Ammonia effect on hydrogen embrittlement mitigation and induction

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In the face of climate change and the energy crisis, the transition to a carbon-neutral society is becoming more and more urgent. Hydrogen is one of the promising ways to achieve carbon neutrality. However, hydrogen embrittlement (HE, degradation of material strength due to hydrogen) is one of the barriers to deploying hydrogen. Hydrogen uptake in the material is an essential process of HE. In a gaseous hydrogen (H_2) environment, hydrogen uptake occurs following the dissociation of the hydrogen molecule into hydrogen atoms aided by the catalytic action of the iron (Fe) surface. Specific gas impurities such as O_2 and CO, which have a stronger affinity with the Fe surface than H_2, preferentially adsorb on the Fe surface and deactivate the catalytic action of the Fe surface for the H_2 dissociation. As a result, HE is mitigated. This technology enables to use of hydrogen incompatible steels in the hydrogen environment. Hydrogen can be transported by the existing natural gas pipelines. The impurity technology can bring us both safety and economic benefits.

In this study, we investigated the NH_3 impurity effect on HE during the fracture toughness test. The material was a low-alloy steel SCM440. **Fig.1** shows the experimental setup. The results are shown in **Fig.2**. Interestingly, it was found that NH_3 had both mitigation and induction effects on HE. Also, NH_3 showed reverse NH_3 concentration dependence in its HE induction effect. To elucidate the mechanisms, H_2 dissociation, NH_3 adsorption and decomposition, electron density, and reaction rate with Fe surface were investigated by Density Functional Theory (DFT). The kinetics model was established to calculate the coverage of each species on the Fe surface according to the Langmuir theory.

The mechanisms considered are shown in **Fig.3**. The reaction rate of NH_3 with the Fe surface was significantly higher than that of H_2. Therefore, NH_3 covers efficiently the Fe surface and hinders hydrogen uptake in the material. As a result, HE was mitigated. Oppositely, NH_3 induces HE when its decomposition to H and other compounds like NH_2 with the help of the Fe surface catalytic action takes place. Either mitigation or induction of HE is dominant is determined by the NH_3 adsorption rate and NH_3 decomposition rate. The DFT revealed that the NH_3 adsorption rate coefficient is significantly higher than the NH_3 decomposition rate coefficient. Therefore, NH_3 induces HE only when the time for decomposition is secured, in other words, the loading rate was sufficiently low. Regarding negative NH_3 concentration dependence in the HE induction effect, it is caused by the reduction in the H supply by NH_3 decomposition with an increase in NH_3 concentration. The kinetic model for the whole steps of NH_3 decomposition that I established revealed that the second step of decomposition ($NH_2 \rightarrow NH + H$) restricts the decomposition process.

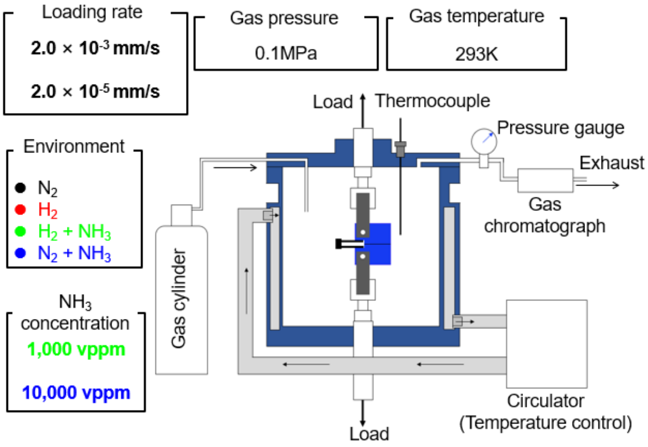

Fig. 1 Fracture toughness test

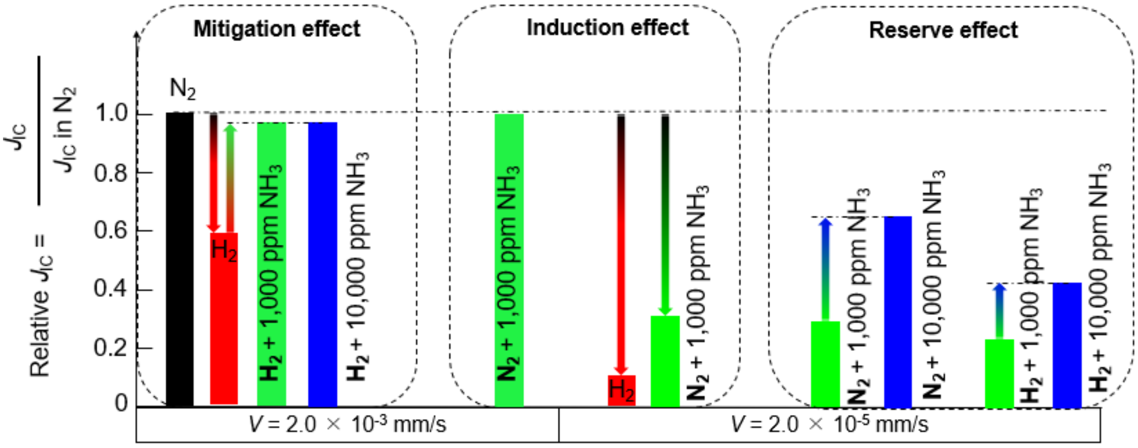

Fig. 2 NH_3 effects during fracture toughness test

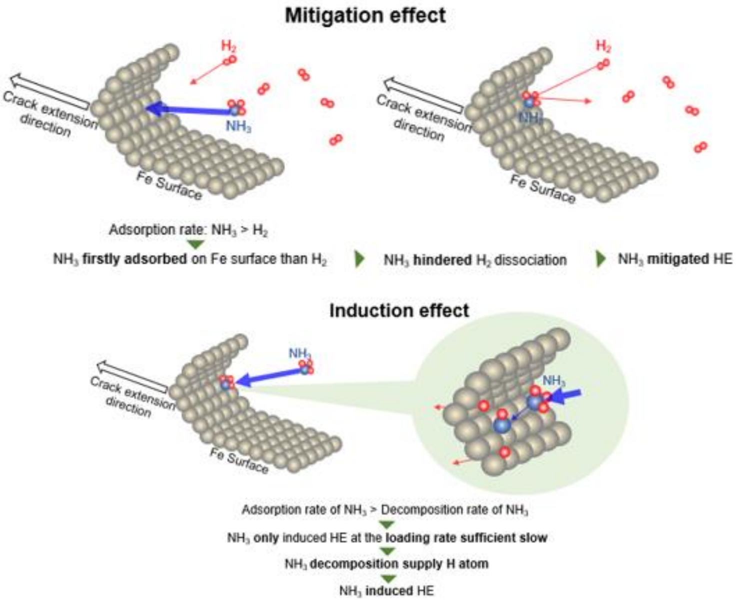

Fig. 3 Mechanisms of NH_3's HE mitigation and induction effects