ON ASPECTS OF NUCLEAR PRODUCTS

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ABSTRACT

Reaction product measurements are not yet conclusive and often appear contradictory. Still the measurement of tritium in several different laboratories appears most convincing. However, there is growing evidence that the tritium production rate can only account for a few percent of the heating rate while the neutron rate is only 10^{-5} to 10^{-8} times the tritium rate.

Various mechanisms proposed to explain these observations are reviewed. The often favored candidate for tritium production, $D + D \rightarrow (^4He)^* \rightarrow T + p + 3.7$ MeV, with the excess energy carried by phonons or electrons, would not explain heat production rate that would require $(^4He)^*$ decay by collective dissipation of 23.8 MeV. Thus it appears likely that several different reactions are involved. The need for improved diagnostics to provide a time correlation for the appearance of products and to measure X rays, gamma rays and energetic charged particles is stressed. The significance of the search for transmutation of Pd and Li isotopes on electrode surfaces is also discussed.

INTRODUCTION

The key objective for measurements of nuclear products is to identify the reaction mechanism (or mechanisms) that are occurring in the current cold fusion experiments. In addition to this identification, an understanding of the nuclear products is essential for the determination of future applications of cold fusion. For example, are we developing small radiation free cells for heat and electrical production or does this device better lend itself to tritium production?

The degree of correspondence between experimental measurements and theory is generally viewed as a gauge of the maturity of any new scientific endeavor. In that sense it appears that, as of today, cold fusion is not very mature; certainly when compared to hot fusion. However, cold fusion has an important advantage in that experiments can be done relatively cheaply in small cells in a variety of laboratories. Consequently, a wide data base can develop rapidly. Thus, it seems reasonable to expect that the field will quickly reach the point where measurements and theory converge to provide a clear understanding of the phenomena involved.

COMMENTS ABOUT REACTION PRODUCT MEASUREMENTS

As shown in Fig. 1, measurements in any experiment should cover a wide variety of possible reaction products[1]. These range from neutrons, gamma rays, X rays and neutrinos on to energetic charged products and isotopic abundances of electrode and electrolytic materials. Also, the importance of continuing the search for helium-4 and helium-3 in both the off gas and electrodes is widely recognized. In addition to measurement of specific products, it is essential that time correlations between various particle and radiation products and also heat production be established. As seen from the present meeting, several attempts to do this have been made[2]. However, due to complications associated with the "burst nature" of the phenomena, time correlations have been difficult to obtain. Not only are time correlations an essential element in the effort to identify the mechanism involved, but they should help resolve the issue of whether different mechanisms occur at different times in a given experiment or in uniquely

Neutrons

Gammas, x-rays, neutrinos

Charged Products

protons, ^4He, ^3He, tritium, betas (β^+ and β^-)

Transmuted Pd isotopes, Li isotopes, O isotopes

Figure 1. Possible Reaction Products from Experiments. Note: Different mechanisms may occur in various experiments, resulting in a different grouping of products.

Tritium/neutron rates ~ ratio of 10^5 to 10^8

Tritium Rx heat rate/heating rate < 5%

Burst character

neutron bursts { sec to 10s min duration; up to 100 n./50 μs bursts }

heat bursts — long term "bursts," delayed initiation

Surface enrichment of ^{106}Pd ? ^7Li

appearance of Rh, Ag, Cu ?

3-MeV protons (bursts) ?

Autoradiograph: active surface "spots," K X rays

Neutron "avalanche," > 10^8 n/s, in "on-off" exps.

Gamma-rays ?

14-MeV neutrons doubtful

Figure 2. Characteristics observed in various experiments.

different experiments. Specifically, a key current issue is whether or not tritium production is correlated with heat and neutron production.

GENERAL OBSERVATIONS

At present it is difficult to provide firm conclusions about reaction products based on experimental data due to the non-reproducible nature and the intermittent bursts of heat and neutrons encountered in most cells. However, a rough impression is summarized in Fig. 2.

Most important, a pattern appears to be emerging where the tritium to neutron production rates occur in a ratio of 10^5 to 10^8[3,4]. Further, the heating rate that would accompany tritium production appears to be less than 5% of the overall heating rate. Indeed, not all experiments have demonstrated both tritium production and heating, so there may not even be a connection between the two. (For example, experiments at Oak Ridge National Laboratory reported at this meeting[5] were cited for excess heat but no measurable tritium. The Los Alamos National Laboratory experiments cited for tritium production[6,7] have not been instrumented for simultaneous heat measurements.) Still, the ratios cited here may be viewed as a "time average" over various experiments.

In addition to tritium production, a variety of other miscellaneous characteristics listed in Fig. 2 have been noted in various experiments. These include, both neutron and heat bursts[4,8]. Also the possible redistribution in surface layers of palladium and lithium isotopes as well as trace impurities in the electrodes has been observed in some instances[9,10]. MeV proton bursts have been reported in several instances. Further, the spotty nature of X-ray and/or gamma-ray production has been demonstrated in autioradiographic measurements[3]. Other phenomena include the observation of massive neutron avalanches in "on-off" experiments and occasional reports of gamma emission in other cells[5,11]. It is important to note that, to date, no experiment hasreported 14-MeV neutron production. Neither has there been a measurement reported which would verify ^4He or ^3He production -- the measurements attempted being inconclusive[12].

Since many of these observations occur on different cells and are not always reproducible, an interpretation is difficult. For example, the segregation of trace impurities and enrichment of some isotopes on electrode surfaces observed in several experiments appears to be associated with electrically driven transport processes rather than a nuclear reaction. However, some of these phenomena may enable the reaction, or alternatively quench it. More work needs to be done to clarify these issues. Thus for present purposes, we will mainly concentrate on tritium and heat reactions since data for these phenomena has been most widely reported.

When searching for reaction mechanisms, it is important to keep in mind that there are two key issues that require resolution. The first is a quantitative explanation of the reaction rates, i.e., the tritium production rate and also the heating rate in the cell. The second issue though, is the relative production of the reaction products, i.e., to the branching ratio between reaction "channels."

TRITIUM PRODUCTION MECHANISMS

As seen from the list of possible two-body reactions, given in Table 1 (taken from Ref. 13), very few reactions can lead to tritium production. The D-D reaction itself is the most obvious candidate. If this is not the source of tritium, then we must jump to other reactions such as:

$$^{2}_{1}D + ^{7}_{3}Li \rightarrow ^{3}_{1}T + ^{6}_{3}Li, \text{ or:}$$

$$^{2}_{1}D + ^{17}_{8}O \rightarrow ^{3}_{1}T + ^{16}_{8}O$$

which, due to smaller abundances of the Li-7 and O-17 isotopes and smaller classical reaction cross sections, seem less likely. While the possibility of tritium production by a multibody mechanism cannot be ruled out, there does not appear to be a reasonable candidate. Thus we conclude here that the D-D reaction is the most probable source of tritium. This conclusion must be viewed in light of additional observations summarized in Fig. 3 that are connected with tritium production. The many orders of magnitude higher tritium production rate vs. neutron rate suggests that the D-D reaction must proceed through the t + p channel rather than the He^3 + n branch. Also, if the absence of 14-MeV neutrons is confirmed, it appears that the tritium must be born at abnormally low energies. In the "conventional" D-D reaction, corresponding to Fig. 3, tritium would be released at 3 MeV. Then, as it slows down in the dense deuterium medium, D-T reactions would occur providing a measurable 14 MeV neutron signal (see, for example, Ref. 14). The failure to observe 14-MeV neutrons is not completely conclusive, however. Such measurements are difficult in view of the erratic behavior of cells. Also the relatively low yields make detection difficult. Thus continued stress should be placed on a search for possible 14-MeV neutrons. Another issue which should be considered is that anomalous slowing of the fast tritium due to collective effects could result in a very low yield of 14-MeV neutrons. At the moment, however we will simply assume that no neutrons are observed and that a classical Fokker-Planck behavior of tritium slowing-down occurs. In that case, cf. Fig. 3, there are limited possible explanations for tritium production. One popular explanation for the strong tritium channel (originally advocated by the authors), is an Oppenheimer-Phillips type D-D reaction. However, this reaction would still proceed with the production of 3-MeV tritium so the 14-MeV neutron argument seems to rule it out. This leaves the possibility of "deuteron tunneling" as proposed earlier by G. Collins[15] and discussed in detail by J. Schwinger[16] at this conference. As indicated in Fig. 3, this reaction produces tritium at 0.08 MeV with the transfer of 3.97 MeV of energy to the surroundings (see the energy level diagram of Fig. 4). G. Collins originally proposed that this excess energy was carried off by electrons while J. Schwinger's recent theory is based on transfer to phonons. In either case, the reaction products are identical. Thus an identification of the exact carrier of the excess energy is not crucial to the present discussion. Further, it should be stressed that in either theory the energy released per triton formed is also identical. Thus the heating rate associated with tritium production is unambiguous, and this is important in any comparison of the tritium production rate to the cell heating rate.

While tritium might be produced via various reactions with lithium, experiments appear to make this possibility unlikely. For example, lithium isotope substitution experiments have not confirmed the need for a specific isotope in order to produce tritium[17]. Substitution of the sodium equivalent for

TABLE I

Possible Two-Body Reactions, Their Products, and Q Values

Reactants	Product 1	Product 2	Product 3	Q Value (MeV)
$p + d$	^{3}He	γ		5.4
$d + d$	t ($\beta t_{1/2} = 12.3$ yr)	p		4.03
	^{3}He	n		3.27
$p + {}^{6}$Li	^{7}Be ($\epsilon t_{1/2} = 53.3$ days)	γ		5.61
	^{3}He	α		4.02
$p + {}^{7}$Li	^{8}Be ($2\alpha t_{1/2} = 0.07$ fs)	γ		17.25
	α	α		17.35
$d + {}^{6}$Li	^{8}Be ($2\alpha t_{1/2} = 0.07$ fs)	γ		22.28
	α	α		22.37
	^{5}He (α, $n\Gamma = 0.6$)	^{3}He		0.84
	^{3}He	α	n	1.80
	^{7}Li	p		5.03
	^{7}Be ($\epsilon t_{1/2} = 53.3$ days)	n		3.38
	^{5}Li (α, $p\Gamma = 1.5$)	t ($\beta t_{1/2} = 12.3$ yr)		0.60
$d + {}^{7}$Li	^{9}Be	γ		5.61
	^{8}Be ($2\alpha t_{1/2} = 0.07$ fs)	n		15.03
	^{5}He (α, $n\Gamma = 0.6$)	α		15.16
	α	α	n	15.12
$p + {}^{12}$C	^{13}N ($\epsilon t_{1/2} = 9.96$ min)	γ		1.94
$d + {}^{12}$C	^{14}N	γ		10.27
$p + {}^{16}$O	^{17}F ($\epsilon t_{1/2} = 64.5$ s)	γ		0.6
$d + {}^{16}$O	^{18}F ($\epsilon t_{1/2} = 110$ min)	γ		7.53
$p + {}^{102}$Pd	^{103}Ag ($\epsilon t_{1/2} = 1.10$ h)	γ		4.24
	m (IT$t_{1/2} = 5.7$ s)			4.11
$p + {}^{104}$Pd	^{105}Ag ($\epsilon t_{1/2} = 41.3$)	γ		5.01
	m (ϵ, IT$t_{1/2} = 7.2$ min)			4.97
	^{101}Rh ($\epsilon t_{1/2} = 3.3$ yr)	α		2.85
	m (ϵ, IT$t_{1/2} = 4.34$ days)			2.69
(Similar reactions in ^{106}Pd, ^{108}Pd, ^{110}Pd: see Ref. 1)				
$d + {}^{102}$Pd	^{103}Ag ($\epsilon t_{1/2} = 1.1$ h)	n		2.01
	m (IT$t_{1/2} = 5.7$ s)			1.88
	^{103}Pd ($\epsilon t_{1/2} = 17.0$ days)	p		5.38
(Similar reactions in ^{104}Pd, ^{105}Pd, ^{106}Pd, ^{108}Pd and ^{110}Pd: see Ref. 1)				

Nomenclature

d = deuteron

IT = isomeric transition (gamma-ray and conversion-electron decay)

m = isomeric state

n = neutron

p = proton

t = triton

$t_{1/2}$ = half-life

α = alpha decay

β = negative beta decay

ϵ = positive beta decay including electron conversion

Γ = level width for particle-unstable nuclides (MeV)

Tritium

T/n rates ~ 10^{5-8}

no 14-MeV neutrons ?

$$T + D \rightarrow \alpha + n + 17.6\ MeV$$
$$\uparrow$$
$$D + D \longrightarrow T + p + 4.02\ MeV$$
$$D + D \searrow {}^{3}He + n + 3.27\ MeV$$

Low energy T ? Possible Explanations

- D + D Oppenheimer-Phillips Reaction - predicts high energy T however.
- D or n tunneling (Δ carried by electrons or phonons)

$$D + D \rightarrow {}^{4}He\ (20\ MeV) + \Delta(3.7\ MeV)$$
$$\downarrow$$
$$T(.08\ MeV) + p(0.23\ MeV)$$

- D + Li reaction ? (Inconsistent with tritium reported in non Li containing cells)

$^{6}Li(n,\alpha)T$ $\quad$ $^{6}Li(d,\alpha,p)T$

$^{7}Li(n\alpha,n)T$ $\quad$ $d(^{6}Li,\alpha,p)T$

- D + O reaction ? $^{2}_{1}D + ^{17}_{8}O \rightarrow ^{3}_{1}T + ^{16}_{8}O$

Other mechanism issues:

Surface vs Volume

Local fields via surface projections vs. Surface Potential

} crucial to scaling relations

How to account for the heating observed? Second reaction independent of tritium producing reaction needed?

Figure 3. Observations connected with tritium production.

the lithium electrolyte did, however, reduce excess heat production[17]. Thus the role of the electrolyte is not clear. Further, there are some reports of tritium in cells that do not contain lithium at all. (An example is T. Claytor's experiment at Los Alamos National Laboratory[18].) In summary then, the D-D reaction associated with n/D tunneling shown in Figures 3 and 4 must be viewed as a possible explanation.

A number of associated issues also need resolution. The most obvious is the location of the reaction, i.e, whether it occurs only on the surface versus in the volume of the electrode. The surface is favored in several theories, e.g., deuteron acceleration by local fields created by dendrites or by a double potential layer at the surface[2,19]. Indeed, thus far, some experimental data appears to favor a surface reaction, but there are contradictions and the data is ambiguous.

Perhaps the most important issue is whether the tritium reaction can also account for the observations of excess heat; i.e., is there a second independent heat producing reaction? Indeed, the latter seems likely since the energy release in the tritium reactions of Figs. 3 and 4 does not account for more than 5% of the heating reported in strong

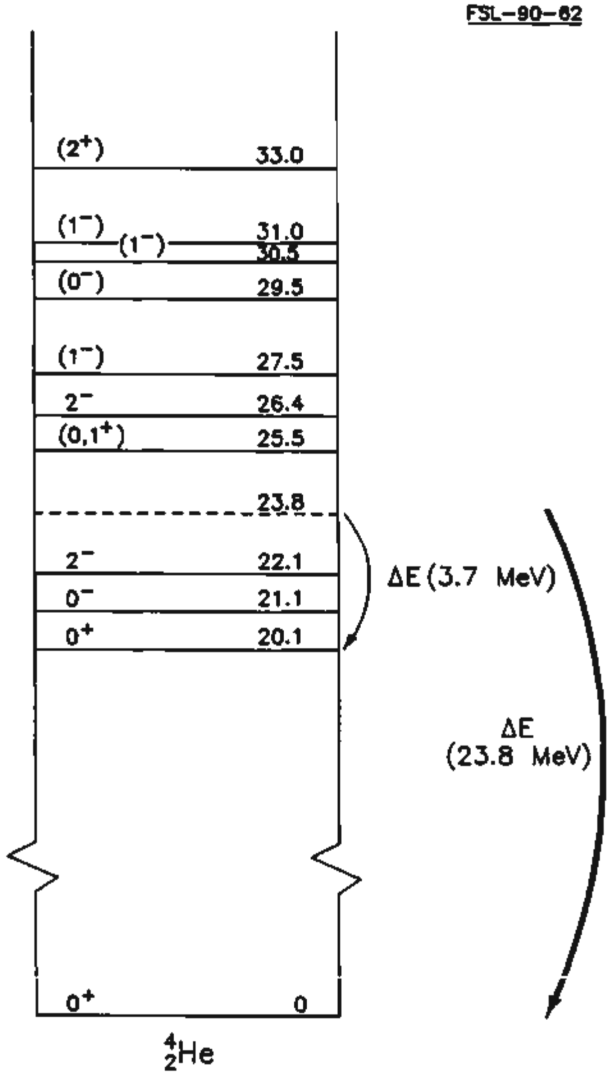

Figure 4. Energy Level Diagram for the ^4He Compound Nucleus.

"heating" experiments[2,3]. Although the lack of correlated measurements admittedly clouds this issue, we will choose to treat heat production as a separate issue in the present discussion.

HEAT PRODUCTION REACTIONS

The most crucial, but to date, the least understood mechanism associated with cold fusion experiments is the observation of excess heat. The observation of tens of megajoules energy release in various experiments seems to rule out all possible sources except nuclear reactions[2,20].

Characteristics observed in calorimetric experiments of heat production are summarized in Fig. 5. As stressed earlier, the ratio of heat production to the heating rate associated with tritium production appears to be a factor of 20 to 100. In addition, the heat production must be quite aneutronic since the tritium reaction itself is so much stronger than the neutron rate.

Various reaction mechanisms which have been discussed in connection with heating are also outlined in Fig. 5. The "deuteron tunneling" reaction in deuterium is quite similar to one discussed earlier in connection with tritium production. However, now, as shown earlier from the energy level diagram in Fig. 4, 23.8 MeV must be dissipated (as opposed to 3.7 MeV[1]). Initial theoretical studies of cold fusion proposed several mechanisms for dissipation of this large amount of energy[21,22], but the present consensus is that this is unlikely. The point is that the lifetime of the ^4He compound nucleus is extremely short[13] so that a collective dissipation of the 23.8 MeV's would have to occur to distribute order of an eV/photon (or multi-eV/electron) quickly enough to prevent decay to other products, i.e., to avoid the "classical" D-D reaction.

Another reaction that has been considered is D-^6Li with the release of two alpha particles carrying a total of 22.4 MeV[30]. In addition to the appearance of high energy alphas, this reaction should produce copious X rays. A search for such emission is just starting. A dependence on ^6Li should also be apparent in the cells, but this has not been confirmed yet, and, indeed there is some evidence to the contrary.

Another mechanism which has been considered in several different forms involves a chain reaction carried by neutrons[23,24]. The "closing" of the chain typically assumes a gamma interaction with deuterium which reproduces the chain carrying neutron. However, calculations of the relative reaction rates do not appear to justify this closure, i.e., it appears that the chain would be very short. Thus, this mechanism does not seem likely.

A third possibility which has been discussed by the present authors is an Oppenheimer-Phillips reaction between the deuterium and the palladium itself[1,25]. (In this case, an Oppenheimer-Phillips reaction involving D-D would occur

Heating Observations

- $\dot{H} / \dot{T} \sim 20 - 100$ x ?
- aneutronic

Reaction Possibilities

- D + D → + ^{4}He + 23.8 MeV ?
- D + ^{6}Li → 2^{4}He + 22.4 MeV ?
- neutron chain chain rate ?
- O-P

  D + ^{A}Pd → $^{A+1}$Pd + p ~ 4.8 MeV

  Pd isotope shift ??
- n-D tunneling

  ^{6}Li + ^{6}Li → ^{7}Li + p + α + 3.6 MeV

  ^{6}Li + ^{6}Li → 2α + t + p + 1.1 MeV
- Coherent
  Cluster (Nattoh Model)
  Bose Block Condensate (BBC)

  Various products } ?
  MeV particles/alphas X rays }
- X^- catalysis

  Xd + α → X^- + ^{6}Li + 1.48 MeV } ?
  (tritium production: ??) → simulated tritium: Xp }

Figure 5. Characteristics observed in calorimetric experiments.

simultaneously, producing tritium and suppressing the neutron branch. However, as discussed earlier and also in Ref. 1, this view would require rationalization of the relative D-Pd and D-D reaction rates as well as an explanation of the lack of observed 14-MeV neutrons from the energetic tritium.) Again there is considerable difficulty in justifying the reaction rate that would be needed to match the observed heating. However, this reaction would be a prime candidate if palladium isotope transmutations are found[1,25]. Indeed, as indicated by D. Rolison et al.[9] at this meeting, palladium isotopic shifts may have been observed in one instance, but this result has not been reproduced. Further, no indication of such a shift has been found in several cases where surface measurements of electrodes have been carried out in other laboratories[10]. Unless experimental confirmation of an isotope shift is found, this mechanism must be ruled out.*

A fourth possible mechanism is the counterpart of the deuteron tunneling mechanism proposed by Collins[15] as applied to lithium. For example, lithium-6 reactions would yield lithium-7 plus a proton and alpha particle. Something of this nature might be inferred from some data. For example, lithium isotopic shift

*Note, however, that as discussed in Ref. 1, transmutations by the Oppenheimer-Phillips reaction will occur in different ratios than those due to "free" neutrons. The relative shifts in the former will be less, so a very careful examination of relative isotopic abundances is essential to evaluate this possibility.

on electrode surfaces noted by several workers at the present meeting[10] seems suggestive of such a reaction. However, again there is no explanation for the reaction rate that would be required to provide the heating rate reported in various experiments. Also this explanation would not be consistent with experiments where lithium-6 was not present when heating was obtained[2,17]. Thus this mechanism also seems unlikely.

Another category of mechanisms includes a variety of multibody reactions with energy being carried off by coherent effects. These include the general theory of coherent reactions[16,26,27], the multibody cluster ("Nattoh") mechanism[28], Bose-Block Condensate[29], the transmission resonance concept[30], etc. These various mechanisms have not been explored in depth yet and require more study to fully evaluate, however. Reaction products have not been clearly identified in all of the proposed mechanisms either, although helium production is a common product which could be detected. Also, these phenomena generally result in MeV charged particles with accompanying gamma-ray and/or X-ray emission. Thus experimental studies of these radiations are crucial to the evaluation of possible mechanisms.

Another hypothesis revolves around the issue that a strange particle, namely the X^- particle, could catalyze cold fusion[31]. The X^- particle is postulated as a remnant from the early formation of the universe. If it exists, the bombardment of the earth by a flux of Xp could result in a finite concentration of Xd in heavy water. Thus, in the cold fusion cell, energy could be released by the interaction of Xd with alpha particles contained in other nuclei, freeing the X^- and yielding Li^6 and heat. The X^- particle would continue to interact in a chain-like reaction. This theory might even provide an explanation for tritium observations since an Xp particle would have an electronic structure capable of simulating tritium emission. While such a phenomena cannot be ruled out, much more effort would be necessary to provide convincing evidence, especially since the X^- has not previously been observed.

An alternate theory involves the formation of a new neutral particle, designated as Neutrium (Nu) which consists of two bound neutrons [32]. The Nu can be absorbed by deuterium, Pd or other heavy nuclei, producing a source of heat. Nu, alternately called the dineutron, was originally studied with respect to the T-T reaction

$$ {}^{3}_{1}T + {}^{3}_{1}T \rightarrow 2\,{}^{1}_{0}n + {}^{4}_{2}He \quad . $$

In conclusion then, the search for the heat producing reaction in cold fusion remains as the greatest mystery of cold fusion. Of the various possibilities discussed here, perhaps the D-D reaction with 23.8 MeV coherent dissipation remains as the most plausible but has no complete theoretical justification. To solidify this position, we must find a theory for rapid dissipation of this huge amount of energy and intensify the search for He^4 production. On the other hand, if the report of an isotopic shift of either palladium or lithium is confirmed, obviously a strong focus would develop on the reactions involving these species.

The measurement of charged particles and of gammas should provide a strong insight into the various multibody reaction possibilities. The observation of high-energy charged particles, in particular, would provide a strong clue. Indeed, several new experiments aimed at charged particle detection are being developed (see, for example, Ref. 33). These include plans to incorporate solid-state detectors into special design cells. Another approach which has started to receive some emphasis is the possible use of CR-39 plastic foil for charged particle tracking[34,35]. The advantage of such foils is their low cost and the relative ease of inserting them into the experiment.

As stressed by several investigators, any mechanism which is proposed must be consistent with the frequently observed burst behavior and long wait prior to initiation which is observed in many experiments. The explanation for these characteristics must be an inherent feature of the mechanism. An example is the high sensitivity of the tritium production rate to key parameters in the exponential functions in Schwinger's theory[16]. On the other hand, this "delay" could be due to other physical conditions that must be brought together in order to enable the mechanism to proceed. For example, a special surface composition might be required. Along that line, some comment is worthwhile about the difference between microscopic hot fusion and cold fusion. In some theories, such as crack propagation, the electric field accelerates deuterium causing reactions by a high energy particle interacting with the cold background (known in the hot fusion community as beam-target interactions). This might help explain the reaction rate but does not explain how the branching occurs. A complete theory must resolve both issues. Indeed, attempts are now being made to incorporate aspects of both reaction rate and branching phenomena in a unified theory (e.g., see Rice, et al.[36]).

SUMMARY

In summary, there is not a consistent picture as to the mechanism and the associated reaction products. However, with the rapid rate of advance and accumulation of experimental data there is every hope that the situation should be cleared up in the near future. One difficulty revolves around the danger that mistakes will be made due to the possible existence of multiple mechanisms which might occur simultaneously during a given experiment or at different times in different experiments.

ı.. level of sophistication of diagnostics of cold fusion has increased dramatically in the past year. There is strong hope that this will help in the resolution of the reaction mechanism. Of course, to fully understand the cell operation, one needs a full array of diagnostics capable of covering a variety of particle and radiation as indicated earlier in Fig. 1. This must be combined with the capability for extensive surface and volume metallurgy. Finally, all these diagnostic techniques must be combined with good control of the experimental conditions in the cell.

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