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Methods and Preliminary Results about Microscopic Hydrogen Diffusion in Metals

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Abstract

The final purpose of this research is temporal and spatial analyze of diffusible hydrogen distribution for welding joints. This paper presents the results of the initial research concerning quality and quantity analyze of diffusible hydrogen distribution in metals. The experiments study diffusible hydrogen distribution on microscopic level, on surface of granule and also on space between granules, for an OL37 piece test electrolytic filled, using a performing microscope with immersive objective. On piece test was added one single cider oil drop, witch have hydrogen capture properties and also this race the rate of microscope visibility.

Key words: *hydrogen diffusion, granule limit, metallic piece test, microscopic observation, welding*

General considerations about microscopic hydrogen diffusion

Referring to aleatory displacement theory, the hydrogen diffusion means individual gallop of the diffusible atoms in to the solvent crystalline network atoms.

The hydrogen in easier solve in metals and diffuse by interstitial mechanism. In this mechanism, the atoms of the solve element jump from a place at network in another, with implications above tension local stage. If the interstitial atom does not "wait" to be taken by a network trouble (vacancy, granule limit, dislocation) for jump, the diffusion coefficient can be relative big [2].

In the real network shows interactions of diffusion element with the network troubles, in scope to temporarily retain or longer retain of this, in function of connection energy value – fig.1.

The most frequent diffusion coefficient measurement methods are based by relaxation techniques or degas methods. For interstitial elements, on infinite dilution, the displacement of atoms make independent, by interstitial mechanism – fig.2.
Grace to surrounding atoms, the activation energy is more and more big and the rate of diffusion is relatively low. For all that, the atoms can diffuse along granule limits, interfaces and free surfaces (creaks, voids) of material [3].

The atoms diffuse easily on granule limits because of cracks pack witch determine large spaces between atoms. Because of preference diffusion ways generate by miss order of atoms from this area, the activation energy is low and the diffusion coefficient is bigger. One small activation energy determine the race of diffusion coefficient an also of interstitial element flow, because a small quantity of external thermal energy is necessary for cross the energetic removal barrier.

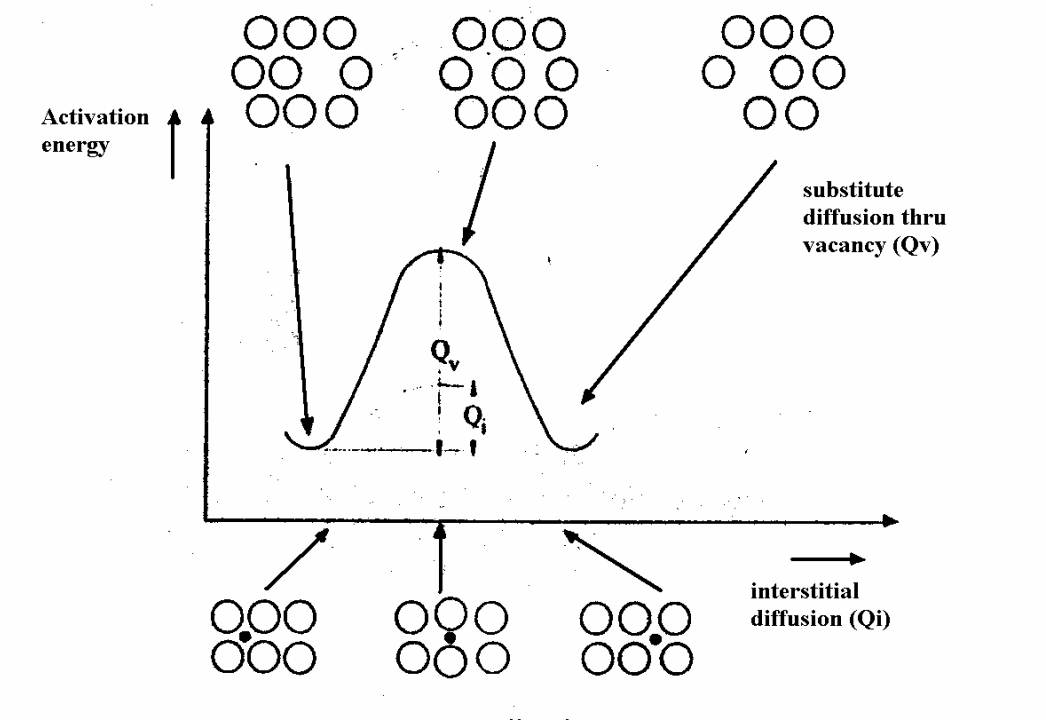

Fig.1. Displacement of interstitial atoms versus activation energy [3]

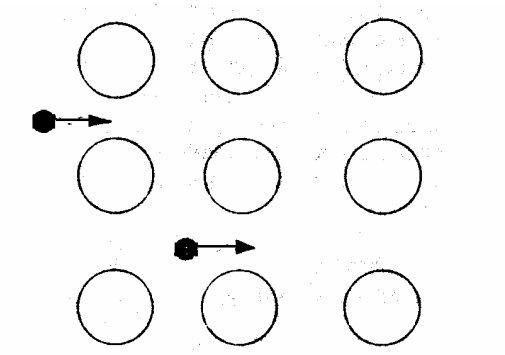

Fig. 2. The interstitial diffusion mechanism [3]

The interstitial diffusion, with low activation energy, is usually with one order bigger than vacancy diffusion or substitution diffusion. The activation energy is low for interstitial atoms solved in metals with low melting temperature.

In the particularly case of welding seam and thermo mechanical influence zone, the grained can be different for very small distances (tens of μm), witch means the diffusion to be much more affected. A microscopic structure compose by more granules will contain a big number of diffusion areas (granule limits, witch represent a big energy zone) resulted by faster and inefficient atoms packing. The local energy can be reduced by reducing granules surfaces, that is the race of grained.

The race of grained means granule limits movement, witch permitted some granule to grow up between other. The diffusion along granule limits is necessary in the granule race process.

Consecutively, the race of granule limits number is tie by activation energy witch means some atoms can jump over energetic barrier represented by that. So, for a material, by racing of granulation the diffusion is accelerating, thanks to decrease of energy on granule limits level.

Experimental determination of electrolytic filled level for OL37 piece test

This experiment was necessarily to follow the time evolution of hydrogen diffusion process, on macroscopic level, for OL37 piece test, with parallelepiped configuration, having the next dimensions: 60 mm length, 10 mm width and 20 mm height. For this purpose was designed an experimental stall, presented in the next paragraph.

Experimental stall

The experimental stall for electrolytic filled on homogeneous OL37 piece test is presented in figure 3. This composed by: glace recipient; 8 graffito elements, serial connected on c.c. power source with 2,5V voltage and 0,7A current; OL37 piece test connected with graffito elements; the power source is presented in figure 4.

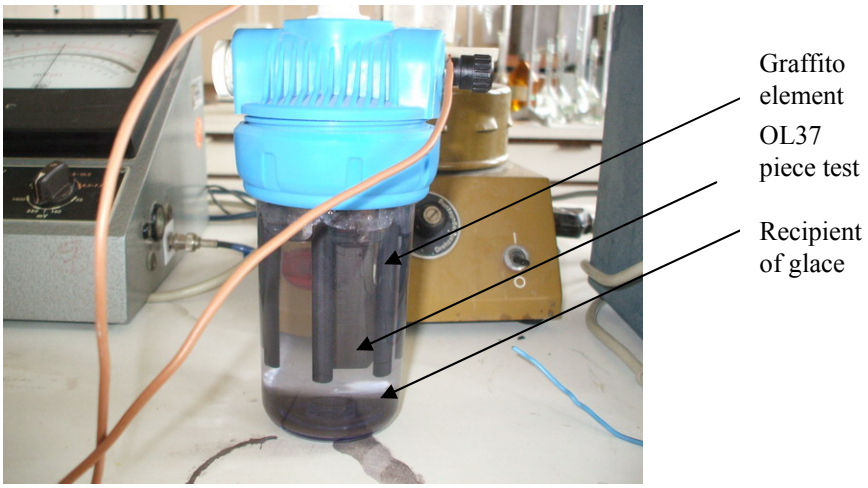

Fig.3. Experimental stall

Fig.4. Power source of experimental stall

To realize electrolytic process on consider OL37 piece test like anode and the cathode is graffito electrodes battery. The piece test is immersing in distillation water solution.

This electrolytic filled was realized in approximate 48 hours.

Quality verification of hydrogen filled for piece test

On the end of filled period, the piece test was cleaned and submersed in a recipient with glycerin, like in figure 5. After 5 minutes, the first balls of hydrogen begin to appear on glycerin surface.

The observations were made for 72 hours. Based on these, we can say that for the first 48 hours, the diffusion was accelerated, corresponding for an exponential function. Until that, the diffusion was deferred, the hydrogen diffuse much more slowly, constantly, until a minimum level.

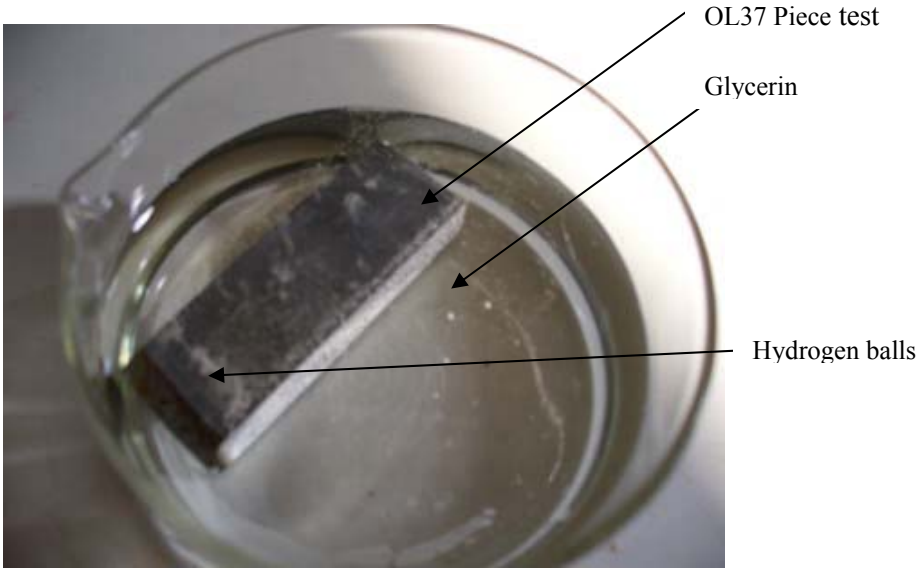

Fig.5. Quality test with glycerin

The experiment was repeated more times, every time was obtained the similar results.

Microscopic observations about hydrogen diffusion on granular and inter granular level

After the macroscopic observations and quality determinations, was appropriate the microscopic evaluation of hydrogen distribution. We know the zones with big concentration of hydrogen inside the metal, is the most important causes of micro cracks.

The microscopic observations were made for the same homogenous OL37 piece test, with parallelepiped configuration, having the next dimensions: 60 mm length, 10 mm width and 20 mm height.

To realize this objective, were necessarily to following the next steps:

- effect metallographic luster for one of piece faces;
- electrolytic filled for piece test using the presented experimental stall;
- fast piece test extraction from electrolytic bath, drying and metallographic attack with 2,3% HNO_3 to distinguish the microscopically structure of that;
- microscopic observation using a performing microscope with immersive objective: for preparing the face we applied a small cider oil drop, witch have the propriety to capture the hydrogen balls and also to race the rate of microscope visibility.

The observations were made for 72 hours. All the pictures were captured with a video camera attached by the microscope. These images were made for different moments of time, shown for witch image. In figure 6 is presented the metallic structure for the moment *time = 30 minutes*. On every image is attaching one measure scale witch we can approximate the dimension of granule and the number of hydrogen balls on surface of granule and also to the limit between granules.

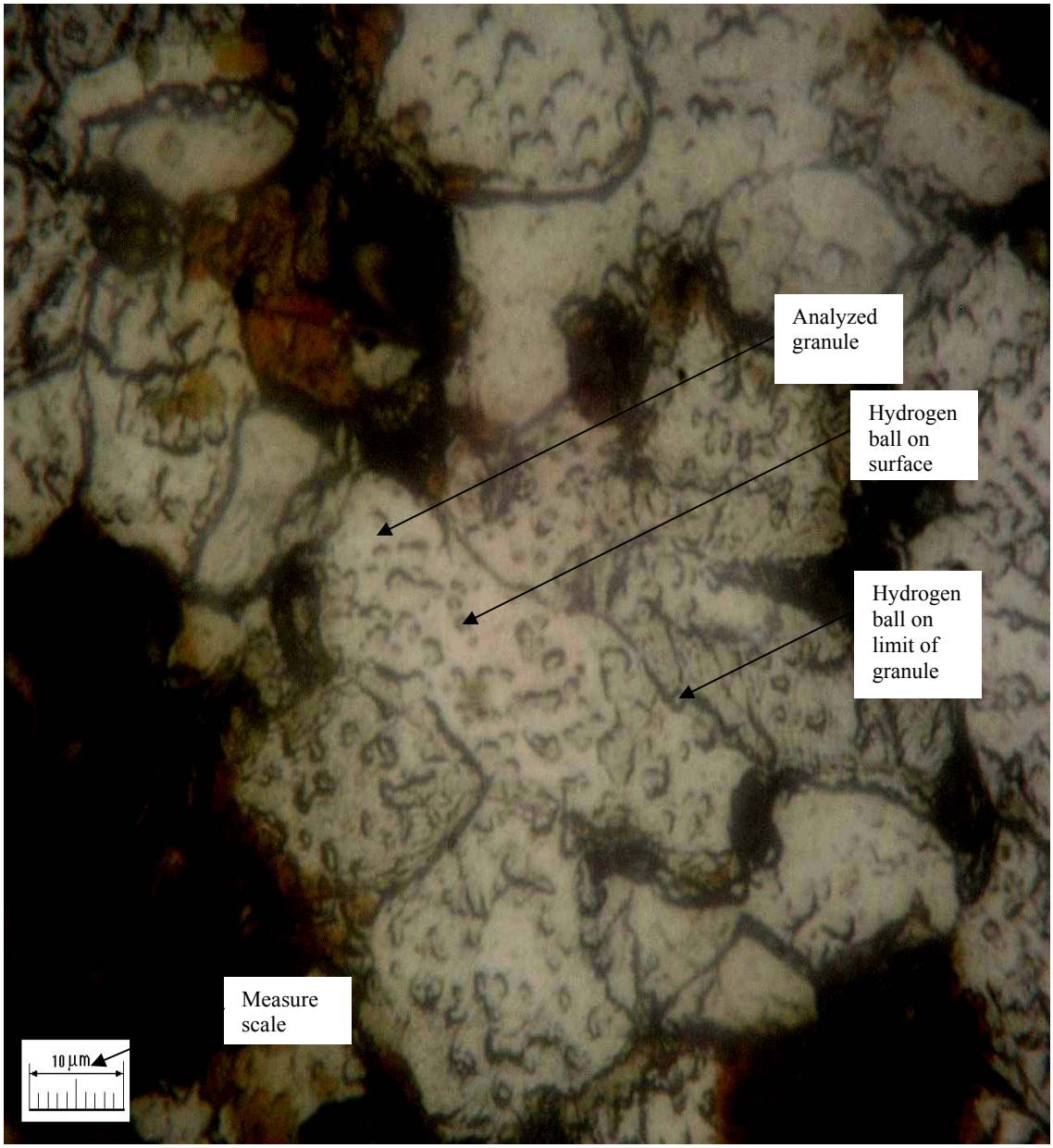

Fig.6. Microscopic image for time = 30 minutes

Determination of hydrogen concentrations on surface and inter granular space for a metallic granule

As a result of microscopic observation we can approximate the dimensions of granule like this: 60 μm – length, 14 μm – width and 2 μm - the ball hydrogen radius; inter granular volume 46,472 x 10^{-6} μm^3; inter granular weight – 30,98 x 10^{-26} g; surface granule – 840 μm^2.

First, we can approximate the number of the hydrogen balls diffused on surface of granule and also to the limit between granules. The results are presented in table 1.

Table 1. The number of hydrogen balls on surface and to the limit between granules

Time [min]	**On surface granule**	**On inter granular space**
0	21	26
5	38	28
15	49	34
30	51	55
60	60	60
120	62	65
360	67	73
720	72	81
1440	73	84
2880	74	86
4320	76	86

Then, we determine the hydrogen concentration values like rapport between number of hydrogen balls multiplied with volume of hydrogen ball and weight of a prism with 4 μm high and the indicate granule surface indicated in figure 6, corresponding for 100 grams of metal. I mentioned that the high of that prism is equal to hydrogen ball diameter. We obtained the value of prism volume like 3360 μm^3. The weight of prism is approximate 224 x 10^{-19} g. In the same way, we calculate the hydrogen concentration for inter granular space, considering that space like cylinder ring, with length equal to perimeter of granule and ring width equal to hydrogen ball diameter.

The values of hydrogen concentrations on surface and to the limit between granules are presented in table 2.

Table 2. The concentration of hydrogen on surface and to the limit between granules

Time [min]	**On surface granule [cm^3/100g]**	**On inter granular space [cm^3/100g]**
0	3,15	7,02
5	5,7	7,56
15	7,35	9,18
30	7,65	14,85
60	9	16,2
120	9,3	14,75
360	10,05	19,7
720	10,8	21,8
1440	10,95	22,68
2880	11,10	23,22
4320	11,40	23,22

Conclusions

As the results shown, the hydrogen diffusion is more intense on inter granular space then surface granule. On mathematical side, the presented results are in concordance with the quality observation from glycerin experimental work shown in this paper. Considering the diffusion equation for a homogenous and isotropic corps, we can observe that variation speed of concentration, for a know point, is directly proportionally with the diffusion coefficient value.

In the future, using these dates, we will determine the volume concentration of granules with finite element method.

The results of these experiments will be used for determination the microscopic hydrogen concentration on limit between welding seam and thermo-mechanic influence zone.

References

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Metode şi rezultate preliminare privind difuzia microscopicã a hidrogenului în metale

Rezumat

Această lucrare prezintă rezultatele cercetării inițiale privind distribuția hidrogenului difuzibil în metale, ştiind că zonele de concentrație maximă a acestuia, reprezintă unul din principalii factori de producere pentru fisuri şi microfisuri. Experimentele pun în evidență distribuția hidrogenului la nivel microscopic, pe suprafața acestuia şi pe spațiul intergranular, pentru o epruvetă din OL37, încărcată electrolitic. Pentru analiza microscopică, pe epruvetă a fost adăugată o picătură de ulei de cedru, acest ulei având proprietatea de a capta bulele de hidrogen difuzibile şi totodată de a mări coeficientul de vizibilitate al microscopului cu obiectiv cu imersie. Utilizând imaginile microscopice, cu ajutorul unei scale de măsurare, a fost determinată concentrația hidrogenului pe suprafața unui grăunte şi pe limitele sale. Rezultatele experimentului vor fi extinse, ulterior, pentru determinarea concentrației de hidrogen la limita dintre cusătura sudată şi zona de influență termo-mecanică.