

# Changes of the breaking strain from content of tritium and cycling in a palladium cathode membrane

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## ABSTRACT

In this work, it is examined the breaking strain in the function of the tritide phases, of depth and surfaces of silver alloyed or not alloyed palladium cathode membrane in a sequential process for penetration of tritium. Examining the parameters, a mapping model is given. Firstly, the introduction of tritium in the membrane by electrolytic cracking of tritiated water with cycling leads to high constant breaking strain in the adsorption surface and an increase of the breaking strain in the desorption surface. The tendency towards numerous cracks formation on the two surfaces is indisputable. Secondly, a progressive decrease of breaking strain in the depth of membrane, independent of cycling, signifies decelerating cracks and fractures in the volume of palladium. Before shutdowns of electrolysis and temperature, tritium purging process is recommended to avoid inversion of phase in entry and exit surfaces then collapse to increase the life of the palladium membrane. Porous ceramic support prevails on the life of palladium membrane.

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## 1. Remember and more centering of specifications

There are two tritium-charging techniques in metals as indicated by Lässer [1]. Firstly, charging tritium on cathode metals from tritiated water is an easy procedure, which is in regard of this paper. The second technique is the production of metal tritides and hydrides directly from the gas phase. These two techniques: from tritiated water or gas lead to a pure mixture of hydrogen isotopes to be separated. Concerning the cathode charging, the paper entitled “Optimization for the tritium isotope separation factor and permeation by selecting temperature and thickness of the diffusion Pd–Ag alloy cathode” [2] and the publication [3] give an aspect of cracking, surface blistering, thickness stretching with width shrinking, of hardening and solubility tritium and internal pressure and  $\beta \leftrightarrow \alpha$  phases changing by electrolysis cycling. The problem remaining in this subject is the aspect of the breaking strain due to tritium responsible of stress corrosion in the alloyed or not alloyed cathode. In this topic, maintaining availability of operating industrial electrolyzer is strategically important in response to demand for recycling the tritium from tritiated water. Therefore, it is imperative to obtain relevant data combined with a predictive framework to understand the breaking strain changing. To clarify how cracks

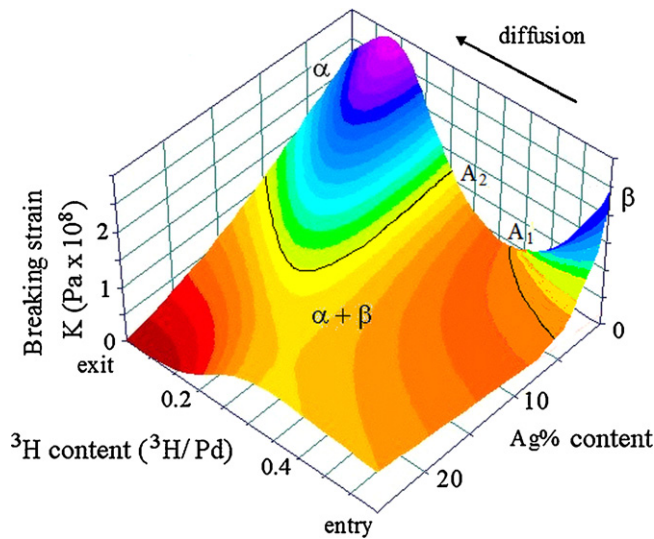
evolve, we chose to study the worse situation in taking pure palladium no-alloyed, not the more resistant palladium-25%silver alloy. Results obtained lead to write this complementary Short Communication to bring more details on the protection of the diffusion electrode in complementary to Bellanger [2].

In the present section of report, we remember engineering items and data for palladium-25%silver alloy and pure palladium cathode industrial electrolyzer. Results of stress are given in the next section “results and specification” using the worse situation with pure palladium. For evidenced the situation, phase diagram of palladium and Pd–Ag alloys in function of stress is given in Fig. 1 and Table 1 obtained using the ETBX digital grabber program (Engrasp™). As seen, stress depends on the silver content and the tritium concentration in the metal. In Fig. 1, it can be seen for pure palladium, the  $\alpha$ - and  $\beta$ -phases are present with high local breaking strain in the entry and exit surfaces where the tritium concentration are respectively high and low. In contrast, with increasing the silver content, breaking strain decreases with a reduced region of the  $\alpha$ - and  $\beta$ -phases encouraging the coexisting  $\alpha$  and  $\beta$  phase formation. At 25% silver, the  $\alpha$ - and  $\beta$ -phases disappear. There appears a bi-model of cracking at each surface side, thereby the compelling evidence to introduce silver in the metal base. In the published report [2], palladium-25%silver alloy cathode membrane electrolyzers have proven to be highly effective in process recycling tritiated water. These are a technology of choice in the nuclear industry for providing maximum tritium purity compared to any other technology using iron or uranium cracking with chromatog-

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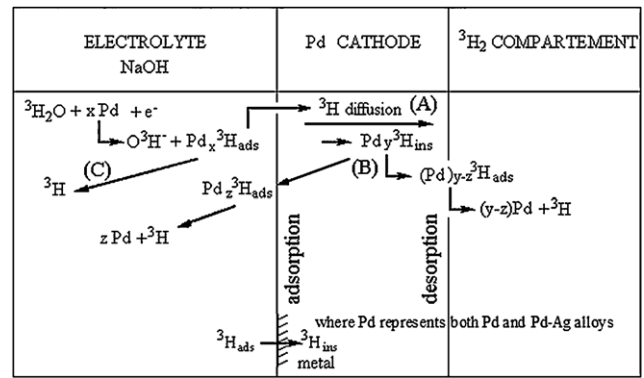
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**Fig. 1.** Diagram representing the solubility gradient of tritium with repartition of  $\alpha$ ,  $\beta$  and  $\alpha + \beta$  phases and breaking strain through relative palladium membrane thickness in function of silver content in the palladium, – lines  $A_1, A_2$ : limit of phases – entry: adsorption face for tritium in the cathodic membrane, exit: desorption face for tritium in the gastight compartment for recuperation after diffusion.

raphy separation and palladium–silver alloy diffuser purification as independent three steps complicating the tritium process [4–7]. The palladium–silver alloy cathode membrane is typically a hollow finger possessing the unique property of allowing both the cracking of tritiated water and the diffusion of tritium to pass through its crystal lattice. In Fig. 2, the tritiated water coming into contact with the outside side cathode membrane surface is cracked into monatomic tritium to pass through the membrane. On the inside surface of the membrane, the monatomic tritium is recombined into molecular tritium. The mechanism of tritium involves a series of steps: (1) cracking of tritiated water, (2) adsorption of tritium in the cathode, (3) its atomic ionization, (4) its ionic diffusion, (5)



**Fig. 2.** Diagram of the cathode membrane.

molecular arrangement of tritium in the inside surface (exit) and (6) desorption. Within the palladium or palladium–silver alloy, tritium loses its electron to the metal structure and diffuses through it as an ion (triton). Various examples of technical evidence have been advanced to substantiate this mechanism in references [8–10]. Only tritium and its isotopes possess the ability to diffuse through the metal. Assuming no pinholes or micro-cracks, the tritium issuing to the inside side of a membrane may be looked upon as of absolute purity. Analysis to detect the presence of impurities show only traces of contaminants in the parts-per-billion range (<1 vpm). These trace impurities probably reflect an incomplete outgassing of the inside walls of the downstream of parts of the system. The ultrapure tritium and hydrogen recuperated can be used in the ITER process (International Thermonuclear Experimental Reactor). Integrity of the cathode shaped as a hollow finger (Fig. 3) is further enhanced with the insertion of pre-cleaned porous ceramic shaped as the cathode (Fig. 4A). In Fig. 4B, it can be seen deformation of palladium in absence of porous ceramic. To make the finger shape, hammering of a palladium–silver alloy foil was at first used, but this technique was quickly discarded because embossing (Fig. 3B) of

**Table 1**  
Breaking strain distribution in the membrane with phases in function of silver content and tritium content—essential results through the membrane, in the bulk of membrane, near the inside surface and near the outside surface.

| Phase                      | Silver effect            |      |      |      |      |                            |                      |                  |      |      |      |                            |                      |      |      |      |      |
|----------------------------|--------------------------|------|------|------|------|----------------------------|----------------------|------------------|------|------|------|----------------------------|----------------------|------|------|------|------|
|                            | Near the inside surface  |      |      |      |      |                            |                      |                  |      |      |      |                            |                      |      |      |      |      |
|                            | $\alpha$                 |      |      |      |      |                            |                      | $\alpha + \beta$ |      |      |      |                            |                      |      |      |      |      |
| Ag%                        | 0                        | 2    | 4    | 6    | 8    | 10                         | 12                   | 14               | 16   | 18   | 20   | 22                         | 24                   |      |      |      |      |
| H content                  | 0.05                     | 0.05 | 0.05 | 0.05 | 0.05 | 0.05                       | 0.05                 | 0.05             | 0.05 | 0.05 | 0.05 | 0.05                       | 0.05                 |      |      |      |      |
| Stress (Pa $\times 10^8$ ) | 2.53                     | 2.39 | 2.07 | 1.77 | 1.45 | 1.15                       | 0.91                 | 0.67             | 0.56 | 0.44 | 0.35 | 0.27                       | 0.23                 |      |      |      |      |
| Phase                      | In the bulk of membrane  |      |      |      |      |                            |                      |                  |      |      |      |                            |                      |      |      |      |      |
|                            | $\alpha + \beta$         |      |      |      |      |                            |                      |                  |      |      |      |                            |                      |      |      |      |      |
| Ag%                        | 0                        | 2    | 4    | 6    | 8    | 10                         | 12                   | 14               | 16   | 18   | 20   | 22                         | 24                   |      |      |      |      |
| H content                  | 0.3                      | 0.3  | 0.3  | 0.3  | 0.3  | 0.3                        | 0.3                  | 0.3              | 0.3  | 0.3  | 0.3  | 0.3                        | 0.3                  |      |      |      |      |
| Stress (Pa $\times 10^8$ ) | 0.8                      | 0.75 | 0.68 | 0.62 | 0.57 | 0.53                       | 0.5                  | 0.48             | 0.46 | 0.46 | 0.46 | 0.46                       | 0.46                 |      |      |      |      |
| Phase                      | Near the outside surface |      |      |      |      |                            |                      |                  |      |      |      |                            |                      |      |      |      |      |
|                            | $\beta$                  |      |      |      |      |                            |                      | $\alpha + \beta$ |      |      |      |                            |                      |      |      |      |      |
| Ag%                        | 0                        | 2    | 4    | 6    | 8    | 10                         | 12                   | 14               | 16   | 18   | 20   | 22                         | 24                   |      |      |      |      |
| H content                  | 0.5                      | 0.5  | 0.5  | 0.5  | 0.5  | 0.5                        | 0.5                  | 0.5              | 0.5  | 0.5  | 0.5  | 0.5                        | 0.5                  |      |      |      |      |
| Stress (Pa $\times 10^8$ ) | 1.79                     | 1.57 | 1.34 | 1.14 | 0.99 | 0.86                       | 0.77                 | 0.71             | 0.67 | 0.64 | 0.63 | 0.62                       | 0.62                 |      |      |      |      |
| Phase                      | Through the membrane     |      |      |      |      | Phase                      | Through the membrane |                  |      |      |      | Phase                      | Through the membrane |      |      |      |      |
|                            | $\alpha$                 |      |      |      |      |                            | $\alpha + \beta$     |                  |      |      |      |                            | $\alpha + \beta$     |      |      |      |      |
| Ag%                        | 0                        | 0    | 0    | 0    | 0    | Ag%                        | 12                   | 12               | 12   | 12   | 12   | Ag%                        | 25                   | 25   | 25   | 25   | 25   |
| H content                  | 0.1                      | 0.2  | 0.3  | 0.4  | 0.5  | H content                  | 0.1                  | 0.2              | 0.3  | 0.4  | 0.5  | H content                  | 0.1                  | 0.2  | 0.3  | 0.4  | 0.5  |
| Stress (Pa $\times 10^8$ ) | 2.53                     | 1.33 | 0.87 | 1.21 | 1.79 | Stress (Pa $\times 10^8$ ) | 1.15                 | 0.98             | 0.79 | 0.62 | 0.83 | Stress (Pa $\times 10^8$ ) | 0.56                 | 0.46 | 0.53 | 0.62 | 0.64 |

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