Conference Proceedings Vol. 33
«The Science of Cold Fusion»
T. Bressani, E. Del Giudice and G. Preparata (Eds.)
SIF, Bologna 1991

SURVEY OF GAS LOADING EXPERIMENTS

F. Scaramuzzi
ENEA, Area Innovazione, Dipartimento Tecnologie di Punta,
C.P. 65 - 00044 Frascati, Italy

WHY GAS LOADING ?

In March 1989 the results of two experiments (1,2), claiming for nuclear reactions taking place, at room temperature, in metal lattices (Pd and Ti) charged with deuterium, were presented. In both cases the technique chosen for charging the metals with deuterium consisted in using an electrolytic cell, containing heavy water, in which the cathodes were made out of Pd or Ti.

Soon later, in April, the Group led by the writer addressed a very straightforward question: if nuclear reactions take place in a metal lattice because of the interaction between the deuterium nuclei and the lattice, is electrolysis the only route to be followed, in order to produce them? Wouldn't it be possible to perform experiments, having the same purpose, by letting the lattice to interact with deuterium in the gaseous phase? The question seemed quite appealing, mostly for one reason: the physical system consisting in an electrolytic cell is a very complicated one, and has to take into account a great number of parameters, while the system consisting in a metal and a gas looks much simpler. The latter would permit much cleaner experimental conditions, and thus it would be possible to analyze more clearly the experiments; it would also favour a higher reproducibility, and would enable testing the proposed theories. Experiments were performed at the Frascati Laboratory of ENEA following this alternative route, using titanium: furthermore, it was decided that, in order to favour nuclear reactions, temperature cycles should be performed on the system (from 77 K to room temperature). Positive results were obtained, consisting in the detection of neutron bursts, and were soon published (3).

Since then, many laboratories have used this technique, called "gas loading", with various success, detecting the emission of neutrons and charged particles and the accumulation of tritium in the metal. The basic idea is that the unknown mechanism that produces nuclear reactions in the system is connected to the thermodynamic transformations taking place in it, while the temperature of the sample, the pressure of the gas, and the related parameters, such as deuterium absorption, are changing.

This is presently one of the most used techniques in Cold Fusion research. Many results have been reported also in this Conference, and will be reviewed in the following. But, at two and half years from the starting of this experimental technique, it is worth to address a question, in the light of the lack of reproducibility that still characterizes its results: is the system under study really a simple system, as it appeared to be at the beginning?

The answer to this question is not easy: in a first approximation, it can be stated that the system is simple in principle, but that most of the particular solutions chosen up to now to study it, including the Frascati experiment of April 1989, show a very high complexity, that tends to cancel the advantage of this "theoretically simple" system. In order to enlighten this concept, I will examine in detail the intrinsic complexity of a typical "gas loading" experiment, then I will survey the experiments pertaining to this class presented in this Conference, and finally I will try to suggest, also in the light of the papers presented here, new possible routes, that could help reducing the complexity of the studied system, taking thus advantage of the real intrinsic simplicity of the metal-gas system.

INTRINSIC COMPLEXITY OF A GAS LOADING EXPERIMENT

I will try to list here (being aware that the list is not exhaustive) the many operations on the sample, while performing a gas loading experiment, and the relative parameters: in the hypothesis that the studied phenomena are connected with thermodynamic transformations (in particular phase transformations) taking place within the sample, each of them can influence the outcome of the experiment. I choose to refer to the experiment of the Frascati Group at the Gran Sasso Laboratory of INFN, presented at this Conference. The list is reported in Table 1: on the left I described briefly the procedure or the parameter, on the right I reported the solution chosen for this experiment. It must be clear that in many cases the choice of the procedure or of the particular value for a parameter did not follow from a thorough knowledge of the role played by that parameter or that procedure in the experiment: it is often suggested only by practice, mostly acquired in preceding measurements. Thus, changing that procedure or the value

Table 1. Complexity in gas loading experiments. The data refer to the paper by De Ninno on these Proceedings.

PROCEDURE/PARAMETER	FRASCATI CHOICE
A. Choice of the sample	
1. substance	Ti-662
2. purity	99.6%
3. impurities	?
B. Shape and amount of sample	
4. shape	shavings
5. size	section: 0.05 x 1.00 mm^2
6. amount	$\approx$ 120 g
C. Preparing the sample	
7. machine used	lathe
8. cutting tool	sharpened at beginning of machining
9. cooling while cutting	flow of pure ^4He gas
10. after cooling	sample exposed to air
D. Cleaning the sample	
11. acid etching	no
12. organic solvent	no
E. Pretreatment of the sample	
13. cell used	stainless steel
14. vacuum on sample	turbomolecular pump, p $\approx 10^{-6}$ torr
15. degassing the sample	pumping at 100°C, 2 hrs
F. Charging D_2 gas (at Gran Sasso Laboratory)	
16. time from step 15	$\approx$ 18 hrs
17. charging circuit	uses a zeolite trap in LN_2 in order to clean the gas
18. charging pressure	20 bars
19. time at room temperature before heating	2 hrs
G. Heating sample to favour absorption	
20. temperature vs. time	up to $\approx$ 300°C in 1 hr
21. control of absorption	see Fig.1
22 restored pressure	P = 30 bars
H. Thermal cycle (repeatable)	
22. time to return to room temperature	$\approx$ 1 hr
23. cooling procedure	immersion of the cell in LN_2; 3-5 min to reach 77 K
24. warm-up procedure	cell immersed in a dewar full of LN_2, in the well of the detector; $\approx$ 24 hrs to warm up to room temperature

of that parameter would be a proper way to learn about its role: the multiplicity of them, however, renders this task quite difficult.

It is clear, looking at Table 1, that this experiment has quite a number of undefined features. Let me try to point out those that seem to be more relevant:

- Temperature is measured with a thermocouple, within the cell, touching the upper part of the tangled mass of shavings. Due to the poor heat conductivity of titanium, the deuterium gas gives a better contribution than the titanium itself to a uniform distribution of temperature. In spite of this, since non-equilibrium conditions are purposely searched in the experiment, temperature gradients and time variations of temperature and of temperature gradients are likely to be created.
- Other parameters that are presumably relevant to the experiment are the D/Ti concentration, its gradient and the time evolution of both, which depend on the absorption procedure. Tests made on samples used in this type of experiment have shown the presence of high concentration TiD_x, while the average concentration is usually in the order of a few percent D atoms per Ti atom.
- The treatment of the Ti shavings before putting them in contact with deuterium is also most probably relevant, including the thermal treatment produced when making them on the lathe, the successive chemical cleaning, the degassing, and so on.

To control all these parameters in this kind of experiment is extremely difficult. Most probably, during one run a great number of different experiments, in which each of the relevant parameters has a different value, unknown to us, are performed in different sites of the sample. This feature probably increases the probability of positive results, but puts a severe limit to the reproducibility of the experiment.

In order to improve reproducibility, a more rational approach to the problem is paramount. On one side one should try to better understand the processes that take place in a metal (in particular titanium and palladium) when interacting with deuterium. The literature is rich, and we had in this Conference a very interesting lecture by Professor Schlapbach on this subject. Nevertheless, the information available, in particular for titanium, seems to require more investigation, in order to gather information useful for cold fusion experiments. An extremely inspiring paper has been presented at this Conference by Professor Sanchez, in which he showed us a correct route to follow in order to better understand the systems on which we are working.

Another possible approach consists in the search for much better characterized systems on which to perform experiments. Many interesting suggestions in this direction have come out in this Conference: I will address this subject in the last part of this paper.

GAS LOADING EXPERIMENTS PRESENTED AT THIS CONFERENCE

It would be difficult, and at the same time pointless, to review here in detail all the papers referring to gas loading experiments presented at this Conference: they are published in these Proceedings. I tried to list them, in Table 2, reporting a few descriptive data, and I will propose some comments.

The first feature that appears is the large number of experiments belonging to this general area; at the same time, it has to be noted that there is a great variety of particular features, that, with a few exceptions, renders every experiment different from the other. The high quality and accuracy of some of them has also to be pointed out: in particular, the neutrons' energy measurement of Bressani, the time resolution in neutron detection of Menlove and of De Ninno, the neutron measurements at Dubna, the tritium measurements of Lanza.

As far as neutron detection is concerned, it has to be pointed out the "dilemma" about whether or not to measure the energy of the emitted neutrons. The measurement of the energy, beyond giving an important information on the physics of the problem, has the advantage of guaranteeing a better identification of the neutron, but requires the accumulation of many data, i.e., long times. The choice of privileging the analysis of the time structure of the emission (the "bursts"), on the other side, requires the use of a moderator, in order to enable the detector to see also neutrons emitted within a very short time interval (μs, ns ?), and to increase the overall efficiency of the system; thus, all information about the energy of the detected neutrons is lost. The latter, at the same time, has the advantage of correlating in real time the neutron emission with the evolution of the thermodynamic parameters of the system. Both approaches are important and should be followed. It seems to me, to summarize the issue, that the second type of approach could be more helpful in the search for a better reproducibility; but there is no doubt that, as soon as a more reproducible experiment will be at hand, it will be paramount to measure the energy of the emitted neutrons.

Going back to the problem described in the first part of this paper, the complexity shared by most experiments, it has to be noted that, among the papers presented at this Conference, many show possible routes to follow in order to reduce it. I will try to list them in the following, looking for systems and procedures that are likely to be used in gas loading experiments, aiming to a

Table 2 - Gas Loading experiments presented at this Conference.

Author	Metal	Part.	Temp.	Notes
Bressani	Ti	n	T_a-540	energy = 2.5 MeV
Cecil 1	Ti	cp	77-T_a	bursts; high DC current in sample (400 mA)
Cecil 2	Ti	cp	T_a	glow discharge
Celani	YBCO	n	77-T_a	superconductive transition, stimulation by n
Claytor	Pd-Si	t,n	T_a	pulsed electric current
De Ninno	Ti	n,t	77-T_a	low background lab, bursts
Duan	Ti	n	T_a	implantation
Ikegami 1 (Hitachi)	Ti	n	77-T_a	bursts
Ikegami 2 (NIFS)	Pd	n	T_a	electric discharge
Ikegami 3 (NTT)	Pd	n	T_a	oxide/Pd:D/Au interface,electric current
Lanza	Ti,Zr, Hf,T_a	t	77-T_a	careful control of deuteration
Li 1 (Tsinghua)	Pd	cp	77-T_a	CR-39 used,search of precursors, E > 5 MeV
Li 2 (At.En.)	Ti	n	77-T_a	bursts,low background laboratory
Li 3 (various)	Pd	cp	T_a	CR-39 used,gas discharge
Menlove	Ti	n	77-T_a	low background lab, bursts
Miley	Ti,Pd Th,Fe	-	T_a	proposal:thin films, implantation
Seeliger 1	Ti	n	77-T_a	correlations,p,dp/dt
Seeliger 2	Pd	n	Ta-650	electrolysis-charged
Tsarev 1 (Dubna)	Ti	n	77-T_a	bursts

Legend:
PART = type of particles detected; n = neutron; cp = charged particles; t = tritium; T_a = ambient temperature. Temperatures are expressed in K

Note. In the column entitled "author" one of the authors, in most cases the speaker, is quoted, and the same name is used in the text when commenting the Table. In the whole paper the references to the presentations appearing in these Proceedings will not be reported: the reader will easily find out the corresponding paper in the Proceedings by looking at the alphabetic index.

Table 2 - Gas Loading experiments presented at this Conference.

Author	Metal	Part.	Temp.	Notes
Tsarev 2 (Kharkov)	Pd,Ti	n,cp	77-1300	D_2 implanted at low T then rapid heating
Tsarev 3 (various labs)				various techniques, including mechanical treatment, discharge
Wang	Pd,T	cp	77-T_a	CR-39 used
Will	Pd	t	T_a	electric discharge

Legend:
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better reproducibility of the measurements. In particular, the issues at stake are a better characterization of the sample, and a better definition of the procedure to follow in order to obtain the non-equilibrium conditions necessary for the phenomenon to take place.

- A procedure that looks very promising is the D and D_2 ion implantation on metals, in order to achieve high values for the D-concentration, which can be definitely higher than those obtained both with electrolysis and gas loading. The damage produced by the implanted ions in the lattice, however, has to be taken into account. Also the use of "plasma focus" plants belongs to this category. (Tsarev/Kharkov, Srinivasan/BARC ((4),not presented in this Conference), Duan, Miley (proposal), De Ninno (proposal))

- The experiment of the Los Alamos Group (Claytor) with Pd and Si stacks pulsed with high electric current represents an interesting compromise between electrolysis and gas loading, still keeping the "intrinsic simplicity" of a gas-solid system, but at the same time realizing a much simpler system than the traditional one described here (Table 1).

- The experiment on Pd films coated with "barrier layers" (Ikegami/NTT) shows as well a very interesting route to follow, in order to have a well characterized system, and investigates one of the possible mechanisms for the nuclear reactions to take place.

- Also the use of gas discharge, being careful not to produce "hot fusion" events, is an interesting possibility for cold fusion investigation. (Cecil, Li, Will)

- Finally, referring to the issue of producing well characterized samples, it would be interesting to realize particular structures, like superlattices (Miles), or growing a film on a support, by evaporating or sputtering deuterium and the chosen metal at the same time in the wanted proportion (5).

I would like to remember here the suggestion of Prof. Li to give more attention to the detection of charged particles, in particular with integrating detectors, such as C-39, which are simple to use and not expensive. The detection of tritium should also be pushed forward. Gas loading experiments are well suited for the detection of all these particles.

References

1. M. Fleischmann, S. Pons, M. Hawkins, Journal of Electroanal. Chem., **261**, 301 (1989)

2. S.E. Jones, E.P. Palmer, J.B. Czirr, D.L. Decker, G.L. Jensen, J.M. Thorne, S.F. Taylor, J. Rafelski, Nature, **338**, 737 (1989)

3. A. De Ninno, A. Frattolillo, G. Lollobattista, L. Martinis, M. Martone, L. Mori, S. Podda, F. Scaramuzzi, Europhys. Letts, **9**, 221 (1989)

4. P.K. Iyengar, M. Srinivasan, Proceedings of the First Annual Conference on Cold Fusion, Salt Lake City, March 28-31, 1990, edited by the National Cold Fusion Institute, pag.62 (1990)

5. S. Scaglione, P. Zeppa, private communication