Experimental investigation on Hydrogen embrittlement of EN47 Spring steel

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Abstract. Carbon steel is one of the significant engineering materials used in industrial and automotive sector. Most of the engineering metals are susceptible to hydrogen embrittlement (HE). It is an inherent phenomenon, which will occur during the course of service life and leads to premature failure of the component. In view of this, an experimental study was conducted to investigate the failure behavior of EN47 in a hydrogen simulated environment. The immersion type hydrogen charging method was adopted. The effect of HE was explored through conventional tensile testing. Experimentations were carried out according to ASTM E8 Standards. The tensile behavior of spring steel and effect of hydrogen interaction with materials was studied. The treated test material experience brittle to ductile transition exhibiting mixed mode fracture morphology on fracture surfaces, often considered being confirmation of failure through HE. Hydrogen charging causes the samples to become brittle, and reduced the plastic deformability. From the test results it can be interfered that the hydrogen charged specimen exhibits loss of ductility and also reduced the ultimate strength.

1. Introduction
Steel is an alloy of carbon, iron and other metals and non-metals. Because of their versatile achievable properties, availability and a relatively low production cost, they are most widely used in engineering and automotive industries viz., transportation, construction, Energy, Industry and appliances. They are especially important as engineering construction materials. They are widely used in boilers, pressure vessels, heat exchangers, piping, and other moderate-temperature service systems in which good strength and ductility are desired [1-3]. They are used due to their availability, relative inexpensiveness and they may be tailored to have a wide range of mechanical and physical properties. There is wide range of steels used as spring steel in automotive to aerospace industries for their resilience and elastic properties with high yield strength. Majority of the spring material requirement are met with the carbon steel, alloy steel and Corrosion resistant steels. Some of the desired mechanical properties are yield strength, toughness and fatigue strength to withstand the high stress, pulsating load, shock or impact loading. In these applications, the long-term static and dynamic behavior plays a prominent role. In the automotive industry, it is recognized that effective life of most components may be affected by yielding, not fracture.
Absorption and release of energy during the lifetime of the usage of a spring is a cyclic loading and unloading process. Therefore, spring failures generally occur by a fatigue mechanism although the root cause may be related to material and processing factors [4,5]. Damage of metals due to the influence of hydrogen is quite frequent and leads to dangerous failures as well as to loss of property and large compensational payments by insurance companies. High stresses combined with a corrosive environment can cause critical components inside power plants and other systems to crack and fail, sometimes with little warning. Hydrogen embrittlement is a delayed catastrophic failure that occur at ambient temperatures, in many steel and other metal alloys when when atomic hydrogen diffuses into the metal structure and interacts within the metal to form a brittle material. It is characterized by mechanical properties degradation due to hydrogen ingress, such as loss of ductility and, consequently, a loss of strength. Most of the engineering metals are susceptible to HE during manufacturing, processing and service environment. HE is considered as one of the primary cause of the premature failure in engineering structure and must be considered in determining the life expectancy of an engineering structure [6,7]. It can be a very complicated process and reliability life prediction is often difficult. This kind of intrusion of hydrogen will produce a disastrous reduction in the ductility and toughness of steel. For evaluation of HE resistance, hydrogen must be charged to specimens. The immersion type hydrogen charging method was adopted [8]. In the present study the effect of hydrogen on the mechanical behaviour of EN47 spring steel has been investigated by means of slow strain rate tensile test of pre-stressed and hydrogen charged specimens. The results are discussed concerning their tensile behavior to HE.

2. Material and Methods

EN 47 is selected as the testing material, as it is used extensively in the aviation, aerospace and automobile industries, among others, due to its good extrudability and weldability and excellent corrosion resistance. Spring steel is medium low alloy, medium carbon steel (EN47/AISI 6150/SUP 10) which is suitable for over-all engineering purposes with the higher strength than the mild steel. As procured material has been chemically tested for the compositions are represented in table 1. HE is the degradation of structural properties of metals due to interaction with hydrogen. It results in loss of ductility and reduced load carrying capacity which usually due to the presence of dissolved hydrogen. The interaction of hydrogen with microstructural features can strongly influence fracture resistance and sub-critical cracking of a material. As HE occurs in most metals, investigation of mechanisms failure under the tensile load is studied and mechanical properties are evaluated. Ammonium thiocyanate (NH4SCN) is one of the widely used reagent for charging steel materials with hydrogen because of its effect on promoting hydrogen absorption and its ease of use [7]. To charge hydrogen, the specimens were immersed in a 35% aqueous solution of ammonium thiocyanate at an ambient temperature for a period of 120 hours. The ammonium thio cyanate solution for the immersion test was prepared by diluting with distilled water to concentration of 35%w/vol at room temperature. A typical test setup was shown in figure 1. Laboratory immersion tests were conducted in accordance with ASTM standard practice. The hydrogen charging carried out with a conventional laboratory method by immersing the pre-stressed specimens.
2.1 Test Specimens and Experimentation

The tensile tests were performed according to the ASTM E8 specification. The configuration and the dimensions of the smooth specimens used in tensile testing are shown in Figure 2. The test cylindrical bar samples of Alloy EN47, gage length ‘5d’ with a diameter ‘d’ of 6 mm were polished and cleaned to obtain the required metallographic finish before test. For the test a fully integrated universal testing machine 50 KN machine was used. The deformation rate was 0.3 mm/min at room temperature. The tensile properties: yield stress, ultimate stress, percentage elongation were evaluated. The toughness is calculated from the area under the true stress–true strain curve. The tensile tests are performed on cylindrical specimens before end after exposed to a hydrogen charging environment. The decrease in mechanical properties between treated and untreated samples at the same strain rate is an indication of the hydrogen embrittlement susceptibility. In laboratory investigations, the specimens were pre-stressed beyond their yield stress to experience a plastic deformation (70% of $\sigma_{ult}$). These pre-stressed specimens were subjected to hydrogen charging in a simulated corrosion media and then subjected to complete failure by tensile testing. Failures, when they occurred, were in regions of stress concentrations resulting from the combined action of stress and the general attack of the corroder. A typical untreated and hydrogen charged specimen is presented in figure 3.

### Table 1: Chemical composition of test specimen material EN47/SAE6510

| Elements     | Carbon | Silicon | Manganese | Phosphorus | Sulphur | Chromium | Vanadium |
|--------------|--------|---------|-----------|------------|---------|----------|----------|
| %            | 0.451  | 0.247   | 0.660     | 0.018      | 0.010   | 1.0      | 0.155    |

Figure 1 Typical experimental setup for immersion test

Figure 2 Typical Tensile Specimen
3. Results and Discussion

The research was based on the material's response to an externally applied load in conjunction with the laboratory simulated conditions. Under the applied load and service conditions, the macro/microstructural mechanical behaviour, the interaction between structure and mechanical properties, material instabilities, and fracture of the material were investigated. The tests were performed on spring steel specimens. The experimental study involves hydrogen charging the specimen and mechanical testing and characterization of the charged specimens and SEM observations of the deformation processes of materials at the micro- and macro-level.

3.1 Stress/strain behavior of treated and untreated material

The images in figure 4 (a) show the fracture surface of a steel with multiple cracks triggering portion (Radial Zone) that failed in a brittle manner with little deformation. It is roughly perpendicular to the applied tensile stress direction. The fan shaped ridges like pattern on the fracture surface indicates an unstable crack growth without increase in the applied stress. At the centre, a fibrous zone depicts overload fracture morphology. The figure 4(b) shows a fractured surface of pre stressed hydrogen charged specimen which depicts delayed fracture of brittle to ductile transition; arrow marks indicate the crack initiation site and shear lip formation and overload fracture. There is a mixed mode of fracture forming and flat surface with a shear lip at failure zone. Figure 4(c) illustrates the engineering stress/strain curves resulting after hydrogen charging and untreated tensile test specimens. A stress/strain curve results in a linear relationship, as shown in Figure 4(c), of treated and untreated steel. It can notice that a variation in the yield strength, ultimate strength and final strain (ductility) exists. It is seen from Figure 4(c) that the tensile strength and the ductility decreased with reference to the untreated specimen. This is in accordance with the observations made in the test results. The slope of the initial linear portion of the stress strain curve shows that the ability to resist deformation in the treated specimen is increased indicating an increase in yield strength, but significant reduction in maximum load carrying capacity. The slope of the treated specimen curve indicates increased stiffness with greater slope confirms that material is embrittled. The figure 5(c), stress strain curve illustrates area under the curve, ability to absorb energy in the plastic range and is increased by approximately 10 percent. As the treated specimen is solution treated for 120 hours for hydrogen charging, might have led to precipitation of impurities which impede the movement of dislocation and to harden the material [9-12]. This might be the reason for increase in the yield strength which needs further investigation.
3.2 Influence of HE on Volumetric changes and toughness

Displayed in figure 5, are the apparent values of the percentage elongation and area reduction; engineering yield stress and ultimate stresses; area under the curve as toughness of the untreated and pre-stressed hydrogen charged specimens. The hydrogen charging does affect the ductility of EN47 steel as depicted from figure 5(a) with variation in percentage change in elongation and reduction in area. The test result shows that the percentage of change in elongation and percentage reduction in area were 17 and 28 respectively. The effect embrittlement is greater upon the values of reduction of area than elongation. The yield stress is given as 0.2% strain. The yield strength of the hydrogen charged specimen increased but reduced in ultimate strength pointing towards the inception of plastic deformation. From figure 4(c) and figure 5(c) has indicated Hydrogen charging causes the samples to become brittle, and reduced the plastic deformability. It is observed that a sharp drop in the ultimate strength of charged specimen, pointing towards embrittled failure. The interaction of hydrogen with the dislocations in the plastic zone of pre-cracks in specimen degraded the ultimate tensile strength (UTS).

3.3 Micro-mechanism failure analysis

One of the characteristics that have convinced investigators that hydrogen embrittlement is related to the presence of internal molecular hydrogen evolved in a mono-atomic form as part of the cathode reaction of almost any electrochemical reaction, which are almost damaging cause of post-processing failure through hydrogen embrittlement[11]. The introduction of excess hydrogen can result in crack nucleation and growth and in some cases failure under load, in both brittle and ductile materials. It is assumed that dissolution is concentrated at the crack tip because of the effects of yielding at that point, and that the cathodic reaction occurs only on the surface of the specimen. In general the stress at the tip of a crack exceeds the yield stress.
The susceptibility was attributed to segregation or precipitation at grain or sub-grain boundaries [12]. As discussed in the previous section, the macrograph provides substantial evidence of failure initiation site. The SEM micrographs analysis (Figure 6) shows the fracture morphologies at the failure area of untreated specimens. In an untreated material there won't be any preexisting cracks. By the virtue of the applied load, the material is pulled to certain extent with the slant surface because of induced crack. Here in this region there won't be any striation or any triggering sites. In this region the material act as aggregate till the crack initiates towards the core. From this point onwards the material will fail by the virtue of material constituents. At the mid region, where there is more resistance, the material will scoop. This is can be witnessed in the micrograph figure 6b, here it exhibits almost a flat surface beyond which normal shear cracks develops. The fracture occurred due to overload. The SEM micrographs analysis (Figure 7) shows the fracture morphologies at the failure area and pre existing cracks induced due the interaction of pre-stressed condition and hydrogen charging of specimens. Fracture surfaces of treated samples revealed pre-exist crack induced due the chemical treatment give raise to slant surface at the early stage itself. The cracks were induced because of embrittlement process. Fracture surfaces of treated samples revealed distinctive mixed mode of fracture indicating a brittle–ductile fracture transition as in case of precipitate hardening. There are indication of cleavage facets and ductile tearing on the fracture surface. Both equiaxed and elongated quasi-cleavage facets were observed. The microvoid coalescence during ductile failure can be witnessed. The fracture surface of the treated specimen suffered from deep and uniformly distributed dimples, which indicates the ductile nature of fracture at the mid region [13,14].

![Figure 6. SEM micrograph of untreated fracture surface; (a) Initiation of Multiple cracks due tensile loading (b) surface showing coalescence of void and fibrous fracture overload zone](image)

![Figure 7. a) SEM micrograph treated fracture surface; (a) Showing dimples of varying size and a noticeable number of dimples. (b) distribution of voids and void coalescence on the overload fracture surface.](image)
4. Conclusions
HE can be defined as the hydrogen-caused deterioration of the mechanical properties of most metallic materials and alloys. It leads loss of ductility, and tensile strength from the presence of dissolved hydrogen. Hardening by hydrogen is more general than softening for commercially pure iron, low-alloyed and stainless steels tested for bulky specimens at room temperature. Based on the tests, the following conclusions can be drawn:

- The effect of hydrogen charging is noticed in-terms of variation in yield strength, ultimate strength and ductility of the untreated and treated tensile test specimens. This could be due to Hardening by hydrogen.
- The failure is due to hydrogen embrittlement. It is observed that a sharp drop in the ultimate strength of charged specimen, pointing towards embrittled failure.
- The treated test material experience brittle to ductile transition exhibiting mixed mode fracture morphology on fracture surfaces.

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