

Constraints on energetic particles in the Fleischmann–Pons experiment

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Abstract In recent Fleischmann–Pons experiments carried out by different groups, a thermal signal is seen indicative of excess energy production of a magnitude much greater than can be accounted for by chemistry. Correlated with the excess heat appears to be ^4He , with the associated energy near 24 MeV per helium atom. In nuclear reactions, the energy produced is expressed through the kinetic energy of the products; hence, it would be natural to assume that some of the reaction energy ends up as kinetic energy of the ^4He nucleus. Depending on the energy that the helium nucleus is born with, it will result in radiation (such as neutrons or x-rays) that can be seen outside of the cell. We have computed estimates of the expected neutron and x-ray emission as a function of helium energy and compared the results with upper limits taken from experiments. Experimental results with upper limits of neutron emission between 0.008 and 0.8 n/J are found to correspond to upper limits in alpha energy between 6.2 and 20.2 keV.

Keywords Fleischmann–Pons effect

Introduction

From the initial announcement of the Fleischmann–Pons experiment in 1989 (Fleischmann et al. 1989), there has been much controversy from problems associated with reproducibility and measurement error to the suggestion that nuclear reactions are involved. After 20 years of

research in the area, some of scientific issues have been clarified to some degree, although at present, there is no consensus on what new physical process is responsible.

We recall that in the Fleischmann–Pons experiment, a palladium cathode is loaded electrochemically with deuterium in heavy water (0.1 M LiOD), and an anomalous thermal response is observed in some experiments. The cell temperature in Fleischmann and Pons experiments showed a sustained increase which could not be accounted for without the presence of very significant energy input. The thermal energy excess in one experiment reported in Fleischmann et al. (1990) was about 4 MJ over about 80 h (see Fig. 10a in that paper), with a rod cathode 1.25 cm long, 4 mm in diameter, and volume of 0.157 cm^3 . If this cathode had been replaced by trinitrotoluene and detonated, the energy liberated would have been about 1.2 kJ.

Since there was little change in the chemical composition of the cell after the experiment (which would perhaps be expected if the excess energy was of chemical origin), Fleischmann and colleagues proposed that this energy was of nuclear origin. In recent times, it has become customary to refer to this as the Fleischmann–Pons effect (or, from our perspective, one aspect of it), which means a significant thermal increase in Pd loaded electrochemically, of a magnitude much greater than can be accounted for from chemical processes.

In a nuclear reaction in which energy is produced, we would expect that the reaction energy would appear as kinetic energy of the products. Consequently, if the energy in the Fleischmann–Pons experiment were due to an unknown nuclear reaction, then it should have been possible to tell from the observations of the energetic products. The absence of commensurate neutrons indicates that the known deuterium–deuterium reaction $^2\text{H}(^2\text{H},n)^3\text{He}$ is not responsible. To date, no energetic products in amounts consistent with the energy produced have been identified.

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There are different conclusions which might be drawn from this. One possibility is to assert that if no commensurate energetic particles are present, then no nuclear reactions occur. This has been the point of view of most of the physics community since 1989 and suggests that the thermal effect are due to measurement error or an artifact. This point of view is supported by one of the foundations of nuclear physics, which holds that energetic reaction products are a necessary consequence of energy and momentum conservation.

However, the great many subsequent observations of excess heat in the Fleischmann–Pons experiment (McKubre et al. 1994; Storms 2007) argue for a different conclusion. In addition, ^4He has been observed in the gas phase in amounts in proportion with the energy produced (Miles et al. 1993, 1994; Miles 2004; Hagelstein et al. 2005). The latter observation was unexpected and remains even now an astonishing result. Such a large amount of excess energy produced with commensurate ^4He as a product can be interpreted as indicative of a new physical process. Although there is general agreement on the part of those working on the problem that this is the case, there is much less agreement as to what specific process is responsible.

At recent workshops at the Naval Postgraduate School, some of our physics colleagues argued strongly in favor of mechanisms in which two deuterons fused to make ^4He , carrying away most of the energy by pushing off of a much heavier nucleus. Such an approach seemed to make the most sense to them when faced with a discussion of the new observations, given their point of view about what can happen in a nuclear reaction. If the alpha were produced deep within the cathode, then it would be “hidden” in the sense that it would not make it out to the cathode surface to be measured.

However, it is in general not so easy to “hide” energetic particles in PdD. It is the case that an energetic alpha has a range between microns and tens of microns in PdD, so that one could reasonably argue that it might not exit the cathode and register as an energetic particle. While slowing down, one would expect it to give rise to various kinds of secondary radiation which can make it out of the cathode and which can be observed. We consider in this work such secondary processes that lead to neutrons, x-rays, and γ -rays. *As detailed below, we have found that energetic alphas ($E > 20$ keV) are not a possibility in this context.* Therefore, little of the 24-MeV per ^4He can be carried away by this particle's kinetic energy.

This problem is interesting for a number of reasons. On one hand, there have been reported excess heat events which have occurred when neutron detectors are present (the most stringent constraints come from secondary neutron emission). In most cases, no neutrons were seen

correlated with excess heat production. As a result, we are able to develop upper limits on the number of neutrons per unit energy produced from these measurements, which can be used to develop upper limits on the initial kinetic energy of the reaction product helium nuclei. The result of this argument is that the helium nuclei must be born with a very low kinetic energy relative to the inferred reaction energy. On the other hand, we can check to see whether a proposed model is consistent with the experimental upper limits. From the results, we will see that the naive model outlined above with the “hidden” alpha particles is inconsistent with experiment because easily detected secondary radiation would be produced as the alpha particle was stopped by surrounding material.

Materials and methods

To make the case outlined above, we require theoretical estimates for the yields of neutrons, x-rays, and γ -rays assuming an energetic alpha particle is created. For the direct reactions summarized below, we have found results for cross sections in the literature. Yields were computed by integrating the cross sections over an average trajectory as determined by the stopping power from the SRIM code of Ziegler et al. (2008). In the case of secondary neutron emission resulting from collisions of alpha particles with deuterons, we started with accurate tabulated cross sections, computed the neutron yield for fast deuterons, and then we developed secondary neutron cross sections using a classical path model for alpha-deuteron collisions. We searched through more than a thousand papers in the published and unpublished literature on the Fleischmann–Pons experiment to find results we could use to develop estimates for upper limits of particle emission per unit energy. By comparing the experimental results with the theoretical results, we are able to develop upper limits on the alpha energy.

Results

We summarize results below for the yield of energetic neutrons from alpha-induced deuteron break up, and for secondary neutron emission resulting from alpha-deuteron elastic scattering; for alpha-induced x-ray emission from Pd and Pt; and for gamma emission from ^6Li resulting from alpha capture on deuterons, and for 478 keV emission from ^7Li resulting from alpha excitation. We next summarize relevant experimental results and find that the most stringent upper bounds can be obtained from measurements of neutrons from experiments where excess power was observed.

Alpha-induced deuteron break up

The alpha-deuteron break up reaction ${}^4\text{He}({}^2\text{H},\text{np}){}^4\text{He}$ has the potential to produce energetic neutrons above the alpha energy threshold at 3.34 MeV in the lab frame. That this process could be used as a diagnostic for fast alpha emission in the Fleischmann–Pons experiment was discussed previously by Takahashi et al. (1995). For this reaction, the integrated cross section has been determined in the literature at only a few energies. We have constructed an empirical cross section of the form

$$\sigma(E) = A \left(\frac{E_r - E_0}{E_r + E_0} \right)^s e^{-bE_r^2} \tag{1}$$

with

$$\begin{aligned} A &= 10.3 \text{ b} & s &= 3.17 \\ E_0 &= 2.40 \text{ MeV} & b &= 0.015 \text{ MeV}^{-2} \end{aligned}$$

where E is the alpha energy in the lab frame, and where E_r is the center of mass energy. This cross section is matched to a measurement by Kambara et al. (1978), which gives 290 ± 60 mb for an incident deuteron energy of 7.8 MeV. This model reflects the near threshold dependence for the zero-angle neutron production cross section of Henkel et al. (1955), which becomes small at energies above the threshold for break up at $E_r=2.22$ MeV. The ${}^4\text{He}({}^2\text{H},\text{np}){}^4\text{He}$ cross section is discussed in (Shanley 1969).

The yield of break up neutrons from this reaction is computed by integrating the deuteron density and cross section over the path of an alpha that slows down in PdD and in D_2O according to

$$Y(E) = \int_0^{R(E)} N_d \sigma[E(x)] dx \tag{2}$$

where N_d is the number of deuterium atoms per unit volume.

We used the alpha stopping power from the SRIM code of Ziegler et al. (2008). To compare with experimental results for the upper limit on the number of neutrons per Joule, it is useful here to present the result of this computation as yield divided by energy.

Results are shown in Fig. 1. We see that if the excess power were expressed as energetic alpha particles with an energy above about 7.5 MeV, then readily measurable fast neutron signals would be present.

Alpha-deuteron capture

It is possible for an alpha to be captured on a deuteron with the emission of a gamma in a ${}^4\text{He}(\text{d},\gamma){}^6\text{Li}$ reaction. The associated cross section has been studied by (Nolette et al.

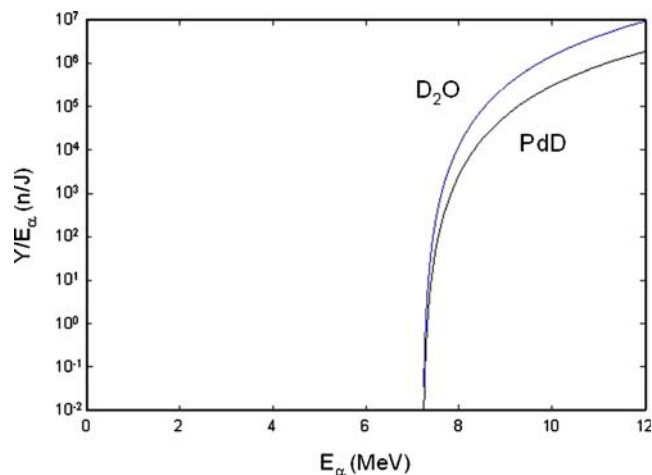


Fig. 1 Number of neutrons produced per unit alpha energy from deuteron break up reactions as a function of incident alpha energy

2001). Although the cross section is finite to much lower alpha energy than for alpha-induced deuteron break up, it is much smaller since it involves an electromagnetic interaction rather than a strong force interaction.

We show the resulting yield per unit alpha energy in Fig. 2. The gamma emission that would result from this process is potentially observable if the average alpha energy were a few megaelectron volts.

Secondary neutrons resulting from knock-on deuterons

Energetic alpha particles will collide with deuterons to produce energetic deuterons, and if these have sufficient energy, then deuteron–deuteron fusion will occur producing secondary neutron from the ${}^2\text{H}({}^2\text{H},\text{n}){}^3\text{He}$ reaction. We have computed the yield of secondary neutrons from this process using an impact parameter formulation based on classical

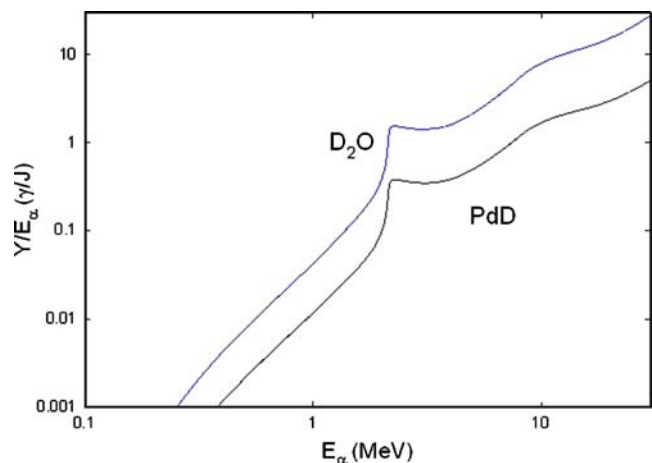


Fig. 2 Number of gammas produced per unit alpha energy from alpha capture on deuterons as a function of alpha energy

scattering trajectories. The secondary neutron cross section $\sigma(E)$ in this model is evaluated from

$$\sigma(E) = \int_0^{\infty} Y[E_d(E, b)] 2\pi b db \quad (3)$$

In this model, an alpha particle with incident energy E collides with a deuteron on a trajectory described by an impact parameter b , and the resulting scattering problem is solved. The deuteron gains energy E_d (which depends on E and b) during the collision, and given the deuteron energy, we can compute the resulting secondary neutron yield Y . By weighting the impact parameter integration by the secondary neutron yield, we can compute the secondary neutron cross section. We have used the stopping power for PdD and for D₂O from the SRIM code, and the cross section for the ${}^2\text{H}({}^2\text{H}, n){}^3\text{He}$ reaction from the LANL ENDFB-VI online data library, to compute the secondary neutron yields.

The case of D₂O is of interest since the observation of ${}^4\text{He}$ in the gas phase indicates that it originates near the cathode surface; hence, energetic particles created near the surface would have a roughly 50% probability of slowing down in the electrolyte. Our calculations refer to particles either slowing down in PdD or in D₂O. In the case of PdD, we have used a screening energy U_e of 800 eV (Raiola et al. 2004); and for D₂O, we have used 25 eV. We are able to compute a yield for the secondary neutrons using this secondary neutron cross section. Results for the secondary neutron yield expressed as neutrons per unit energy are shown in Fig. 3. The curves cross in this figure because at higher alpha energy, the yield per unit energy is higher in D₂O due to the longer range of alphas and deuterons; for alpha energies less than about 8 keV, the screening effects in PdD lead to a larger yield.

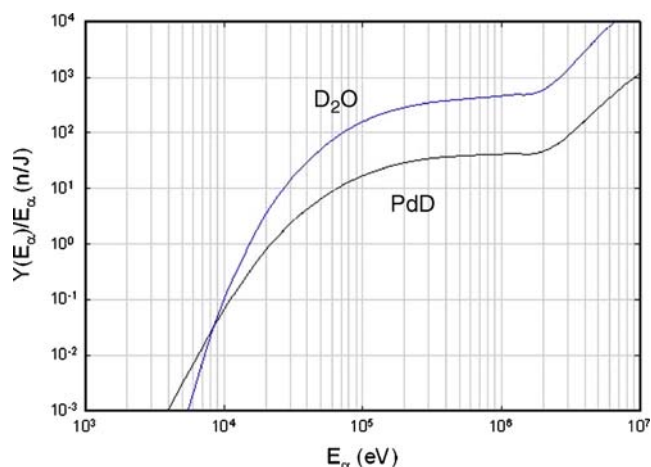


Fig. 3 Number of secondary neutrons produced per unit energy from deuterium–deuterium fusion reactions as a result of collisions between alpha particles and deuterons

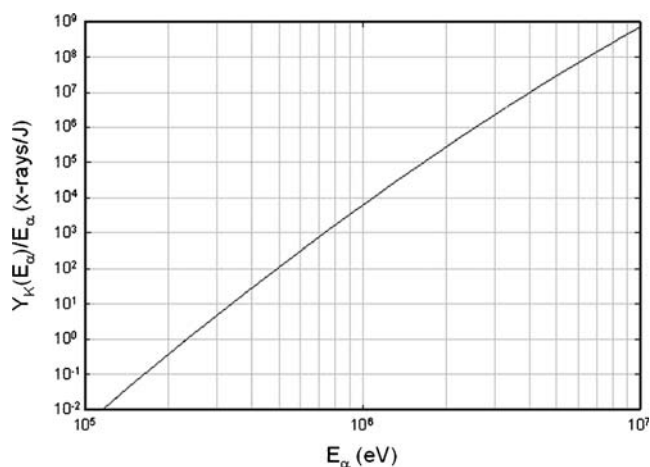


Fig. 4 Number of Pd K_{α} x-rays produced per unit alpha energy from alpha impact ionization of Pd in PdD

We see that a significant secondary neutron yield would be expected if alpha particles are born with as little as 10 keV.

Pd and Pt K-shell x-rays

Energetic alphas are known to cause K-shell x-ray emission, as has been studied both experimentally and theoretically. We have constructed an x-ray production cross section for Pd by scaling data from measurements for Rh and Ag reported by (Wilson et al. 1977). Cross section values outside of the range reported in this work were developed using scaling (Garcia 1971) of other experimental cross sections. Results for the yield divided by the alpha energy are shown in Fig. 4.

As will be discussed, Pt K-alpha x-rays have been observed in Fleischmann–Pons experiments (Pt is often used as an anode, and Pt is usually found on the outer Pd

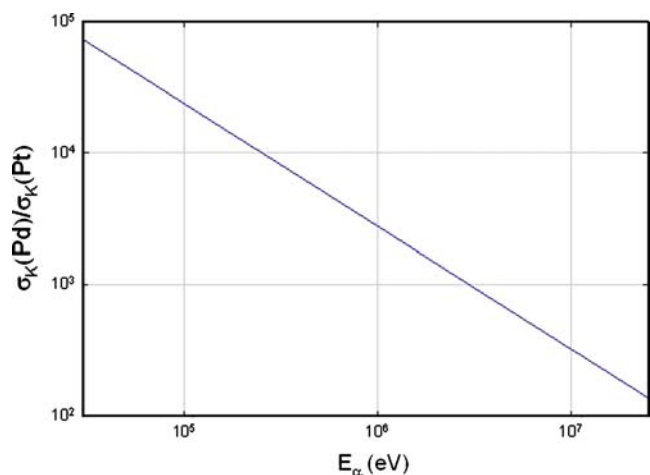


Fig. 5 Ratio of Pd K_{α} cross section to Pt K_{α} cross section as a function of alpha energy

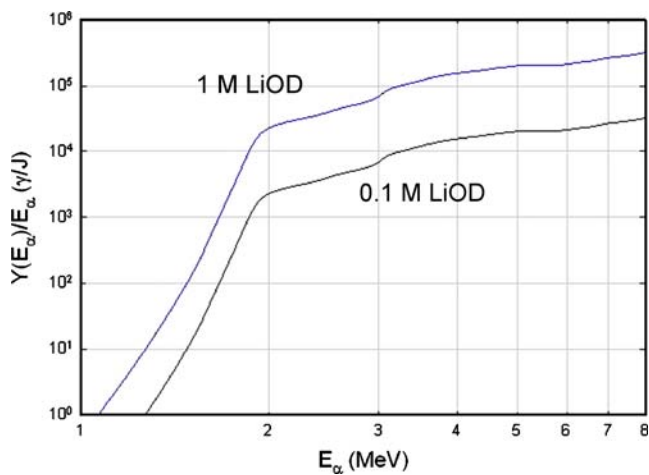


Fig. 6 Yield of ${}^7\text{Li}$ 478 keV gammas per unit energy as a function of alpha energy

cathode). Were these x-rays produced by energetic alpha particles, then one would expect more Pd K_α x-rays than Pt K_α x-rays, since the binding energy is less for Pd. We have plotted the ratio of the K-shell x-ray production cross sections for Pd relative to Pt in Fig. 5. One sees that Pd K-shell x-rays would be produced much more efficiently by energetic alphas in the megaelectron volt range.

We conclude that observable K-shell x-ray emission should be expected if the alphas are born with an energy greater than a few hundred megaelectron volts, and that there should be many more Pd K-shell x-rays than Pt K-shell x-rays.

Alpha-induced gamma emission from ${}^7\text{Li}$

The electrolyte in the Fleischmann–Pons experiment is 0.1 M LiOD, and 1 M LiOD is often used in replications. This motivates us to consider whether there are observable consequences of lithium on the surface or in the electrolyte. Fast alphas can disintegrate Li and produce neutrons; however, the associated neutron signal would be small in comparison with the neutrons produced by deuteron disintegration. Gamma emission from the first excited state of ${}^7\text{Li}$ has been studied over the years, and the associated cross section for alpha excitation (which is relatively large) has been studied. We have used the measurements of

Cusson (1965) combined with the normalization of Li and Sherr (1954), to compute the yield of the 478-keV gamma from alphas interacting with ${}^7\text{Li}$ in the electrolyte. Results are shown in Fig. 6 for 1 and 0.1 M LiOD.

Lithium is also incorporated into the outer surface of the cathode as discussed in Yamazaki et al. (1995) and Uchida et al. (1999). The contribution of gamma rays from the surface Li will depend on the location of the alpha source, but alphas which have sufficient energy to make 478 keV gammas from surface Li will give a larger signal from the Li in the electrolyte. Because the alpha capture cross section is so small, we expect that gammas from ${}^7\text{Li}$ excitation would provide a much stronger signal than gammas from ${}^6\text{Li}$ resulting from alpha capture.

We conclude that if the initial alpha energy is greater than 1–1.5 MeV, then it should produce an observable 478-keV gamma signal.

Neutron measurements during excess heat events

In the years following the announcement of the Fleischmann–Pons experiment, there were attempts to monitor for neutron emission in connection with calorimetric measurements for excess power. In a few cases, excess power was seen, while neutron detectors were operating. In Table 1, we summarize a selection of results of this kind.

In Scott et al. (1990), results are given for an open cell experiment in which excess heat was seen and no neutrons, and also for a closed cell experiment in which there was a weak correlation of excess heat and neutron emission. We have taken the numbers from the first of these to estimate an upper limit. If the weak correlation in the closed cell experiment were used to develop a neutron per unit energy number, the result would be about 0.1 n/J. The uncertainty in the neutron measurement is on the order of 10% of the background count rate. In the experiment of Klein et al. (1990), the largest average excess heat result was for an experiment where the neutron signal was not shown; we have used in this case an estimate of the uncertainty to be about 20% of the background for the other experiments where the data was given. In Wolf et al. (1990), it is mentioned that an excess heat event at the 5–15% level was seen for a Srinivasan cell. The operating conditions of this

Table 1 Summary of relevant experimental results where excess power was observed and neutron measurements were reported

Max P_{xs} (W)	Correlated neutrons?	Detector	Efficiency	Signal or background	Upper limit (n/J)	Correlated (n/J)	Reference
4	Uncorrelated	NE-213	1.46×10^{-3}	40 n/24 h	0.008		Scott et al. 1990
6	Uncorrelated	BF_3	0.01	0.25 n/s	0.8		Klein et al. 1990
0.10	Uncorrelated	NE-213	0.05	0.5 n/min	0.17		Wolf et al. 1990
100	Weak correlation	NE-213	N/A	1 n/s		0.01	Takahashi et al. 1993
15	Uncorrelated	${}^3\text{He}$	0.22	12 (35 n/10 min)	0.021		Gozzi et al. 1994

kind of cell are discussed by Appleby et al. (1990); we have taken a representative number of 100 mW which would be consistent with the discussion in the Wolf et al. paper. We have taken the uncertainty to be 10% of the background count rate for our estimate of the upper limit. In the case of Gozzi et al. (1994), the background count per tube is about 35 per 10 min; we have taken the uncertainty to be 10% of this count rate for our estimate.

There are additional experiments where excess power was observed with neutron detection operative. In experiments described in the Final Report of the National Cold Fusion Institute, excess heat was observed with no correlated neutrons; unfortunately, information as to the detection limit for the neutrons was not given. In Okamoto et al. (1994), there is a report of excess heat and neutron emission, with at most a weak correlation between the two; once again, we do not have an estimate for the source neutron emission rate in this work. In Aoki et al. (1994, 1998), excess power was observed with no detected increase in the neutron count rate; we do not have an upper limit for the neutron emission rate in this work. Finally, in Yasuda et al. (1996), experiments are described in which excess heat is seen with no neutron emission.

X-ray and γ -ray emission correlated with excess heat

There are a small number of observations of excess heat events under conditions where x-rays or gamma rays were monitored. In the experiments reported in Takahashi et al. (1995), excess heat was observed at the level of 2.5–3 W, while a CdTe x-ray detector was monitoring for x-ray emission. No x-rays were seen; the detector in this case was not very efficient, and it was estimated that about 10^8 source x-rays per second would have been required to register a signal. Using a more sensitive detector, Bush and Eagleton reported the observation of K_α x-rays from Pd, Rh, Ag, and Pt from a PdAg alloy correlated with the excess power. In this experiment, the excess power was given as 5.2 W over 64.4 h. About 1,800 Pt K_α x-rays was seen with a detector efficiency of 0.0033, resulting in an estimate of about 1 x-ray/J. In experiments reported in Iwamura et al. (1995), excess heat was observed uncorrelated with x-ray emission; a dominant Pt K_α was seen in the x-ray spectrum. We are not aware of an observation of the 478-keV ${}^7\text{Li}$ line correlated or uncorrelated with excess power in any Fleischmann–Pons experiment.

Discussion

The single most sensitive indicator for the presence of fast alpha particles in the Fleischmann–Pons experiment is secondary deuteron–deuteron fusion neutrons following

elastic scattering of deuterons by fast alphas. From the computation of the secondary neutron yield and from the experimental results of observations of neutron emission during heat bursts, we are able to estimate an upper limit for the energy of the alpha particles. It is useful to understand the limit from the graphical result in Fig. 7, where we have plotted the results extracted from experiment as points on the yield divided by energy curve. The upper limits correspond to upper limits on alpha energy in the range of 6–20 keV. The result of Takahashi et al. (1993) showing a weak correlation would imply an alpha energy of about 6.5 keV. We have also indicated in this figure how things would change if we had used different screening energies. One sees that had we used lower screening energies (which are inconsistent with experiment), which would represent a drastic change in the model, the upper limits are not changed by very much.

Note that energetic particles (including alpha particles) are observed at low levels in experiments with PdD (Lipson et al. 2000; Mosier-Boss et al. 2007). However, these emissions have not been correlated with excess power, and the amount of energy associated with these particles is lower than the energy production observed in other experiments by more than ten orders of magnitude.

There are two experiments where the Pt K_α appears to have been seen. In these experiments, the implication is that the Pt anode is not participating, and that the Pt on the surface is responsible. If so, this needs to be clarified experimentally. In the experiment of Iwamura et al. (1995), the x-ray emission is not correlated with neutron emission, which suggests that the Pt K_α is not produced as a result of fast ions. However, further observations are needed before

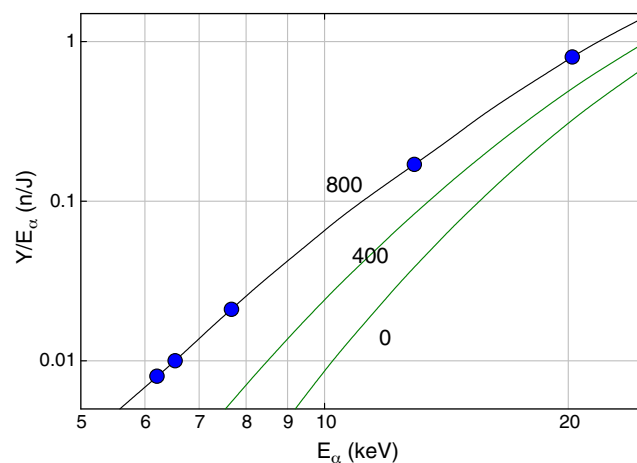


Fig. 7 Number of secondary neutrons produced per unit energy from deuteron–deuteron fusion reactions with circles indicating upper limits and correlated value for the number of neutrons per joule from experiment (see Table 1). Also shown are curves obtained with different values for the screening energy: $U_e=0$ eV (lowest curve); $U_e=400$ eV (middle curve); $U_e=800$ eV, corresponding to experiment (upper curve)

we can be sure. Future experiments in which x-ray, gamma ray, and neutron measurements are carried out in association with excess power measurements have the possibility of clarifying the mechanisms through which the low-level radiation is produced.

Conclusions

As discussed in the “Introduction”, a primary motivation of the computations presented in this work is to better understand the limitations on “hidden” energetic alphas in connection with excess power in the Fleischmann–Pons experiment. The basic conclusion is that as a reaction product, the alpha particle must be born with an energy less than 6.3 to 20.3 keV in order to be consistent with the absence of neutrons between 0.008 and 0.8 n/J as measured in Fleischmann–Pons experiments where excess heat is produced. Measurements of ^4He correlated with energy production in the Fleischmann–Pons experiment suggest that the reaction energy is 24 MeV per helium atom produced. If so, then the experimental results are consistent with the alpha particle having less than 0.1% of the reaction energy.

We are familiar with the reaction energy appearing as kinetic energy of the products in nuclear reactions, which is a consequence of energy and momentum conservation in the equivalent vacuum version of the nuclear system. Although we have not yet learned what the reaction mechanism directly from Fleischmann–Pons experiments, based on the discussion above, we can say that only a small fraction of the reaction energy can be present in the alpha particle at the end of the reaction. Efforts to account for excess energy in the Fleischmann–Pons experiment based on models that involve energetic particles are unlikely to be successful in light of the upper limits discussed here.

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