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Evidence for Tritium generation in Self-heated Nickel Wires subjected To Hydrogen Gas Absroption/Desorption Cycles

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Abstract

The loading characteristics of hydrogen gas in electrically self-heated nickel wires was investigated with a view to maximise hydrogen absorption and thereafter "trigger" it to generate anomalous excess heat as reported by Focardi et. al in early 1994. The nickel wires were found to absorb substantial quantity of hydrogen following several alternate cycles of absorption/desorption. But calorimetric studies conducted with the system so far indicate that we have not succeeded in triggering excess heat generation. However on dissolution and counting using standard liquid scintillation techniques, a number of hydrogen loaded nickel wires were found to contain tritium in the range of 3 Bq to 2333 Bq. This finding corroborates the detection of tritium in light water solutions electrolysed by nickel cathodes reported by the authors first at ICCF - 3 (Nagoya, 1992) and again at ICCF - 4 (Hawaii, 1993), confirming the occurrence of anomalous nuclear reactions in nickel-hydrogen systems.

Introduction

Focardi et al have reported [1-4] observation of anomalous "excess heat" generation in a "reproducible" manner, in a nickel rod (5mm dia x 90 mm long) subjected to repeated cycles of hydrogen gas absorption/desorption. In their experiments the nickel sample was heated by means of a platinum heater coil mounted around it. We report here attempts to replicate this so called "Piantelli experiment". In our experiments the nickel sample was taken in the form of an electrically self-heated wire (either 125 um or 380 um dia), 35 to 50 cm long. The self-heated wire method has the significant advantage that the wire temperature can be easily and precisely controlled through its resistance ratio (Rt/Ro), where Ro is the initial room temperature resistance.

For calorimetric measurements a double walled glass cell was employed. Coolant water was passed through the outer jacket in a once through fashion. A pair of RTDs mounted at the inlet and outlet of the water jacket measured the Delta T rise across the cell. Knowing the water flow rate in milliliters per minute, the heat dissipated in the cell could be easily measured and compared with the power input (product of voltage and current) to the nickel wire coil for detecting "anomalous" excess power production, if any.

Experimental Set up and Methodology

The apparatus comprised of a glass vacuum line provided with a rotary vacuum pump and a gas handling system incorporating a Toepler pump, intermediate gas storage reservoir etc. The cell in which the nickel wire was mounted in the form of a spring was made either of Pyrex or Quartz glass and was provided with a pair of tungsten electrical connecting leads. lolar grade hydrogen gas was used for the absorption experiments. The absolute pressure of H_2 gas in the system was measured by a mercury manometer. The quantity of hydrogen gas absorbed/desorbed during a given cycle could be accurately measured by means of a sensitive silicone oil differential manometer. Fig.l is a schematic diagram of the experimental set up. At the outset the voltage-current characteristics of the nickel wires was studied both in vacuum and in H_2 gas. All the wire samples displayed a unique and characteristic shape for the V-I curve (See Fig.2), composed of three distinct regions:

Region I :	$1.0 < (Rt/Ro) < 1.9$
Region II :	$1.9 < (Rt/Ro) < 3.5$ (up to curie point)
Region III :	$3.5 < (Rt/Ro)$ (above curie point)

Interestingly above the curie point the V-I curve was remarkably linear giving a normalised incremental resistance ratio ((dV/dI)/Ro, a dimensionless number) of ~ (9.0 + 1.0) in all cases.

Hydrogen Absorption Characteristics of Nickel

The first objective of the experiments was to study the hydrogen loading characteristics of nickel samples and optimise the procedure which will lead to maximum amount of net H absorption. It is generally known that repeated cycles of loading/unloading has to be resorted to, to obtain high loading values. The procedure arrived at for this, after several trial runs, was as follows:

After degassing / annealing the Ni wire under dynamic vacuum at glow hot condition for about 5 minutes, Ro the initial resistance at room temperature is recorded. The surface of the wire is then "activated" by first annealing in air at low pressure and then introducing hydrogen gas under glow hot condition. After noting the initial H_2 gas filling pressure at room temperature, the wire is allowed to absorb hydrogen for about 10 to 15 hrs. As pointed out by Focardi et. al[(1)] we also find that the rate of gas absorption is somewhat faster if the nickel wire temperature is set at about 200°C to 300°C corresponding to Region II of the V-I characteristics. When the absorption has reached a saturation value, a cycle of desorption is commenced by evacuating the system and taking the wire to glow hot condition. The quantum of absorption or desorption is measured using the silicone oil manometer. Fig.3 shows a typical variation of the net amount of H_2 gas absorbed by a nickel wire as a function of the absorption/desorption cycle number. In Fig.3 odd numbers on the X-axis correspond to absorption cycles (open circle points) and even numbers to desorption cycles (crosses). It was generally observed that during 4 to 5 hours of desorption, approximately a third of the quantity of gas intake of the previous absorption cycle is released. Table I summarises the quantum of hydrogen gas absorbed, in units of centimetres of silicone oil, in some of the nickel wire samples studied by us. Only data for those wires which gave tritium are included in this table.

The quantity of H_2 gas absorbed in units of cm of oil can be converted into mass of H_2 gas loaded into the nickel wire, provided the volume of the cell plus the dead volume of the system are known. In our experiments for the case of a 60 ml volume cell, the effective system volume was measured to be ~68 ml, and hence 1 cm of pressure drop in the silicone oil manometer corresponds to ~5.3 μg of H gas absorption in the nickel wire. Thus for the case of wire # Ni-27 (48.6 mg weight) for example, the net absorption as per silicone oil manometer reading of 436 cm of oil (see Table I) implies 2.3 mg of H_2 gas absorption. This corresponds to an average H/Ni loading ratio of 2.6 which is even higher than in NiH_2! Such a large loading ratio is obviously very difficult to believe.

In order to verify the exact magnitude of hydrogen loading in the Ni wires, some samples were got analyzed by an "inert gas fusion method". (This instrument melts the nickel and measures the quantum of hydrogen released into the flowing argon cover gas, through a calibrated conductivity technique). The inert gas fusion measurement however indicated a much lower value of H/Ni, in the region of a few hundred ppm only. We are currently investigating the possible causes for the large discrepancy in loading ratio between oil manometer and inert gas fusion results.That there is no basic flaw in the silicone oil manometer measurement was independently confirmed by loading hydrogen in Pd wires[5]. In the case of Pd, three different techniques of assessing the H/Pd loading ratio, namely resistivity method, oil manometer method and inert gas fusion method have all given comparable results.

However, it must be pointed out that even the several hundred ppm level of loading in nickel, indicated by the inert gas fusion set up, is much higher than the 10 ppm value quoted by D.P. Smith[6] for nickel, presumably obtained using "conventional gas loading" techniques. We speculate that the electrically self-heated wire method, is probably helpful in obtaining higher loading values.

Measurement of Tritium Content of Hydrogen Loaded Nickel Samples

The authors have earlier reported [7, 8] observation of tritium in light water solutions of K_2CO_3, Li_2CO_3 etc electrolysed by nickel cathodes wherein hydrogen is loaded electrolytically into the nickel. As such it was suspected that tritium may also be generated during gas phase loading of hydrogen into nickel. To investigate this possibility, the loaded nickel wires in the present experiments were cut into 3 or 4 pieces of 8 to 12 cm length each and dissolved separately in 5 ml of dilute (10%) $HN0_3$. At least 24 hours time was given to enable complete dissolution. Excess acid was then neutralised by adding AR grade anhydrous Na_2CO_3, and the solution distilled under partial vacuum using a microdistillation set up.

Standard liquid scintillation counting technique was adopted for measuring the tritium content of the distilled sample. For this 1 ml of the distilled water sample (derived from the nickel solution) was added to 10 ml of scintillation cocktail (mixture of toluene + Triton X-100 + scintillation chemicals) and counted in a scintillation counting unit, after allowing about 16 ~ 20 hrs for "chemiluminiscence" effects to decay.

So far 6 out of 9 loaded wires have indicated generation of tritium. However not all the cut pieces from a given wire showed tritium, suggesting that tritium production is not uniform over the length of the wire. For example, out of 27 cut pieces which have been dissolved and

counted up to now, only 14 pieces were found to contain tritium. Table II summarises the tritium content of these cut wire samples. The measured tritium activities range from 3 Bq to 2333 Bq. For the lowest activity case of 3 Bq (in 5 ml of dissolved solution) the count rate was ~30% above the background value of ~250 counts/10 mins. The maximum tritium activity was observed in a 11 cm segment of Ni-27 wire which displayed exceptional absorption/desorption characteristics in terms of both rate as well as quantity of H_2 absorption (see Table 1).

In the case of one of the cut pieces of wire # Ni-504 wherein the surface layers were leached out separately prior to dissolving the balance portion, both the solutions (sample nos 504/5/I and 504/5/II) indicated presence of tritium. Blank (or control) samples cut from the two nickel stock spools (125 μm dia and 380 μm dia) dissolved and counted following an identical procedure have not given any counts above background levels.

Two cut pieces from two different loaded nickel wires when exposed sandwiched between a pair of medical X-ray films gave clear autoradiographic images. The images on the upper and lower photographic films were identical. The sample from one wire gave identical but weak spotty images, while the sample from the other wire gave images which were more intense though diffused.

At present we are not in a position to state whether these images are due to presence of tritium or some luminiscence type of phenomena observed in PdH_x [10].

Summary and Conclusions

Hydrogen loading characteristics of self-heated nickel wires have been investigated with a view to try and replicate the anomalous production of excess heat, first reported by Focardi et. al[1]. We seem to be obtaining bulk loading ratios of at least a few hundred ppm. The loading ratio in the near surface region may be somewhat higher. We have however not been able to obtain the conditions necessary for the production of excess heat. About two thirds of the loaded nickel wires indicated presence of tritium on dissolution and counting in a liquid scintillation counting set up, using standard procedures. Tritium generation is however found to be non-uniform along the length of the wire. The quantum of tritium in individual cut wire pieces was in the range of 3 to 63 Bq except for one wire segment which gave an unduly large amount of 2333 Bq. These results corroborate the generation of tritium reported earlier by us[7,8] as well as Notoya et al[9] during electrolytic loading of hydrogen in nickel cathodes. It is possible that a significant amount of tritium is getting lost from the wires during the desorption phase and hence it may be worthwhile to search for tritium in the gas phase also. A couple of loaded nickel wires have given clear autoradiographic images on medical X-ray film.

Although no excess heat has been detected by us to date, the occurrence of some anomalous nuclear process in hydrogen loaded nickel wires stands confirmed in view of the observation of tritium in several samples.

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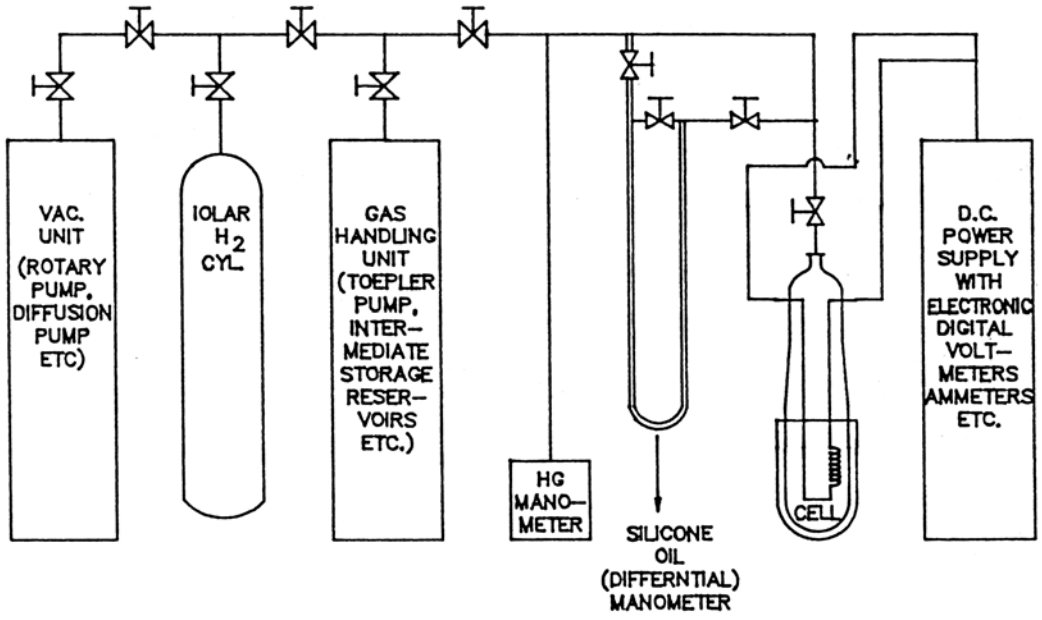

Fig. 1. SCHEMATIC DIAGRAM OF THE EXPERIMENTAL SETUP

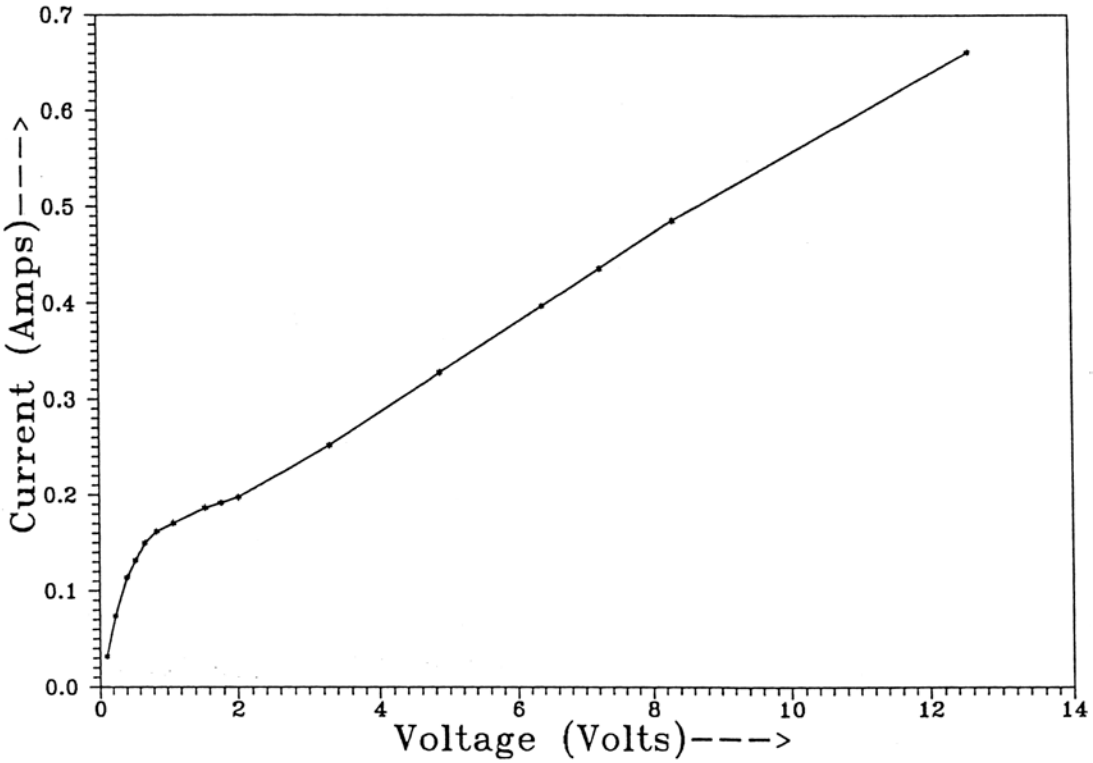

Fig. V-I CHARACTERISTICS OF Ni WIRE IN VACUUM

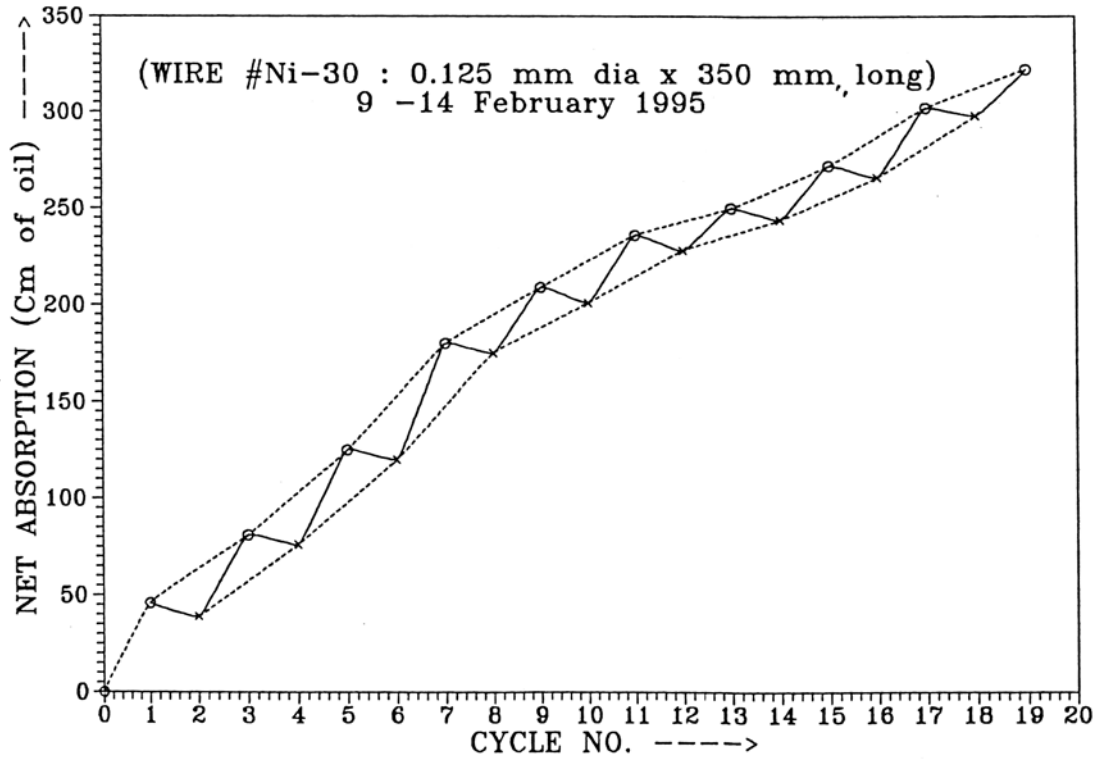

Table 1. QUANTUM OF HYDROGEN GAS ABSORPTION IN NICKEL WIRE SAMPLES THAT HAVE INDICATED PRESENCE OF TRITIUM

Sr. No.	Wire #	Dimensions Dia length mm cm		Mass mg	Duration of loading	Number of cycles	Net Loading
1	Ni-23	.125	50	53.8	7th OCT to 13th OCT	5	*
2	Ni-501	.38	50	500	24th OCT to 27th OCT	5	125
3	Ni-501	.38	50	500	23rd OCT to 9th DEC	15	203
4	Ni-24	.125	50	54.1	12th DEC to 15th DEC	5	58
5	Ni-27	.125	45	48.6	18th JAN to 25th JAN	6	436
6	Ni-30	.125	35	39.4	9th FEB to 14th FEB	10	325

* Not measured

Table 2. OBSERVED TRITIUM ACTIVITY IN DISSOLVED CUT PIECES OF Ni WIRE SUBJECTED TO SEVERAL HYDROGEN ABSORPTION / DESORPTION CYCLES

Sr. No.	Dissolved Cut Wire Sample #	Average Excess Over Background Counts * per 10 minutes (1 ml Solution)	Total Tritium Activity in Cut Wire piece (5 ml Solution) (Bq)
1	23/1	313	13
2	501/1	532	22
3	504/4	152	6
4	504/5/I	70	3
5	504/5/II	103	4
6	24/1	440	18
7	24/2	690	28
8	24/3	1150	47
9	27/1	950	38
10	27/2	704	28
11	27/3	57650	2333
12	30/1	1560	63
13	30/2	220	9
14	30/3	550	22
15	Standard	4200	170

* Background count rate was approximately 250 counts in 10 minutes. 10 % in excess of BG represents limit of sensitivity