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Direct Observation of Hydrogen Distribution in Pearlite

Yi-Sheng (Eason) Chen[1,2,*], Ranming Niu[1,2], Pang-Yu Liu[1,2], Patrick Burr[3], and Julie Cairney[1,2]

[1.] Australian Centre for Microscopy and Microanalysis, The University of Sydney, Sydney, Australia.
[2.] School of Aerospace, Mechanical and Mechatronic Engineering, The University of Sydney, Sydney, Australia.
[3.] School of Mechanical and Manufacturing Engineering, University of New South Wales, Australia
* Corresponding author: yi-sheng.chen@sydney.edu.au

Hydrogen, as a zero-carbon fuel, plays a significant role in a future decarbonized energy portfolio. It would be economically attractive if hydrogen can be used in the current natural gas infrastructure. However, existing gas pipelines are made of steels that are susceptible to hydrogen embrittlement. Failure due to hydrogen embrittlement can lead to catastrophic consequences. As such, understanding how hydrogen embrittles steels is paramount for developing a mitigation strategy to enable the energy transition from fossil fuels to clean hydrogen.

Pipeline steels generally contain a significant amount of pearlite, which is comprised of cementite and ferrite (Figure 1A and B). The interface of the two phases has generally been thought to be where hydrogen trapping and crack initiation take place, despite the high interfacial coherency (Figure 1C) [1,2]. However, there has been no direct observation of the distribution of hydrogen in the vicinity of these interfaces to support such hypothesis. Here we used atom probe tomography (APT) to observe hydrogen in the steel, by using a combination of a cryogenic sample transfer method and deuterium charging to circumvent the diffusion loss and the hydrogen ambiguity, respectively, in the APT vacuum chamber [3]. The result (Figure 2A) suggests that the deuterium (red) is trapped within cementite which is delineated by the blue isoconcentration surfaces of 13% carbon. Furthermore, our data exhibits a hydrogen depletion zone near the ferrite-cementite interface, as green-shaded in Figure 2A. This result suggests the lower tendency of hydrogen to reside at the interface than in cementite bulk and ferrite matrix, which contradicts the general hypothesis of the interfacial trapping of cementite [2].

To further understand the hydrogen trapping mechanism, DFT calculations (Figure 2B) were performed to obtain the lowest energy states of hydrogen around a ferrite-cementite interface. Our model considered the presence of elastic strain in both phases, which is required for the formation of a coherent interface and was experimentally confirmed by a geometric phase analysis of the high-resolution TEM image in Figure 1C (data not shown). Our calculation also considered the presence of carbon vacancies in cementite, which can be confirmed in the APT compositional analysis (data not shown) and have a significant role for trapping hydrogen solutes [1]. As a result, we found the hydrogen in ferrite tends to migrate toward the interface due to the expansion of the ferrite lattice (black solid line in Figure 2B). Subsequently, hydrogen at the interface can be attracted into cementite bulk, enhanced by the presence of decreasing compressive strain toward the interior of cementite (red broken line in Figure 2B). Moreover, hydrogen absorption by cementite is energetically more favored in the presence of carbon vacancies in cementite (blue and green broken lines in Figure 2B). The modeling rationalizes the APT observation of hydrogen depletion at the interface and the trapping in bulk cementite.

In summary, our observations clarify that cementite plays a dominant role in the hydrogen trapping in pearlite, providing a new insight for the design of new pipeline steels that can better withstand hydrogen embrittlement.

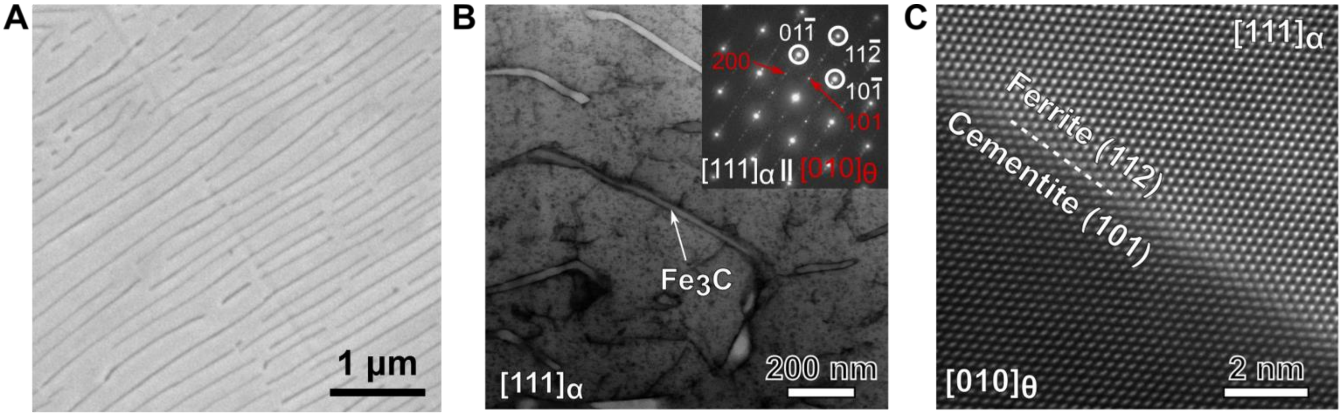

Figure 1. (A) SEM micrograph of the pearlitic steel specimen containing cementite (dark linear features) in a ferritic matrix. (B) Bright-field TEM micrograph of the cementite in the ferrite matrix and the diffraction pattern (inset) which indicates the crystal orientation relationship of the two phases. (C) High-resolution TEM micrograph of the ferrite-cementite interface and their respective cohesive planes.

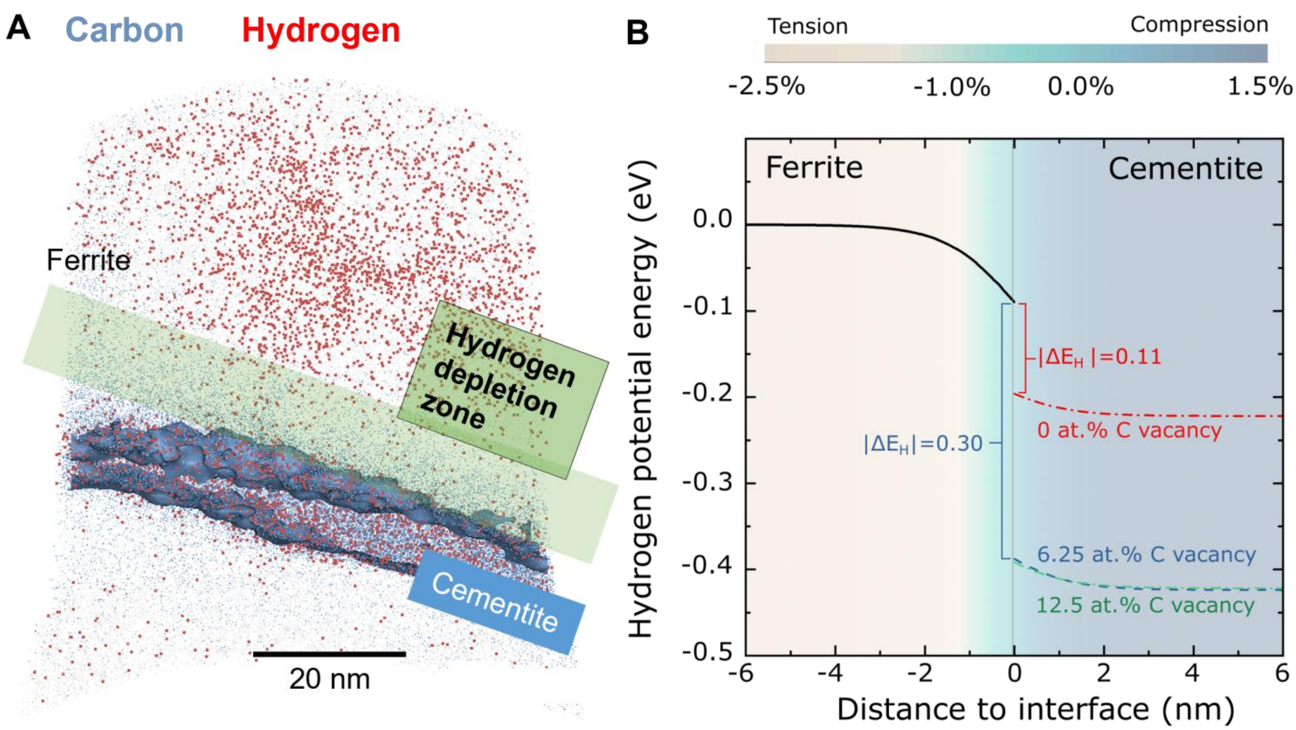

Figure 2. (A) A 10-nm 2-dimentional slice from a 3-dimentional atom map of a hydrogen charged pearlite APT specimen. Blue isoconcentration surfaces delineate the carbon-rich region, indicating the location of cementite in the specimen. Red points are deuterium. Green shade highlights a hydrogen-depletion zone between the ferrite and cementite regions. Deuterium signals are used to represent hydrogen in this dataset. (B) DFT calculation of the energy profile of a hydrogen atom at the proximity of ferrite (ivory)-cementite (blue) interface. The presence of elastic strain, represented by color gradients, in the two phases can alter the energy state of hydrogen solute, leading to their tendency of migration.

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