

OCT 25 1989

Professor Hendrik J. Monkhorst
Department of Physics
362 Williamson Hall
University of Florida
Gainesville, FL 32611

Dear Henk:

Just a line to acknowledge, with thanks, the receipt of the Anghaie/Froelich/Monkhorst manuscript. You may be interested to know that Professor Kim of Purdue University has been thinking along lines similar to yours; you may wish to send him your preprint.

I did appreciate the handwritten note you attached to the manuscript. It was gracious of you to say what you said.

Best regards.

Sincerely,

Original signed by:
Ryszard Gajewski

Ryszard Gajewski, Director
Division of Advanced Energy Projects
Office of Basic Energy Sciences, ER-16

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*Yeong E. Kim
Department of Physics
Purdue University
West Lafayette, IN 47907

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**On Fusion/Fission Chain Reactions
in Fleischmann-Pons "Cold Fusion" Experiment.**

S. Anghaie*, P. Froelich**, H.J. Monkhorst***

*Department of Nuclear Engineering Sciences, University of Florida, Gainesville, Florida 32611

**Quantum Theory Project, University of Florida, Gainesville, Florida 32611

and Department of Quantum Chemistry, Uppsala University, 75720 Uppsala, Sweden

***Department of Physics and Chemistry, University of Florida, Gainesville, Florida 32611

Abstract

The possibility of fusion/fission chain reactions following $d - d$ source reactions in electrochemical "cold fusion" experiments has been investigated. We have estimated the recycling factors for the charged particles in fusion reactions with consumable nuclei d , ${}^6\text{Li}$ and ${}^7\text{Li}$. It is concluded that, based on the established nuclear fusion cross sections and electronic stopping power, the recycling factor is four to five orders of magnitude less than required for close to critical conditions. It is argued that the cross generation of charged particles by neutrons does not play a significant role in this process, even if increased densities at the surface of electrodes do occur.

I. Introduction

Ever since the announcements of alleged evidences for $d - d$ fusion events under electrochemical conditions by Fleischmann and Pons (FP) /1/, and Jones *et al* (J) /2/ an explosion of experimental and theoretical activities occurred. Many experiments (all but a few unpublished) have confirmed at best only part of the FP and J observations, and most have not found any evidence of electrochemical fusion. Theoretical efforts /3/, /4/ have exclusively focussed on finding mechanisms to enhance individual $d - d$ fusion rates. Following the original suggestions by FP and J all attempts dealt with fusion inside the Pd cathode. Scientists concentrated on the possibility of the $d - d$ reaction via quantum mechanical tunneling induced by the "chemical confinement" /5/. It was quickly shown that the mere proximity of metal-infused deuterons is by far insufficient and can not lead to the fusion rates far beyond those typical for D_2 molecules. Invariably the calculated fusion rates fall short of the experimentally claimed values by many orders of magnitude. For example, the most thoughtful analysis by Leggett and Baym /6/ arrives at a upper limit to the $d - d$ fusion rate inside the solid of

$$\lambda_{f,d-d}^{bulk} \sim 10^{-47} /sec^{-1} /d - d \text{ pair} \quad (1)$$

This is to be compared with the experimental values claimed by FP and J

$$\lambda_{f,d-d}^{FP} \sim 10^{-19} /sec^{-1} /d - d \text{ pair} \quad (2)$$

$$\lambda_{f,d-d}^J \sim 10^{-23} /sec^{-1} /d - d \text{ pair} \quad (3)$$

as suggested by neutron and triton productions.

Only unwarranted assumptions about electron effective mass, screening Coulomb interactions, exotic reactions, coherent solid state effects or d concentrations inside Pd have been used by some workers to bring the fusion rate anywhere near the observed

values. The most startling finding by FP is the generation of excess heats of several Watts per cm^3 of Pd cathode. This would require a fusion rate enhancement with yet another factor 10^{10} as compared to Eqs. (2) and (3). Because of that FP ruled out any chemical reactions as a source for this heat. Needless to say that theorists were particularly at a loss explaining its origin, and are confronted with a mismatch between experimental and theoretical $d - d$ fusion rates approaching *forty* orders of magnitude, an uncomfortable discrepancy indeed.

Yet, experiments are continuing, and we are aware of several substantial (albeit not complete) confirmations of the FP and J work. The most telling results are those by one Texas A&M group (TAM) that found tritium production from D_2O electrolysis, under FP conditions, at rates even exceeding their rates (Eq. 2). TAM obtained maximum T activity of 10^6 dpm/ml after 12 hours, corresponding to roughly

$$\lambda_{f,d-d}^{TAM} \sim 10^{-13} / \text{sec}^{-1} / d - d \text{ pair} \quad (4)$$

when expressed as an equivalent $d - d$ fusion rate in the bulk of Pd , significantly higher than Eq. (2).

It appeared plausible that nuclear reactions producing tritium take place, even if no neutrons or excess heat are observed, and that conditions exist in the electrochemical process enormously enhancing the apparent $d-d$ pair fusion rate not explicable in "normal" bulk fusion terms. One was tempted to search for radical alternative explanations.

Below we investigate the possibility of nuclear fusion/fission chain reactions in the surface layer involving d , 6Li and 7Li as consumables; p , t n and 3He as intermediary particles, and 4He (and Be) as products. Some experimental facts seem to suggest that

(i) the nuclear events occur at the cathode surface, rather than in the bulk

(ii) the surface layer surrounding the Pd cathode could have enormous OD^- , D , D_2O and Li^+ concentrations. As explained below, this follows from the current densities reaching $1A/\text{cm}^2$

It has been speculated that these high densities could help to drive the chain reaction process, especially if also neutrons were involved.

In the following section we give largely phenomenological arguments from electrochemistry in support of high surface layer densities, far exceeding those in the bulk electrolyte.

In Section III, the concept of self-sustained chain reaction is introduced, and the most probable fusion/fission reactions which might lead to recycling of energetic charged particles are explored. In Section IV the associated fusion reaction probabilities and the overall recycling factor are calculated.

II. Phenomenology of Electrochemical Transport Situations

Consider the fact that current densities of the order of magnitude $0.1 - 1 \text{ A/cm}^2$ have been used /1/, /2/. Let us consider the upper limit and assume the cathode reactions



to be essentially responsible for the hydrogen evolution and OD^- ion production, and to happen at the cathode surface. It then follows that a flux of OD^- and D outward, and D_2O inward bound of

$$\sigma_d \sim 6.8 \cdot 10^{18} / \text{cm}^2 \cdot \text{sec} \quad (6)$$

is present on surfaces surrounding the electrode surface at distances small compared to the electrode dimensions. It is well known that at layer thicknesses l of the order of 1 to 10 μm away from the electrodes the voltage drops to very small values causing the motion of OD^- towards the anode to be diffusion determined. Typical diffusion drift velocities are $v_d \sim 10^{-5} \text{ cm/sec}$. This then gives an average deuteron density in the surface layer

$$\bar{n}_d = \frac{\sigma_d}{v_d} \sim 10^{24} / \text{cm}^3 \quad (7)$$

some two orders of magnitude higher than in the bulk electrolyte! Since the surface layer attempts to be neutral in the stationary state we conclude that the Li^+ concentrations are

comparable:

$$\bar{n}_{Li^+} = \bar{n}_d \sim 10^{24}/\text{cm}^3 \quad (8)$$

Actual local densities within the surface layer can be higher, since

$$\bar{n} = \frac{1}{l} \int_0^l n(x) dx \quad (9)$$

with x the distance into the layer, and

$$n(l) \sim n(\text{bulk}) \quad (10)$$

However, since we expect something like an exponential profile to $n(x)$, \bar{n} must be close to the highest local density. There are a number of oversimplifications in this picture: (i) electron tunneling to a few D_2O molecular layers for reaction (5) might occur; (ii) not the entire surface area of the Pd cathode can be considered active for conduction. As the electrolysis proceeds the surface becomes microscopically highly irregular, developing cracks, dendrites, sharp edges, etc.. This causes the σ_d , and the associated \bar{n}_d to locally exceed the macroscopic values of Eqs. (6) and (7) by several orders of magnitude /7/.

The resulting densities in the order of $\bar{n} \sim 10^{24}$ to $10^{26}/\text{cm}^3$ are truly astronomical /1/, the upper limit corresponding to that of stellar interiors. These numbers are large, but not unthinkable as they are result of an intrinsically non-equilibrium situation.

One is driven to the conclusion that enormous densities of D , Li^+ and OD^- could occur locally in the surface layer. The details of motion, distribution and relative concentrations of these particles are probably unknowable, but they should not matter for the nuclear physics we will discuss next.

III. Nuclear Processes

We explore the possibility that the results of F and P /1 / and some subsequent experiments can be explained by considering a *chain of nuclear reactions*, which could

proceed as a self-sustained process in a regenerative system. Such a system contains fusible material that can produce additional fast particles as a result of fusion reactions. A certain fraction of those fast particles will cause fusion reactions before being slowed down appreciably (i.e. below the threshold for nuclear reactions).

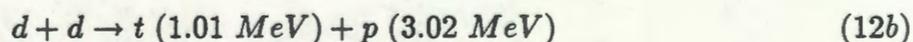
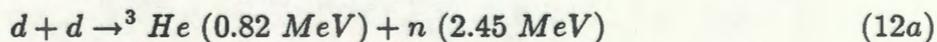
The chain of reactions could be triggered by any weakly-probable fusion process, as for instance the $d-d$ reaction, and could then proceed with help of reactions involving ${}^6\text{Li}$ or ${}^7\text{Li}$ acting as a source of ${}^3\text{He}$ and t . In a way, such a process should not be classified as cold fusion, since the nuclear reactions occur at the high energies characteristic for the fusion products being created and slowed down in the vicinity of the electrode's surface. Under suitable conditions, discussed below, such a reaction chain could lead to the so called *critical system* /8/. The important feature of such a system is that its life-time and efficiency does not depend on the absolute fusion rate, but rather on the *multiplication factor*, being a ratio of production rate λ_p to the disappearance rate λ_d for those fusion or fission products which enter the cycle of regenerative reactions

$$R = \frac{\lambda_p}{(\lambda_d)} \quad (11)$$

The recycling factor factor R for any particular assembly depends on the the size of the system as well as its composition and geometrical configuration.

The requirement for criticality in a finite system for any energetic particle is that $R = 1$ in the absence of an extraneous source; in these circumstances a steady state will be possible since just as many fast particles are produced as are lost in various ways.

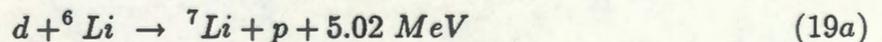
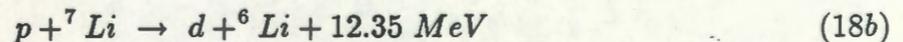
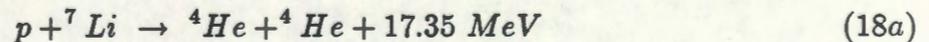
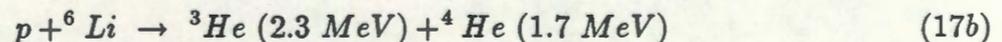
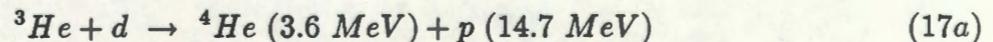
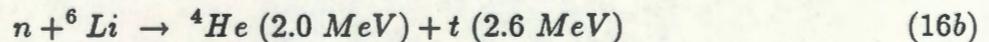
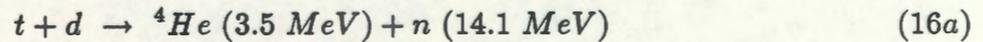
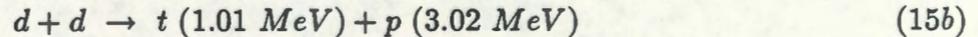
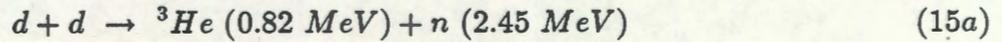
The chain reaction may consist of several cycles, each cycling and multiplying specific nuclei. For instance, assuming that the initial reaction is



the shortest t and ${}^3\text{He}$ cycles would be



${}^3\text{He}$, n , t and p can be considered as the $d-d$ source generated particles and the consumable nuclei are d , ${}^6\text{Li}$ and ${}^7\text{Li}$. Other most important cycles are listed below:



$$d + {}^6\text{Li} \rightarrow {}^7\text{Be} + n + 3.38 \text{ MeV} \quad (19b)$$

$$d + {}^6\text{Li} \rightarrow {}^4\text{He} + {}^3\text{He} + n + 1.79 \text{ MeV} \quad (20a)$$

$$d + {}^6\text{Li} \rightarrow {}^4\text{He} + t + p + 2.56 \text{ MeV} \quad (20b)$$

$$d + {}^6\text{Li} \rightarrow {}^4\text{He} + {}^4\text{He} + 22.37 \text{ MeV} \quad (20c)$$

$$n + {}^7\text{Li} \rightarrow {}^4\text{He} + t + n^* - Q \text{ MeV} \quad (21)$$

$$d + {}^7\text{Li} \rightarrow 2{}^4\text{He} + n + 15.12 \text{ MeV} \quad (22a)$$

$$d + {}^7\text{Li} \rightarrow {}^7\text{Be} + 2n - 3.87 \text{ MeV} \quad (22b)$$

$${}^3\text{He} + {}^7\text{Li} \rightarrow 2{}^4\text{He} + n + p + 9.63 \text{ MeV} \quad (23a)$$

$${}^3\text{He} + {}^7\text{Li} \rightarrow {}^9\text{Be} + p + 11.20 \text{ MeV} \quad (23b)$$

$${}^3\text{He} + {}^7\text{Li} \rightarrow 2{}^4\text{He} + d + 11.85 \text{ MeV} \quad (23c)$$

$${}^3\text{He} + {}^7\text{Li} \rightarrow {}^6\text{Li} + {}^4\text{He} + 13.33 \text{ MeV} \quad (23d)$$

$${}^3\text{He} + {}^6\text{Li} \rightarrow 2{}^4\text{He} + p + 16.88 \text{ MeV} \quad (24)$$

$$t + {}^6\text{Li} \rightarrow {}^7\text{Be} + 2n - 2.88 \text{ MeV} \quad (25a)$$

$$t + {}^7\text{Li} \rightarrow 2{}^4\text{He} + 2n + 8.86 \text{ MeV} \quad (25b)$$

$${}^4\text{He} + {}^6\text{Li} \rightarrow {}^{10}\text{Be} + \gamma + 4.46 \text{ MeV} \quad (26)$$

Regeneration of energetic particles is through series of direct and precursory reactions which are shown in Tables 1-4. (A number of less significant nuclear reactions with very low cross sections are not included in these reaction chains.) The regeneration cycle for p, n, t and ${}^3\text{He}$, and the recycling factor for direct and indirect regeneration of these particles are conceptualized on Figure 1.

For regeneration of each particle, R_D and R_P are recycling factors through direct and precursory reactions, respectively. The total recycling factor $R = R_D + R_P$ can be determined by calculating the probability of each branching reaction (including branch interdependencies). If the total recycling ratio for some particle is equal or greater than one, the system is critical or supercritical on that particle population, respectively. For a *critical system*, the population of the particle increases linearly (and indefinitely) and is proportional to the number of $d - d$ source reactions. Population of particles in a *supercritical system* increases exponentially with a time constant proportional to the life time of the particle in the system.

In a *sub-critical system*, regeneration of fast particles is independent of the source intensity S , and depends on the recycling factor R alone. The subcritical multiplication M is given by

$$M = \frac{S + SR + SR^2 + \dots}{S} = \frac{1}{1 - R} \quad (27)$$

provided that $R < 1$, i.e. that the assembly is subcritical.

Assuming for the moment no leakage, the recycling factor R can be obtained in terms of the fusion probability during the stopping time for energetic particles.

IV. Fusion Reaction Probabilities

Let us consider a flow of monoenergetic and monodirectional particles I_0 (particles/sec). For high particle energies one can accurately assume that the energy loss mechanism is predominantly by electronic interactions. To calculate the upper limit of

the probability for fusion reaction one can further assume that fusion reaction is the only reaction which removes particles. Let us define P_f as the probability of particle fusion before stopping in a homogeneous infinite medium:

$$P_f = \frac{1}{I_0} \int_0^{\infty} R_f \cdot dx \quad (28)$$

where R_f is the rate of fusion reaction per unit length and is provided in terms of the particle intensity at x

$$R_f = n\sigma_f I(x) \quad (29)$$

where

$$I(x) = I_0 \exp(-n\sigma_f x), \quad (30)$$

n is target nuclei number density, and σ_f is the fusion cross section. Transforming the relation for P_f to the energy phase space results in

$$P_f = \int_{E_0}^0 n \cdot \sigma_f \exp[-n \cdot \sigma_f \cdot \int_{E_0}^E \frac{dE}{(dE/dx)}] \frac{dE}{(dE/dx)} \quad (31)$$

Using the Lindhard - Scharf - Schiott (LSS) expression for the electronic stopping power /9/ with the modification proposed by Dickstein *et al.* /10/ to take into account charge (Z_2) oscillations, one arrives at

$$\frac{dE}{dx} = 8\pi\hbar\chi_e a_0 n \left(\frac{Z_2 Z_1}{Z}\right) \left(\frac{2E}{M_2}\right)^{\frac{1}{2}} \quad (32)$$

where

$$\chi_e = Z_2^{\frac{2}{3}} \left(\frac{Z_1}{A_1}\right)^2 [1 + \left(\frac{Z_1}{A_1}\right)^2 \sin(2\pi(Z_1+2)/18)] \quad (33)$$

with

$$Z = \left(Z_1^{\frac{2}{3}} + Z_2^{\frac{2}{3}}\right)^{\frac{3}{2}}. \quad (34)$$

In these relations a_0 is Bohr radius of the hydrogen atom ($a_0 = 5.2918 \cdot 10^{-9}$ cm), \hbar is the Planck's constant, M_2 and A_2 are the mass number and the atomic number of energetic

particles (projectiles), respectively, M_1 and A_1 are the mass number and the atomic number of the target nucleus, respectively, and n is the number density of target nuclei.

For charged particles, the relation for P_f can be reduced to:

$$P_f = 1 - e^{-cE_0^{\frac{1}{2}}} \quad (35)$$

where E_0 is the projectile energy at birth and c is given in terms of the average fusion cross section $\bar{\sigma}_f$ as:

$$c = \frac{1}{4\pi\hbar a_0(Z_1 Z_2/Z)\chi_e} \left(\frac{M_2}{2}\right)^{\frac{1}{2}} \bar{\sigma}_f \quad (36)$$

As is evident from Eqs. (35) and (36), the fusion probability during the stopping time is *independent* of the target density.

Calculation of fusion probability for each charged particle gives a recycling ratio R not larger than 10^{-4} . This result is obviously sensitive to the approximation of the stopping power, especially as different forms are used for different energy ranges. Equation (32) is designed to cover the intermediate to low energy range (keV to MeV), and overestimates stopping at high energies of new-born particles. We will now show that similar result is obtained assuming a stopping power designed to describe stopping at high energies.

Fusion probability during the stopping time can be expressed as /11/

$$P_f = 1 - e^{-\Omega} \quad (37)$$

where

$$\Omega = \int_0^{E_0} n \cdot \sigma_f \cdot v \, d\tau \quad (38)$$

and can be expressed in terms of the stopping power as

$$\Omega = \int_{E_f}^{E_i} n \cdot \sigma_f \left(\frac{dE}{dx}\right)^{-1} dE \quad (39)$$

where n is the target density, σ_f is the cross section for fusion and v is the velocity of the slowing projectile, with energy ranging between E_0 and 0. This energy decreases according to the stopping power which for high energies takes the form

$$S = -\frac{1}{n} \frac{dE}{dx} = \frac{4\pi Z_1^2 Z_2}{m_e v^2} \ln \frac{2m_e v^2}{\bar{I}} \quad (40)$$

where Z_1 is the averaged atomic number of the target, \bar{I} its average atomic ionization energy, Z_2 is the atomic number of the projectile, and m_e is the electron mass. Calculation gives $P_f \simeq 10^{-4}$, in essential agreement with the result obtained with Eq. (32). Therefore, even without leakage, the system can hardly get critical, i.e. reach steady state condition where just as many particles are produced as are lost by the "premature stopping" without fusion reaction.

The total recycling factor R for each charged particle is equal to the sum of probabilities of all branching reactions which regenerate the particle. For example, a pair of cycling reactions which regenerate ${}^3\text{He}$ is given by equations (14a and 14b). The probability of regeneration of ${}^3\text{He}$ through these reactions can be calculated from Eq. (35). For average fusion cross section not larger than tens of barns, $cE_0^{\frac{1}{2}}$ is much less than 1, and P_f can be reduced to

$$P_f \simeq cE^{\frac{1}{2}} \quad (41)$$

Using Eq. (14a) and assuming an average fusion cross section equal to 1 barn, P_f is $4 \cdot 10^{-5}$. For the regeneration of ${}^3\text{He}$ through the reaction given in Eq. (14b), and assuming a similar average cross section, P_f is $4 \cdot 10^{-6}$. Based on these probabilities, the partial recycling factor of ${}^3\text{He}$ through $[d({}^3\text{He}, \alpha)p ; {}^6\text{Li}(p, \alpha){}^3\text{He}]$ cycle is:

$$R_P = P_f[d({}^3\text{He}, \alpha)p] \cdot P_f[{}^6\text{Li}(p, \alpha){}^3\text{He}] \quad (42)$$

which gives $R_P = 1.6 \cdot 10^{-10}$. However, it should be noted that ${}^3\text{He}$ can be also cross generated by the 3.02 MeV protons (which are generated from a $d-d$ reaction, Eq. (12b)

through the fusion process given in Eq. (14b). The probability of ${}^3\text{He}$ generation through this reaction is estimated to be $2 \cdot 10^{-6}$.

A similar analysis can be performed for regeneration of p and t .

Calculation of recycling factors for reactions involving neutrons require development of a fairly detailed transport model including the absorption, scattering and leakage of neutrons as well as the description of the geometry, configuration and composition of the medium. Even under the high density conditions (alluded to in Section II) the probabilities of regeneration and cross-generation of charged particles by neutrons are expected to be small, and significantly less than those for reactions involving fusion.

V Conclusions

We can summarize our results as follows:

1) Based on known fusion cross sections, fusion energies and particle number densities in the system the probability of achieving any form of self-sustained fusion/fission reactions is four to five orders of magnitude less than required for critical conditions.

2) If such cycling reactions would have taken place, then heat generation could have been explained without direct correspondance to the rate of t and n productions.

3) As evidenced by the whole host of reactions in Tables 1-4, the rate of ${}^4\text{He}$ production should directly correlate to the heat generation. Therefore any "cold fusion experiment" reporting steady heat generation should look for abnormal levels of ${}^4\text{He}$.

Acknowledgments

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References

- /1/ M. Fleischmann and S. Pons, *Journal of Electroanalytical Chemistry* **261**, (1989) 301
- /2/ S. Jones *et al.* *Nature (London)* **338**,(1989) 737
- /3/ S.E. Koonin and M. Nauenberg, *Nature (London)*, **339**,(1989) 690
- /4/ K. Szalewicz, J. Morgan III and H.J. Monkhorst, *Phys.Rev A***40**,(1989),2824
- /5/ I. Shimamura, unpublished
- /6/ A.J. Leggett and G. Baym, *Phys. Rev. Lett* **63**,(1989), 191
- /7/ J. O'M. Bockris, *Modern Electrochemistry*, vol. 2, ch. 10 and 11, Plenum Press, New York 1970
- /8/ See, e.g. S. Glasstone and A. Sesonske in "Nuclear Reactor Engineering", van Nostrand Reinhold, New York, 1981
- /9/ J. Lindhard, M. Scharff and H.E. Schiott, *Mat. Fys. Medd. Dan. Vid. Selsk.* **33**,(1989) no. 14
- /10/ P. Dickstein, D. Ingman and N.H. Schafir, *Nuclear Science and Engineering* **98**,(1988) 255
- /11/ S.S. Gershtein *et al.* *Sov. Phys. JETP* **53**,(1981), 872

Precursory Reactions

^3He Production Reactions

^3He Loss Mechanisms

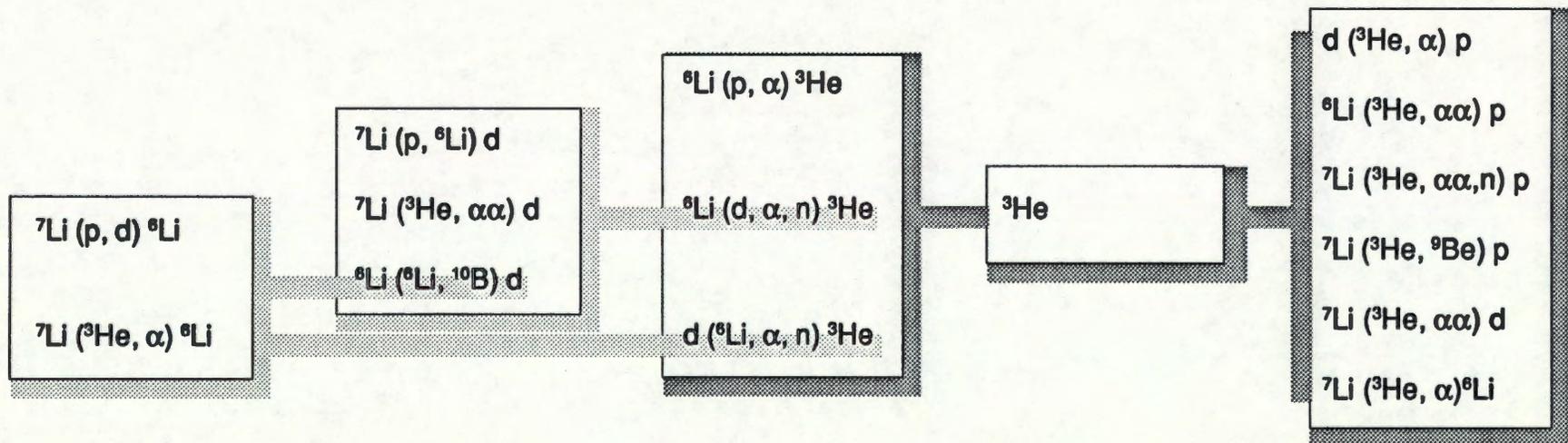


Table 1. ^3He production and loss mechanisms through a chain of direct cycling and precursory reactions.

Precursory Reactions

Neutron Production Reactions

Neutron Loss Mechanisms

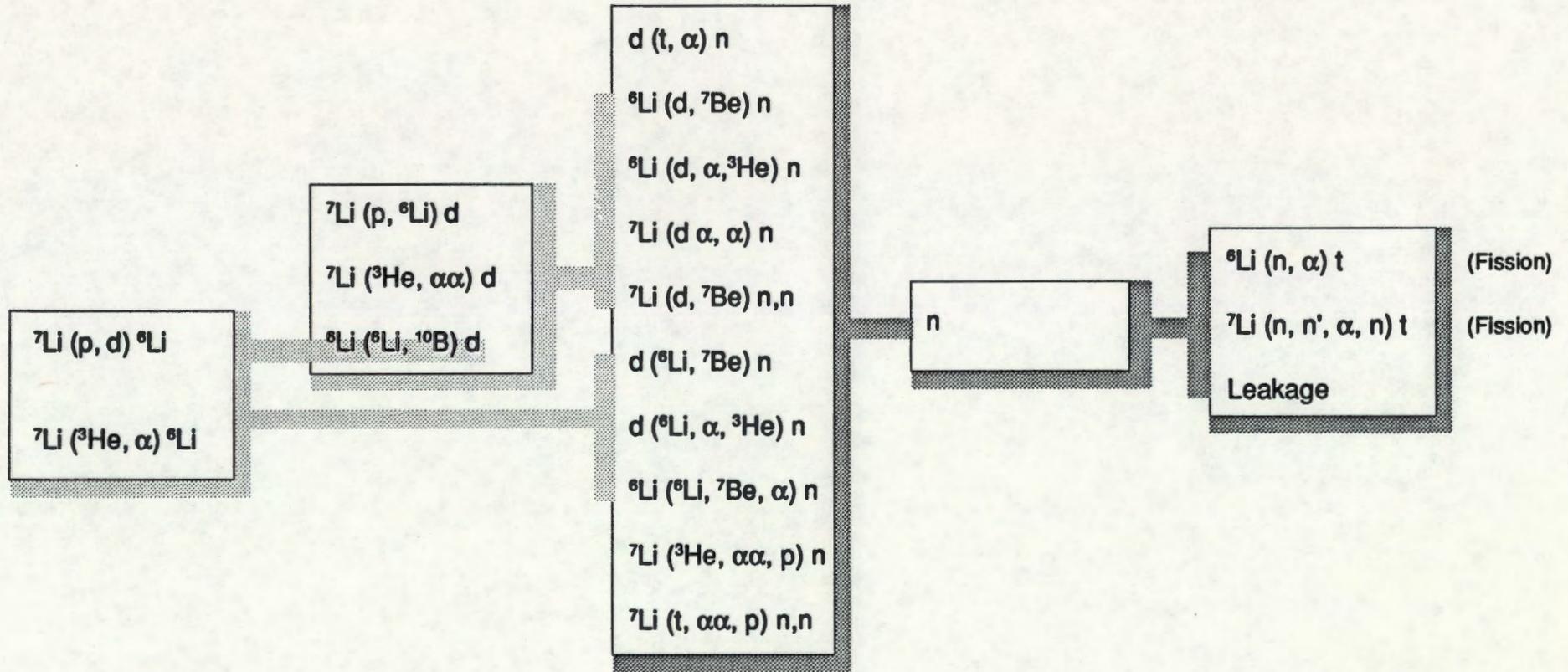


Table 2. Neutron production and loss mechanisms through a chain of direct cycling and precursory reactions.

Precursory Reactions

Tritium Production Reactions

Tritium Loss Mechanisms

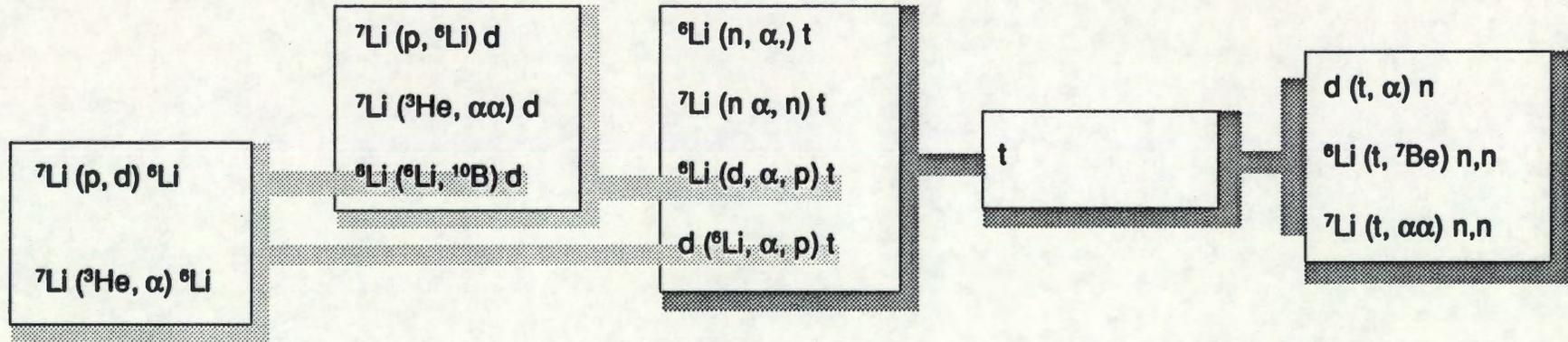


Table 3. Tritium production and loss mechanisms through a chain of direct cycling and precursory reactions.

Precursory Reactions

Proton Production Reactions

Proton Loss Mechanisms

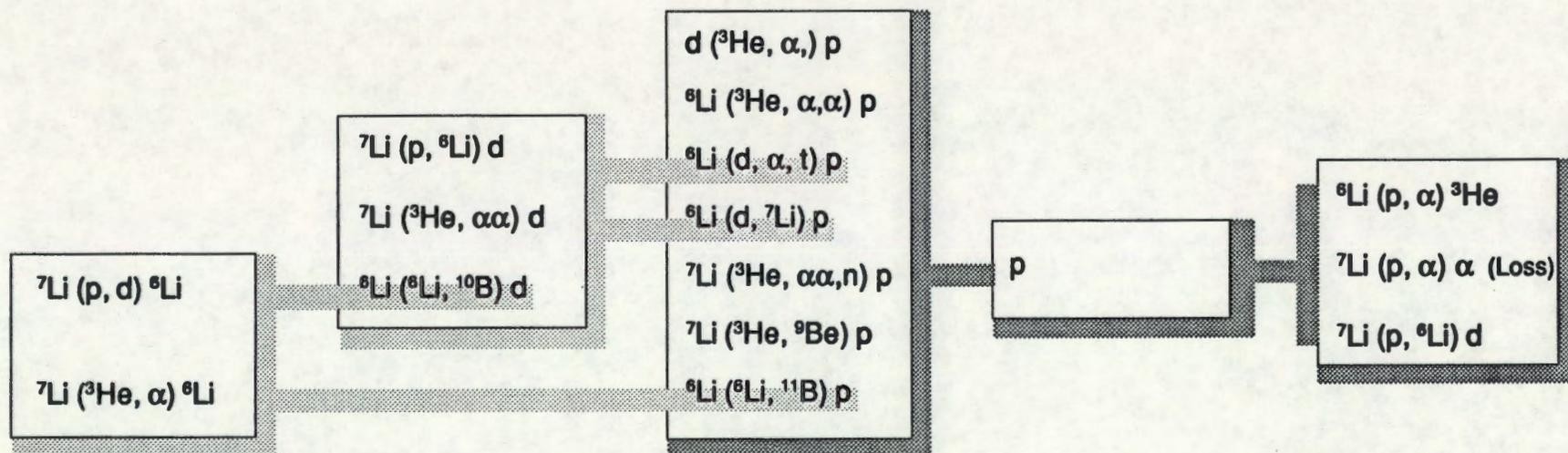


Table 4. Proton production and loss mechanisms through a chain of direct cycling and precursory reactions.

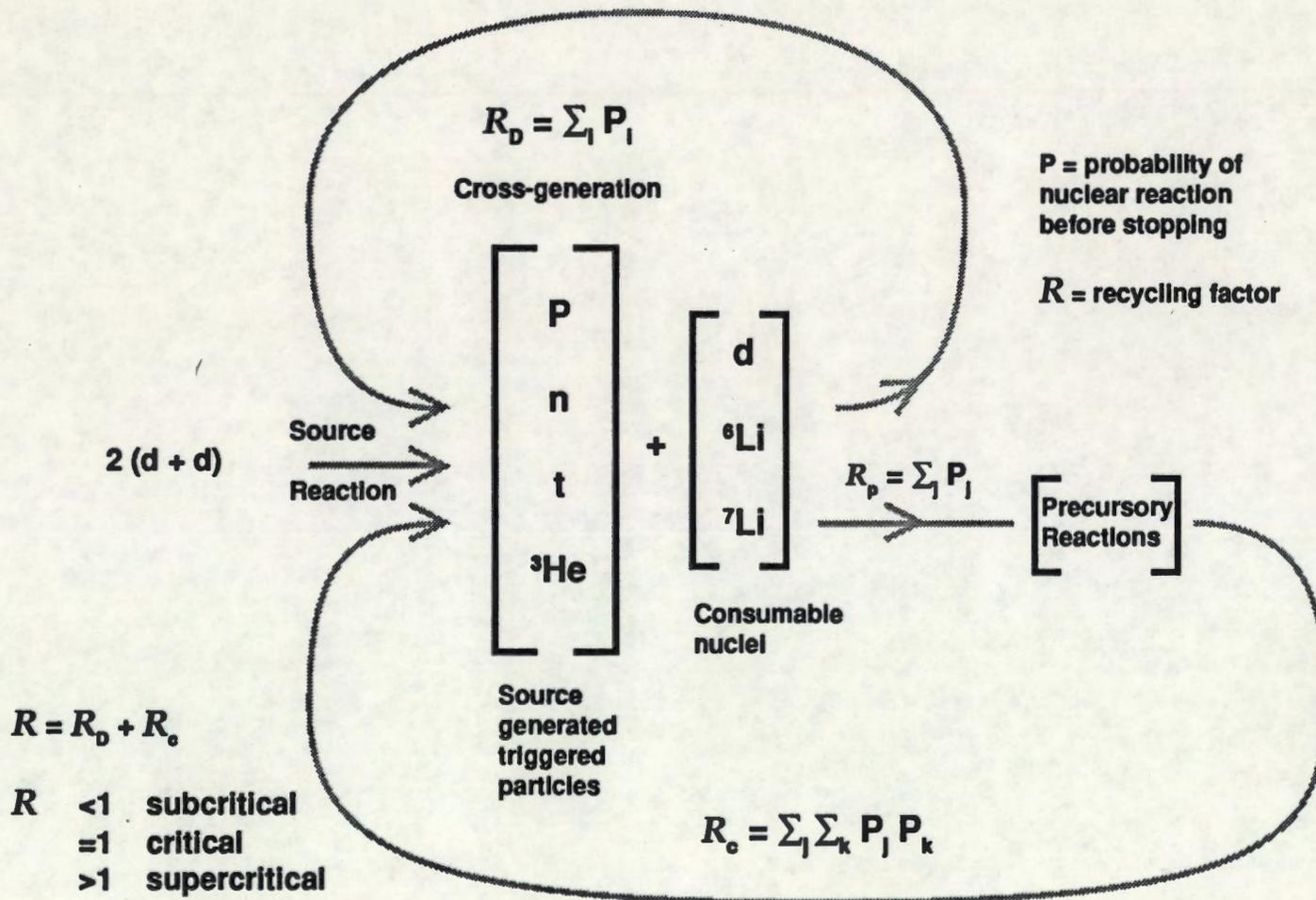


Figure 1. Cross-generation and recycling of energetic particles.