for this reaction was measured by H.L. Schutz in 1938 to be 1 mb at 8 MeV [1]; it has more recently been measured to be 0.3 barn at 19 MeV [2]. The energy dependance of this cross section from 4 to 8 MeV in the center of mass [3] shows no decrease towards low energies and demonstrates that the reaction proceeds via $^{6}Li^{*}$ (5.65 MeV) which further decays into ^{5}He + p. Although the threshold for $^{6}Li^{*}$ production is 4.2 MeV for reaction (1), the width of the state is so large (1.5 MeV [3]) that the effect of this resonance should still be important at threshold (2.2 MeV). Note also that ^{5}He has a large width of .6 MeV.

Due to these arguments and to the fact that no new measurements of the cross section of reaction (1) have been performed near threshold since 1938 (to our knowledge), the following calculations have assumed arbitrarily a value of 0.1 barn for the cross section at 8 MeV, giving an interaction length of 1.5 m in heavy water. If the deuterium is in a chemical form like TiD_2, or inside palladium (D/Pd = .6), the microscopic concentration of deuterium is probably close to that of heavy water.

Uranium and Thorium, the parents of the naturally occuring radioisotopes are normally present in material at the ppm ($\mu g/g$) level. Experiments designed to observe very rare processes (solar neutrinos, double beta decay, dark matter searches,...) must measure many materials to select those with very low radioactivity. Radium can also be present at the level of few pCi per gram, even in the absence of uranium contamination.

In a closed system, uranium is in equilibrium with all its daughters products, and the activity of each of these species is the same. Figure 1a shows a diagram of Uranium decay products. The long half life of ^{238}U (4.5 10^9 years) determines the activity. Radon is gaseous and the equilibrium of the decay chain can be broken if ^{222}Rn escapes. A chemical treatment to obtain pure uranium can also break the equilibrium. The ^{214}Po decays with a half life of 150 µsec by emitting an α particle of 7.8 MeV.

Thorium (^{232}Th, $t_{1/2}$ = 1.39 10^{10} years) is responsible for another family (fig 1b) of radionuclides. ^{220}Rn is a member of this family. Among the daughters ^{212}Po decays with a half life of .3 µsec by emitting an α of 8.8 MeV (branching ratio : 64%). ^{208}Tl decays by emitting a gamma of 2.61 MeV (branching ratio 36%) with a half life of 180 sec.

The energy loss (dE/dx) of α particles in heavy water is 630 MeV/cm for a kinetic energy of 8 MeV, rising to 780 MeV/cm when kinetic energy is 6 MeV. After travelling 28 µm in heavy water, the α hast lost enough energy to be below threshold of reaction (1). In titanium, $\langle dE/dx \rangle \approx 1900$ MeV/cm. If titanium sponge is used, the size of the grains composing the sponge must be considered. The probability of interaction (1) for an α emitted with 8 MeV of kinetic energy can be computed for the cases of interest (assuming $\sigma = 0.1$ barn):

- in heavy water, the probability is 1.6 10^{-5}
- deuterium combined into metal, the probability is 0.6 10^{-5}
- in pure deuterium, the probability is 6 10^{-5}

To reproduce the signal detected by S.Jones et al.[4] of 4 10^{-3} neutron/s it is therefore necessary to assume that the α's interact in heavy water (the efficiency of 1% to detect fast neutrons in the counter is assumed) and that the activity in the apparatus is around 50000 Bq of ^{214}Po (Uranium series) or of ^{212}Po (Thorium series). It is common to find samples of precious metals like palladium, with activities of 1000 Bq/g [5]. Nevertheless this contamination seems high if it is due only to uranium or thorium.

Radon is present in the air naturally. In houses, the walls contain radionuclides and emit radon continuously. Quantities vary strongly, but typical value are above 3.7 Bq/m^3 or a concentration of 1 atom of radon per 10^{19} "air molecules". The half life of ^{222}Rn is 3.82 days. The other short lived isotope ^{220}Ra ($t_{1/2}$ = 55 sec) is also present.

Radon, like other gases, is adsorbed easily in the kind of materials used in "cold fusion" experiments. Sponge or rough surfaces increase dramatically the adsorption capacity, even at room temperature. Table 1 shows measured values of adsorption of rare gases on charcoal at 0°C [6]. Thus the adsorption capacity for radon is clearly not a limiting factor, but rather the amount of radon present in the air. The quantity needed to explain the activity quoted above is around 3 10^{10} atoms of ^{222}Rn or 2 10^9 atoms of ^{220}Rn. Whether radon was present at this level in the materials used in the experiments depends on the adsorption before and during the experiment.

Once radon is adsorbed on the electrode, it decays as shown in figure 1. After some 2 hours ^{214}Po or ^{212}Po is present and produces α particles as explained above. Very different quantities of radon could be adsorbed by different electrodes during their storage prior to use in the experiments. Heating the electrodes is not sufficient to remove the adsorbed gases; the usual cleaning technique is to heat the titanium to 800°C in vacuum.

During electrolysis experiments, a large amount of gas is produced at the electrodes. This gas can remove the radon adsorbed on the electrodes or dissolved in the water, breaking the radioactive chain, so that the radioactive signal could disappear with time. In fact, a very efficient technique for purifying liquids containing undesirable dissolved gases is to bubble any pure gas

through the liquid. The dissolved gases escape with the bubbles of pure gas. As an example of this technique, in solar neutrinos experiments (Chlorine experiment, Gallex) as few as 10 radioactive atoms in a volume of more than 100 tons of liquid can be removed merely by bubbling pure helium or nitrogen through the liquid [9]. Flushing with a volume of gas equal to ten times the volume of the liquid removes the dissolved impurities with an efficiency of 90%.

In the experiment of S.Jones et al. [4] runs 1 to 14 provide evidence for the emission of fast neutrons. Several important conditions change from run to run, the electrolyte is modified, heavy water is added, and the cell and the electrodes are changed. Moreover, the background measurements were made before and after, but not during the series of runs yielding positive results. Different pieces of equipment were used in the background measurement. In particular, if D_2O is replaced with H_2O, the type of background described above would obviously disappear, thus reinforcing the signal of fast neutrons.

Heavy water and light water have also different neutron transmission characteristics. Cells filled with light water could act as a shield for the neutron counter. In addition, if LiOH is used as an electrolyte, lithium may migrate to the cathode where it can also be a source of fast neutrons due to (α,n) reactions. A yield of $4.7\ 10^{-6}$ neutrons per α of a polonium source is quoted by E.Segre [8].

An experiment performed by F.Scaramuzzi et al.[9], which claims to observe fast neutrons emitted from titanium in the presence of pressurized deuterium, can also be explained by natural radioactivity. Titanium cooled at liquid nitrogen temperature is a better adsorbant by several orders of magnitude than titanium at room temperature, so it would trap radon along with other gases present in the laboratory. In the presence of pressurized D_2, reaction (1) can take place, even more efficiently than in the electrolysis experiment, as soon as enough $^{214}Po/^{212}Po$ has been produced from this fresh source of radon (about 2 hours is necessary to reach the equilibrium activity). Once all the radon products have decayed, the signal will disappear as observed in this experiment and also in S.Jones et al.[3]. At the end of the experiment, some radon will be trapped again before the temperature rises, and neutrons will again be produced for a short time until the gases (including D_2 and radon) desorbs from the titanium.

Finally, the mechanism of reaction (1) could nicely reconcile some of the observations reported by M.Fleischmann and S.Pons [10] and by S.Jones et al. [4]. In Fleischmann's experiment, fast neutrons are poorly measured, but there is a convincing γ ray peak at 2.2 MeV. This is interpreted by the authors as coming from the neutron capture reaction in the large surrounding thermal bath of light water:

$$n\ (2.45\ MeV) + p \rightarrow d + \gamma\ (2.5\ MeV)$$

The rate of this reaction is reported to be $4\ 10^4$/sec. We assume that these γ rays are coming from ^{208}Tl present in the thorium chain (2.61 MeV). Such a rate implies that $8\ 10^4$ α/sec are emitted by ^{212}Po.We thus predict a production rate of the order of 1 neutron/sec in the S.Jones experiment, very near the measured value (.41 neutron/s).

Another source of fast neutrons are the (α,n) reaction on ^{17}O and ^{18}O (threshold -0.58 MeV and 0.7 MeV respectively). For α anergies, >5 MeV above threshold (twice the Coulomb barrier), the cross sections are about 1 barn [12] , but the natural isotopic abundances of ^{17}O and ^{18}O are only .037 and .2 % . The average energy of the produced neutrons is 3.5 MeV. Note that these reactions would also operate on light water.

A last source of neutrons come from the reaction :

$$\gamma + d \rightarrow \gamma + n + p \qquad (2)$$

A 2.6 MeV γ has a 10^{-3} probability of photodisintegration of the deuteron (for 10 cm of heavy water),leading however to neutrons of rather low energy (200 keV), below threshold in the experiment of S.Jones et al. [4], but which could have been detected with thermal neutron counters.

Conclusion

As long as the signal of neutrons in "cold fusion" experiments is small, careful experimental procedures are necessary to detect fast neutrons unambiguously in these devices. In particular, all elements of the experimental apparatus should be carefully outgassed before performing the experiment. Materials should be selected for low radioactivity. Background measurement capable of discerning artefacts due to natural radioactivity must be made.

Due to uncertainties in the cross sections of reaction (1) it is necessary to undertake experiments of electrolysis with known contamination of radium or thorium in the electrolytes, or radon in the electrodes. This program is now underway, using the neutron detectors operated by the Bugey collaboration [11] to allow the best neutron identification. With these measurements we will have enough information to evaluate the importance of this source of neutrons.

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Table 1 : Volume in cm^3 (STP) of adsorbed gas by 1gr of charcoal for a partial pressure of 1 torr at 0°C [8]. For radon it is an extrapolated values of the possible capacity. Values in titanium sponge are at least equal.

Gas	Z	cm^3/g
Ar	18	0.058
Kr	36	0.34
Xe	54	1.58
Rn	86	> 2.00

Fig. 1a

^{238}U chain

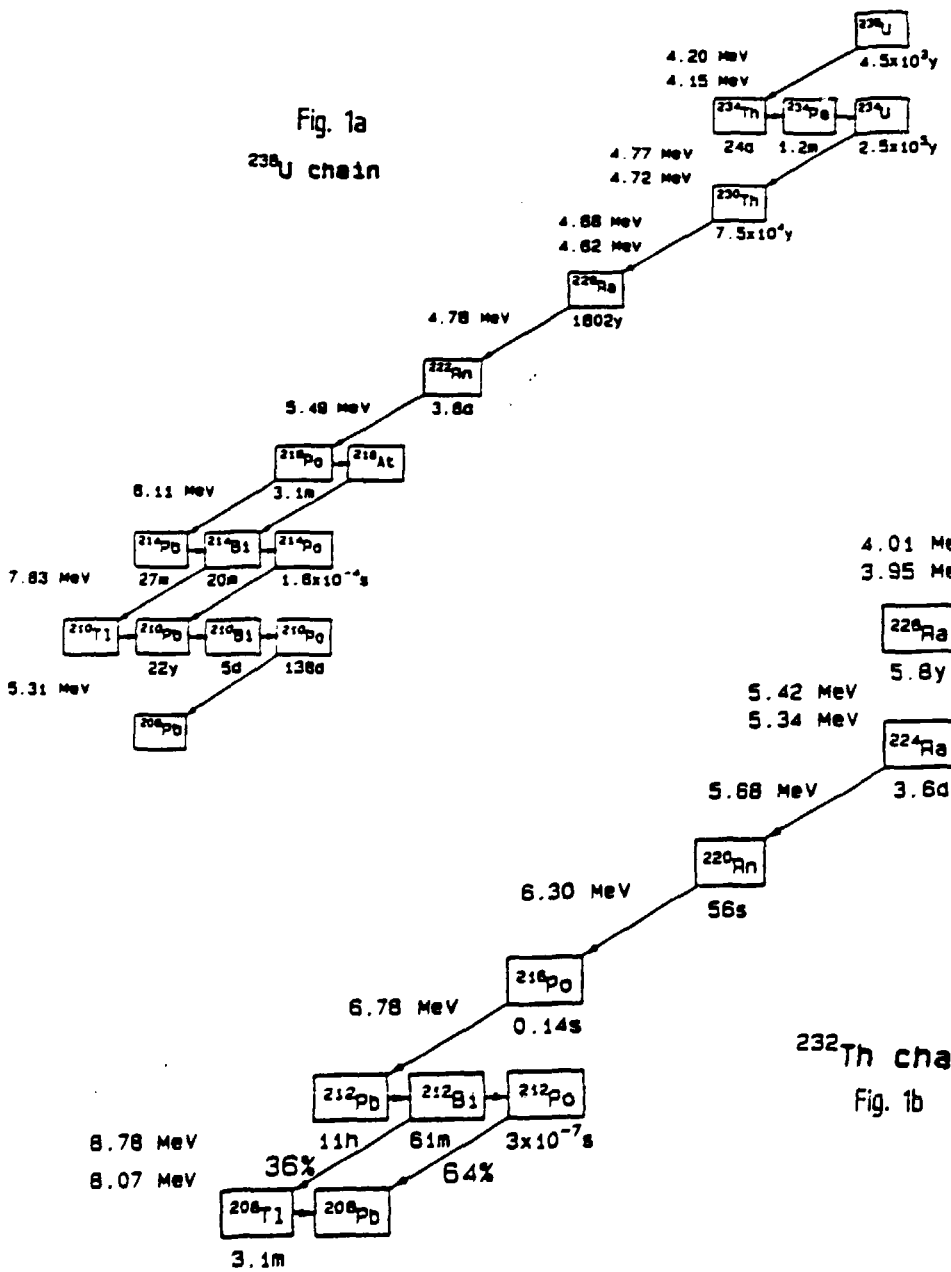

Fig. 1b

^{232}Th chain