



22nd International Conference on
Diffusion in Solids and Liquids
22 TO 26 JUNE 2026 | RHODES, GREECE

ABSTRACT:

Gaseous and Electrochemical Hydrogen Insertion Linked by Impedance Spectroscopy

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Electrochemical modulation methods for investigating hydrogen diffusion in metallic membranes are well-established due to the simplicity of their experimental procedures and since they do not need to measure the absolute flow [1]. We adapted the electrochemical impedance method to measure hydrogen permeation through membranes by adding a modulating dead-volume device to an existing membrane permeation setup [2], using a simplified volumetric method originally proposed by Altunoglu et al. [3]. We demonstrate that this volumetric impedance spectroscopy is superior to steady-state methods, particularly for distinguishing diffusion from non-diffusional properties. We provide a quality parameter based on the frequency dependence (the Montella condition [1]). Experiments on hydrogen permeation in archetypal Pd membranes corroborate the feasibility and advantages of the modulation method.

Finally, we address the possibility of applying the method to measure gases other than hydrogen, e.g., CO₂ and CH₄. This is a particular strength of a volumetric method, as electrochemical methods are restricted to conductive membranes and to gases that can be generated/affected by electrochemistry. In fact, modulated gas experiments may even be applied to catalysis [4].

[1] C. Montella, *J. Electroanalytical Chem.* 480, 166 (2001).

[2] A. Borgschulte and C. P. Canales Oyarzo, *Int. J. Hydr. Energy*, in press (2026)

[3] A. Altunoglu and N. Braithwaite, *J. Alloys Compds.*, 231, 302 (1995).

[4] A. Borgschulte, M. Achermann, and M. Nikolic, *J. Phys. Chem. Lett.* 15, 10451 (2024).