

Mass Spectrometry: Critical Aspects of Particles Detection related to Condensed Matter Nuclear Science

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Abstract. The detection of transmutations in Fleischmann and Pons experiments is a critical issue in order to assess the nuclear origin of the excess heat production. A very accurate study in this field has been carried out in ENEA, aimed to investigate the possible occurrence of nuclear transmutation in nickel hydride thin films. The research work, which took several years and involved an international collaboration between many research laboratories, came to the conclusion that the experiments didn't produce any evidence of transmutation experiments, pointing out the importance of using state of the art instrumentation in performing such a kind of study.

1. Introduction

In the last decades some scientific papers have been published on the study of nuclear transmutation reactions in condensed matter, in which the formation of new nuclear products as a consequence of eventual nuclear reactions was investigated. In this contest, a specific research activity based on electrolysis experiments has been developed at ENEA, by using Instrumental Nuclear Activation Analysis (INAA) [1] and Secondary Ions Mass Spectrometry (SIMS) [2, 3]. SIMS is a very powerful technique to approach this problem, since it is able to measure the atomic composition of a material with very high sensitivity, but also the isotopic abundance of each atomic specie, that, if different from the natural distribution, is a solid evidence of the occurrence of a nuclear reaction. Actually, the "appearing" of new elements which were not detected before, in Fleischmann and Pons (F&P) experiments, can be likely caused by contaminations, which are very critical to be ruled out due to the extremely low signals involved in the transmutation investigation.

In this communication, we review some previous results [2,3] concerning the study of nuclear ashes in nickel hydride thin films undergone to electrochemical experiments and report new experimental data which allow to get a definite and coherent interpretation of the complete research work.

2. Experimental methods

The electrochemical cells used in the F&P experiments were made of pure polyethylene. The anode was made of 99.98% Pt wire wound in the form of a flat spiral and the cathode was a thin film of Ni deposited by ion beam sputtering on a polymeric substrate made of the same pure polyethylene material of the cell. During the sputtering process, two identical films were deposited, one to serve as a control while the other was used for electrochemical loading with hydrogen to form nickel hydride. The electrolyte was 1M Li_2SO_4 in pure H_2O (distilled at 18 M Ω). Electrolysis was performed for time periods varying from 4 min to 20 hours. Additional details of the all the experimental setup can be found in previous publications [2, 3].

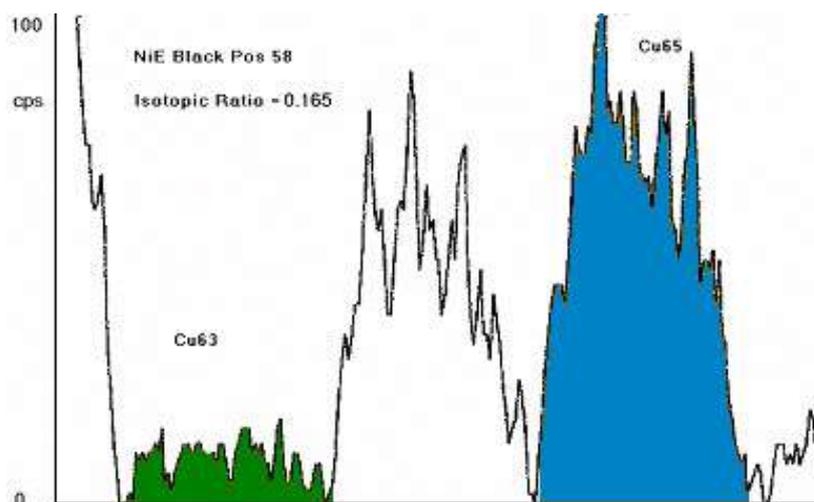


Fig. 1 - SIMS spectrum of the active film in the range of 63-65 nominal masses.

3. Results and discussion

Copper was selected as a marker for the experiment, because it is the most likely product of the reaction between protons and Ni and, in addition, Cu has only two stable isotopes having nominal masses of 63 a.u. and 65 a.u., respectively.

SIMS analysis was performed on the surface of the thin-film cathodes, both on the electrolyzed sample and its relative control one. The used facility was a Quadrupole Analyzer SIMS, which had 0.5 a.u. mass resolution and allowed to analyze both the samples in the same measurements run. Preliminary results showed, “apparently”, a quite big shift of the Cu isotopic ratio (see Fig. 1), being the $^{63}\text{Cu}/^{65}\text{Cu}$ mass ratio equal to 0.165 instead of the natural value of 2.2; in addition, by repeating the analysis in different zones of the sample, the observed discrepancy with the natural value was more significant into the center of the electrode than on the border [2].

In order to confirm the results, the measurements were repeated by a Super SIMS facility, the access to which was granted thanks to the scientific collaboration with the National Research Laboratory (NRL, Washington D.C., USA) [4]. Super-SIMS (also called Accelerator-SIMS) is the combination of Secondary Ion Mass Spectrometry with Accelerator Mass Spectrometry (AMS). The main advantage of this technique compared to conventional SIMS is the virtually complete suppression of molecular background and dark-counts. Detection limits for trace elements can therefore be improved by orders of magnitude and are only limited by contamination and count rates, achieving levels as low as ppb to ppt. The new measurements did not confirm the old ones, reporting a value of the Cu isotopic ratio equal to the natural one, within the measurement accuracy, in both the electrolyzed and the reference samples [3]. Speculations on the possible explanation of the discrepancy between the previous and the new results and on the presence of possible artifacts in both measurements were presented in ref. [3].

The possibility to repeat the measurement with a high resolution Time Of Flight (TOF) SIMS allowed the story to come to a conclusion. The employed instrument was the TOF MiniSIMS manufactured by SAI (Scientific Analysis Instruments), whose used was granted thanks to the collaboration with Assing S.p.A. (Italy). The TOF MiniSIMS has superior mass resolution ($m/\Delta m$ higher than 600 in the mass range of interest), that makes it able to distinguish between single elements and organic compounds. The new analysis revealed that the peak at the nominal mass 65 was constituted by two contributions, the more intense of which was assigned to an organic compound (C_5H_5^+), while the other at 64.93 a.u. well fit $^{65}\text{Cu}^+$ (see Fig. 2, bottom graph). The peak at 63 a.u. was also made by two sub-bands, one at higher mass assigned to C_5H_3^+ and the other at 62.93 a. u. to $^{63}\text{Cu}^+$ (see Fig. 2, top graph). Based on the above identification of the peaks, the mass ratio of the copper isotopes resulted to be 2.0, which well match to the natural value (2.2).

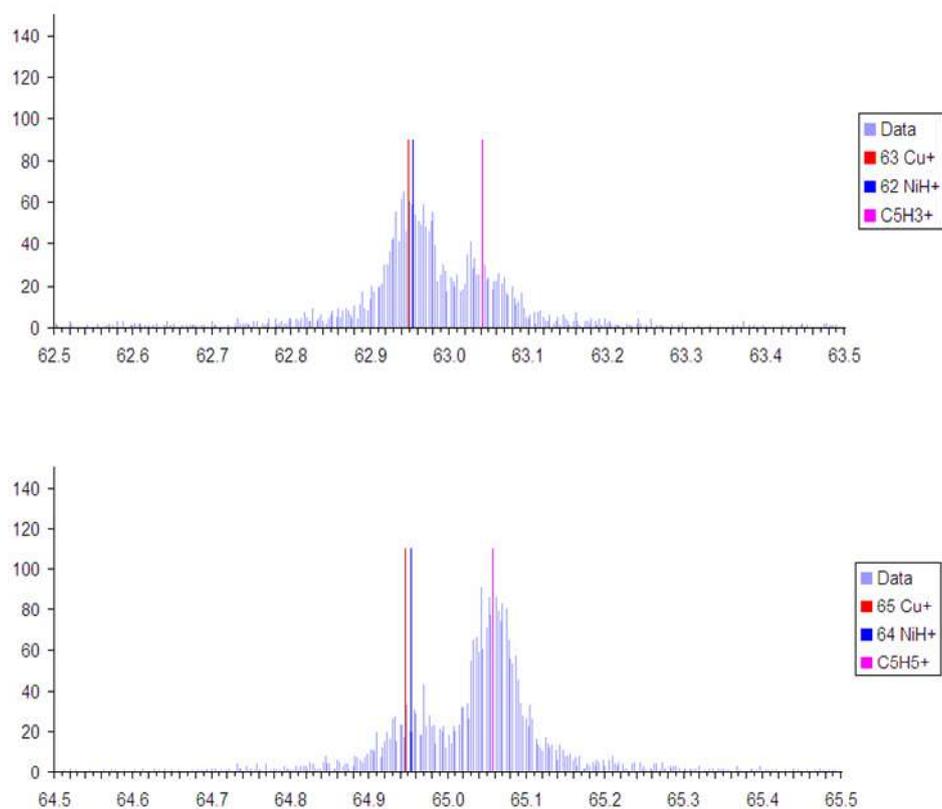


Fig. 2 - TOF-SIMS high resolution spectrum of the active film, in the range of 63 nominal mass (top) and 65 nominal mass (bottom).

The organic compounds detected by the TOF MiniSIMS were probably produced during the SIMS beam bombardment of the sample, emerging from the zones of the polyethylene substrate where the nickel film was more damaged. The higher the exposed area of the polymeric substrate (in the middle of the sample, because of the film micro-cracks) the larger the “apparent” shift.

The reason why the measurement performed by using SUPER-SIMS did not revealed the organic compound, although the instrument had a very high mass resolution, is that the high energy of the impinging ions destroys any organic compounds which may be produced. This did not happen in the case of TOF MiniSIMS and Quadruple SIMS used in the first analysis, which both have the energy of the primary ions in the range of a few keV.

4. Conclusions

In conclusion, after the first experimental campaign we repeated two times the isotopic composition measurements, on the same samples, by using two high performance SIMS. The new analysis revealed that the Cu isotopic ratio was the natural one and that the apparent shift was due to the organic compound C₅H₅. Then, the apparent isotopic shift was an artifact produced by the not enough resolution of the SIMS used in the first measurement performed in 2003. Then the experiments didn't produce any evidence of transmutations.

The work highlighted the importance of using status of the art instrumentation in performing transmutations studies.

5. References

- [1] P. Avino, E. Santoro, F. Sarto, V. Violante, A. Rosada, “Neutron activation analysis for investigating purità grade of copper, nickel and palladium thin films used in cold fusion experiments”, *J. Radioanal. Nucl. Chem.* (2011) DOI 10.1007/s10967-011-1296-3.
- [2] V. Violante, M.L. Apicella, L. Capobianco, F. Sarto, A. Rosada, E. Santoro, M. McKubre, F. Tanzella, C. Sibilìa, “Serch for Nuclear Ashes in Electrochemical Experiments”, *Proc. of 10th Int. Conf. on Cold Fusion (ICCF-10)*, Cambridge, Massachusetts, USA, World Scientific Inc. (2003), pp. 405-420.
- [3] M. Apicella, E. Castagna, G. Hubler, M. McKubre, F. Sarto, C. Sibilìa, A. Rosada, E. Santoro, F. Tanzella, V. Violante, “Progress on the Study of Isotopic Composition in Metallic Thin Films Undergone to Electrochemical Loading of Hydrogen, *Proc. 12th Int. Conf. on Cold Fusion (ICCF12)*, Yokohama, Japan, (2005), pp. 264-271.
- [4] Accelerator Super-SIMS facility at the Swiss Federal Institute of Technology Zurich (ETH, Zurich, Switzerland), by Dr. M. Doebely.
- [5] Millbrook ToF MiniSIMS Analysis Report No 334, Analyst: Dr John Eccles; www.assing.it