

## LOW TEMPERATURE NUCLEAR CHANGE OF ALKALI METALLIC IONS CAUSED BY ELECTROLYSIS

R. Notoya

Hokkaido University, Catalysis Research Center

Kita-11, Nishi-10, Kita-ku Sapporo, 060, Japan

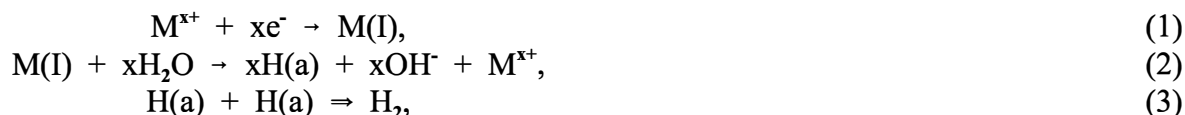
### ABSTRACT

It was found that the hydrogen evolution reaction on platinized platinum and porous nickel in various alkaline solutions was causative of the nuclear reactions. 0.5 mol/l potassium and sodium carbonate, and 0.1 mol/l cesium sulfate solutions of light and heavy water were used for electrolysis in thermally open cells. Analysis of the electrolytes by use of ICP-MS, a flame photospectrometer, a liquid scintillation spectrometer and a  $\gamma$ -ray spectrometer, revealed that the some nuclear products were generated during electrolysis, for example, from potassium and proton to calcium, from cesium-133 and neutron, proton and so forth to some species of mass numbers of 132 to 140 amu, from sodium-23 and neutron to sodium-24, as well as tritium, which were accompanied by an extraordinary large heat evolution. The reaction mechanism for the cold fusion caused by electrolysis is proposed, in which the intermetallic compounds between alkali metals and cathode materials play an important role in making the solid reaction possible.

### INTRODUCTION

In order for low temperature nuclear reactions to occur in solid, the reactants must be present in solid. The cold fusion in heavy water was observed on palladium electrode on which hydrogen electrode reaction was occurring. In such a system, a deuterium which has been regarded as the reactant is absorbed in palladium metal very well [1]. Accordingly the reactant seems to be present in solid in this case. On the other hand in the case of light water cold fusion, there are few works to make a mention of this point of view [2]. But, people do not propose of the low temperature nuclear reactions of alkali metallic species until they show the possibility of these species being present in solid. The author has dealt with this possibility, on the basis of the mechanism for the hydrogen evolution reaction (HER) [3].

The mechanism for HER in alkaline solutions on the so-called low overvoltage metals, namely, platinum group metals, nickel, silver, titanium and so forth has been established by A. Matsuda and his coworkers by use of the galvanostatic transient method (GSTM), as follows [4]:



where  $M^{x+}$ ,  $M(I)$  and  $H(a)$  denoted an alkali or an other metallic ion with the valence  $x+$ , the intermetallic compound between alkali or other metal and the cathode material and an adsorbed or absorbed hydrogen atom on or in the electrode, and  $\Rightarrow$  the rate determining. On the contrary, almost of the electrochemists in the world believe that the electron transfer step of HER in alkaline solutions consists in the discharge of water molecule. It can be denied on the basis of the following experimental result.

The exchange current density  $i_{10}$  of the electron transfer step (1) determined by GST-analysis of the most initial change of overvoltage shows a linear relationship with that of the concentration of alkali metallic ion but not pH with the slope of 0.5 for mono-valent or 0.8 [5] for di-valent discharge. That is consistent with the kinetic equation (4) of a cation discharge:

$$i_{10} = \text{const.} \times [M^{x+}]^{0.5 \text{ or } 0.8, \text{ etc.}} \quad (4)$$

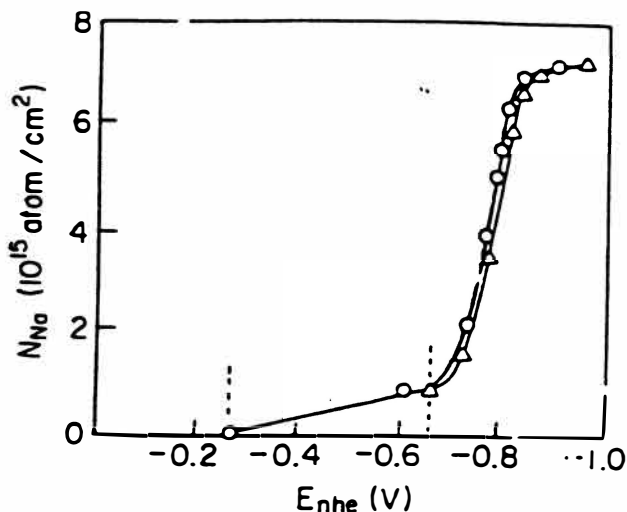
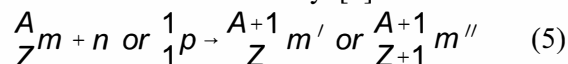


Fig. 1 Relationship between Na atoms/cm<sup>2</sup> of true surface area of Pt and  $E_{nhe}$  in 1mol/l Na<sub>2</sub>SO<sub>4</sub> solutions, pH=4.7(○) and pH=11.38(Δ).

The another result of GST-analysis of the decay process of overvoltage from a steady state to the equilibrium, indicated that the accumulated alkali atoms of M(I) amounted to about  $10^{16}$  atoms/cm<sup>2</sup> of true surface area of a Pt electrode, as shown in Fig.1 [6]. Consequently, alkali atoms have to penetrate into the depth of  $\geq 1,000$  monolayers from the surface, in view of the composition of M(I) and the presence of its diffusion layer.

Hence, taking account of the mechanism of HER as shown above, you can expect the following nuclear reaction to occur easily [3]:



where  $\frac{A}{Z}m$ ,  $\frac{A+1}{Z}m'$ , and  $\frac{A+1}{Z+1}m''$ , and  $\frac{1}{1}p$

denote the nuclei of M(I), of the products of reaction (5) and of an adsorbed or absorbed hydrogen atom H(a), respectively. In general, reaction (5) produces some nuclear particles as the byproducts and then it is thought that some reactions take place among  $m$ ,  $m'$  and these particles in the vicinity in the surface layer of the electrode, as follows:



and  $m$ ,  $m'$ ,  $m''$  and  $m''$ , etc. should become the reactants of the next step. We found a certain amount of tritium simultaneously in the same light water's electrolytes as used for the analysis of nuclear products. At least, the reaction between deuterium atoms,  $d+d$  and/or  $d+d+d$  appeared to be occurring in parallel with reaction (6) [1,7]. Consequently, the mechanism for the nuclear reactions seem to be expressed as a chain reaction which is initiated by the formation of the intermetallic compound M(I).

This paper is concerned with heat evolution and the nuclear products as well as tritium, which were observed in simultaneous or individual experiments.

## EXPERIMENTAL RESULTS

The electrolytic cells used for a series of the experiments were the same as in the previous work. All procedures of the experiments and the accuracies of the measurements were also nearly the same as described previously [8,9].

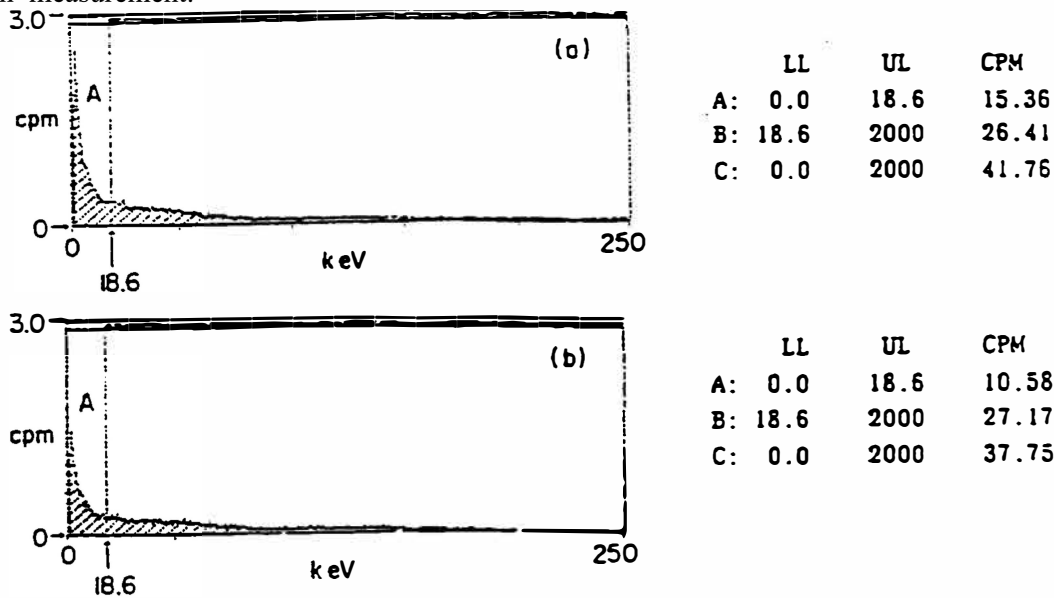
**1. Excess Heat Measurement**

The excess heat which was due to electrolysis in  $K_2CO_3$  light water solution on porous nickel, was found to be in the range from 200 to 400%, in regard to more than 150 observations. In  $Cs_2SO_4$  also light water solution, excess heat of 200 to 300% was observed on platinized platinum. Excess heat on porous nickel in  $Cs_2SO_4$  light water solution might be considerably large as found to be 6,000% estimated by comparison with the foregoing value on platinum, but not reproducible, because porous nickel was often dispersed violently in  $Cs_2SO_4$  solution.

**2. Tritium Measurement**

All experiments concerning the tritium production during electrolysis were carried out in the second basement of a building in order to keep the background value of radiation low and constant. A series of the experiments for the measurement of tritium generated by electrolysis simultaneously with the  $\gamma$ -ray measurements were carried out for 6 hours to 3 days in a dark chamber made of a lead wall of 10 cm thickness equipped with a germanium  $\gamma$ -ray detector, which was settled in an air-conditioned room at  $20 \pm 1^\circ C$ . In this room we couldn't use airflow for cooling the cells, accordingly the observed values of excess heat were slightly small [10]. After electrolysis, the amount of tritium in the electrolyte was measured as described in the previous paper [9].

Typical energy spectra of the radioactivity measurements of the samples prepared by the test electrolyte and control one are shown in Fig. 2. The background value of this scintillation analyzer was found to be  $4.4 \pm 0.4$  cpm in the energy region of emission ray, 0 to 18.6 keV, by use of 1 ml distilled water instead of the subject solution. The background value, which was usually kept very stable, was checked before each measurement.



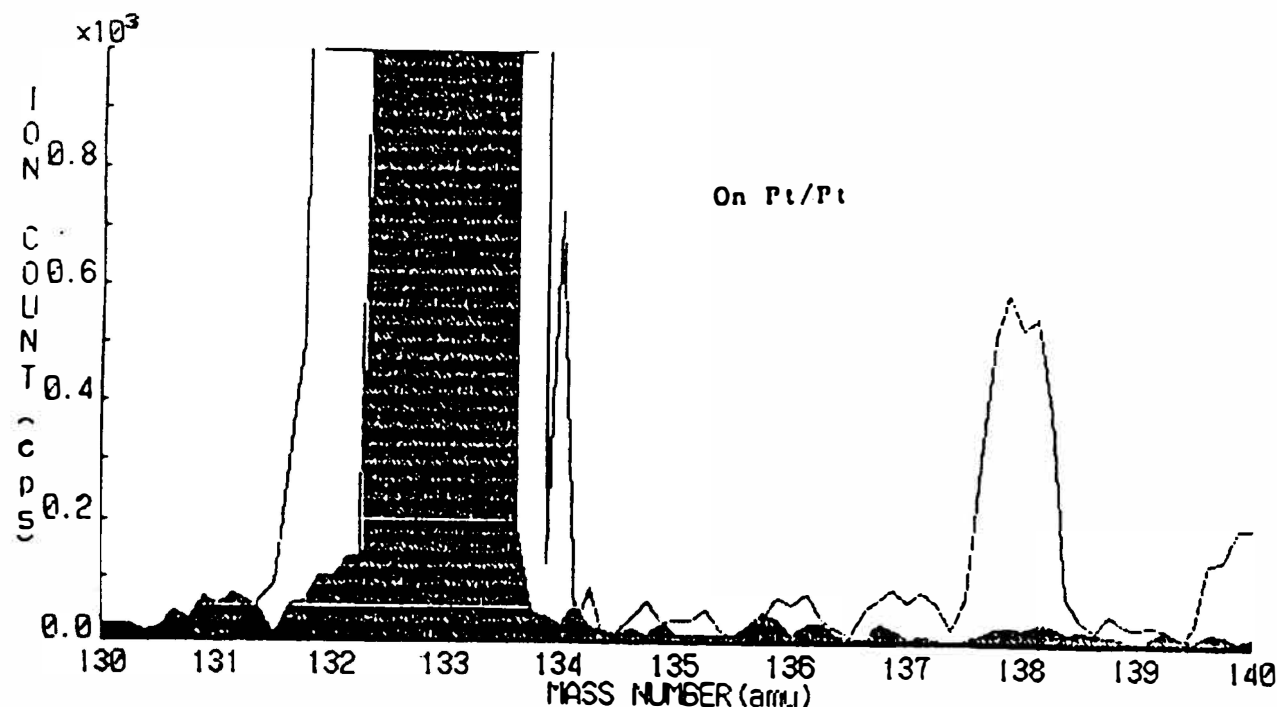
**Fig. 2** Energy spectrum of radioactivity measurements by a liquid scintillation spectrometer: (a) test electrolyte of 0.5 mol/l  $K_2CO_3$  of light water after electrolysis in  $cell_{Ni}$  and (b) control one, region A is due to  $^3t$  with tables of the counts (cpm) in each energy region of interest.

It is apparent from the experimental result observed in  $K_2CO_3$  solutions of light and heavy water [10] that the  $^3t$  amount generated is proportional only to the value of the excess heat, but not the duration of electrolysis and the other factors. If  $^3t$  is the final product of the nuclear reaction, all of the produced  $^3t$  must be accumulated in the electrolyte according to the mechanism shown by Eqs. 1-3, because step (3) is rate determining. Therefore, the above result suggests that  $^3t$  may be one of the intermediates of the nuclear reaction.

### 3. Analysis of Other Products of Cold Fusion

It was confirmed that some nuclear products were generated during electrolysis by use of platinized platinum and porous nickel electrodes, in light water solutions of all alkali metallic ions investigated. For example, calcium of 4 ppm was detected in  $K_2CO_3$  light water solutions by a flame spectrometer [8]. The  $\gamma$ -ray measurement revealed the formation of  $^{24}Na$  during electrolysis in  $Na_2CO_3$  solution. ICP-mass spectra of the electrolytes of  $Cs_2SO_4$  light water showed several peaks of various nuclear products in the region from 132 to 140 amu.

The detection of calcium was described in detail in [8], therefore the present paper is concerned chiefly with the result of the electrolysis of  $Cs_2SO_4$  solution in cells equipped with platinum and nickel electrodes (hereafter, expressed by  $cell_{Pt}$  and  $cell_{Ni}$ ) [11]. Cesium is one of the most appropriate species as a reactant among alkali metals to determine the nuclear products owing to 100% of the natural abundance of  $^{133}Cs$ , as well as  $^{23}Na$ . The above experiment was carried out simultaneously with excess heat measurement. During electrolysis, cells settled in the thermostat box were cooled by airflow, as described in the section of excess heat measurement. After electrolysis, the electrolyte in  $cell_{Pt}$  and  $cell_{Ni}$  as well as the control electrolyte was analyzed by means of ICP-MS (SQR-6500, Seiko Instruments). As the sample for ICP-mass spectrometer, the test electrolyte was diluted quantitatively with pure water 4,000 or 10,000-fold. On the other hand, the control electrolyte was diluted merely 100-fold. Therefore strong contamination with the control electrolyte remained at 133 amu and some others on mass spectra. The shape of the mass spectrum of the residual contamination in the range of mass number from 130 to 140 was not varying during the measurements of the test samples. Fig. 3 shows the typical mass spectrum of test sample of 0.1 mol/l  $Cs_2SO_4$  electrolytes in  $cell_{Pt}$ , which was electrolysed 18.2 hours. The total electricity spent during electrolysis amounted to  $18,000 \pm 200$  coulomb.



**Fig. 3** Mass spectrum of 0.1 mol/l  $Cs_2SO_4$  electrolyte in  $cell_{Pt}$ : duration of electrolysis = 18.2 hours, spent electricity =  $18,000 \pm 200$  coulomb, black shadow indicates residual contamination by the counter electrolyte measurements of the test samples. Fig. 3 shows the typical mass spectrum of the test.

On the other hand, the mass spectrum of the control electrolyte showed only one peak at 138 amu which was found to be 0.4 ppm, except for 133 amu. The amount of the 138 amu species consisted well with the total amount of barium species determined to be 0.6 ppm by a flame spectrometer, in view of the natural abundance of  $^{138}\text{Ba}$  equal to 71.7%.

The amount of the nuclear product of each mass number in the region from 132 to 140 amu was calculated from the area of the peak of mass spectrum of the test electrolyte and compared with that of the control electrolyte, respectively. Thus, the increases of the mass number's species are summarized in Table 1. Concerning the mass numbers 132 and 134 amu the amounts as denoted by + couldn't be determined quantitatively because the resolution of spectra for them was insufficient owing to too large ion counts of 133 amu species, but these amounts seemed to be almost the same as that of 138 amu.

**Table 1** Amount of cold fusion products caused by electrolysis of 0.1mol/l  $\text{Cs}_2\text{SO}_4$  light water solution on platinized platinum.

Mass No.(amu)	: 132	134	135	136	137	138	139	140
$\Delta$ amount(ppm)	: +	+	0.4	0.3	0.5	4.7	0.2	1.8
Expected Species:	Cs Xe	Cs Ba	Xe La	Cs Ba	Cs Ba	Ba	Ba Ce	La Ce
	Ba La	Xe La	Ba Cs	La	La Ce		La	Pr

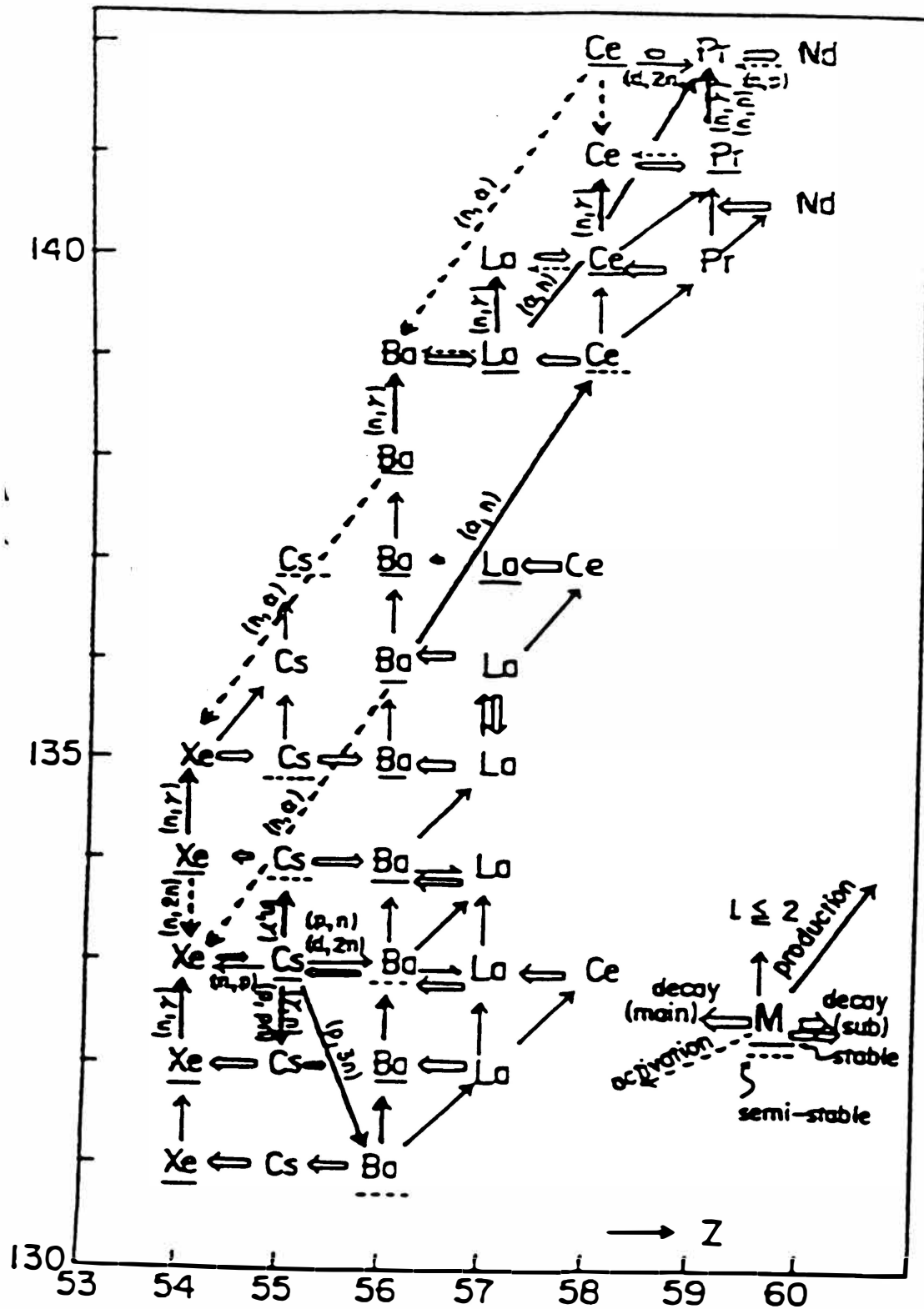
The well-known reactions for nuclear production and activation occurring in a nuclear reactor, were quoted from the literature [12] in the range from 130 to 140 amu and summarized in a diagram (Fig. 4). Moreover, some possible reactions for the changes of nuclei are added in the diagram on the basis of the assumption that the spins  $l$  in the reactions must be less than 2, taking into consideration of the parities and  $Q$ -values. Fig. 4 shows a few possible products of the nuclear reactions at each mass number, which are listed in Table 1.

It was found that cathodic polarization of porous nickel in  $\text{Cs}_2\text{SO}_4$  solution caused its violent dispersion. Such a violent dispersion occurred often, especially when you use a worn electrode.

During fifty hours electrolysis in  $\text{Na}_2\text{CO}_3$  solution of 20 ml by use of a porous nickel cathode, a measurement of  $\gamma$ -ray spectrum was carried out in parallel with that of the excess heat as described at section "excess heat measurement". It was found that there was the peak attributed to the emission of the specific  $\gamma$ -ray from  $^{24}\text{Na}$  at 1368.63 keV in the spectrum and this amount was calculated to be  $1.35 \times 10^{-3}$  Bq from the area of that peak. The spent electricity was 45,000 coulomb. It can be suggest that the reaction  $^{23}\text{Na}(n, \gamma) ^{24}\text{Na}$  is occurring in the surface layer of the cathode by analogy with the radiation reaction occurring in a nuclear reactor.

## CONCLUSION

It was shown by an experimental study that some low temperature nuclear reactions of the intermetallic compounds of alkali metals with neutrons, protons and so forth are occurring in the surface layer of porous nickel and platinized platinum cathodes, during electrolysis in all alkaline solutions investigated as well as  $d+d$  and/or  $d+d+d$  reactions, but the former is predominant.



**Fig. 4** Diagram composed with reactions for nuclear production (bold line), activation (broken one), decay (short or long arrow means slow or rapid) and possible reaction (thin line) as described in the text: Underline (dotted or solid) expresses a stability of each nucleus (half life,  $\tau_{1/2} \geq 10$  days or  $\geq 10^4$  years).

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