

**MEASUREMENT OF THE HYDROGEN DIFFUSION THROUGH VARIOUS  
STEELS WITH AND WITHOUT OXIDE FILMS**



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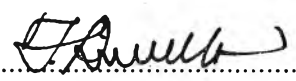
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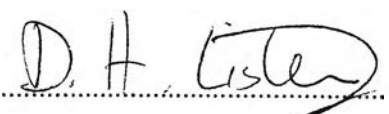
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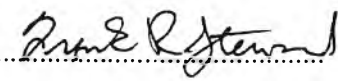
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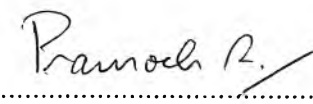
  
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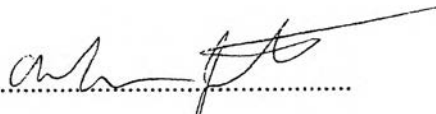
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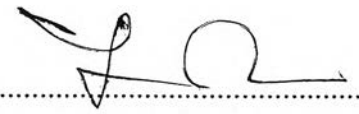
  
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## ABSTRACT

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Chutima Leelasangjai: Measurement of The Hydrogen Diffusion Through Various Steels With and Without Oxide Films

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Hydrogen atoms are produced electrochemically when ferrous metal is exposed to high temperature water. The rate of hydrogen produced corresponds to the rate of corrosion which has been designated as Flow Accelerated Corrosion (FAC). In deaerated conditions, the atomic hydrogen enters the metallic lattice interstitially and permeates through the metal and recombines to form hydrogen gas at the outer surface of the metal. Due to the rapid diffusion of hydrogen through the metal at the temperature of interest, the total rate of hydrogen effusion from the metal is an indication of the instantaneous corrosion rate. The Hydrogen Effusion Probe (HEP) is being developed for on-line monitoring of FAC by measuring the through-wall hydrogen effusion. This study was carried out to better understand the fundamental principles of corrosion and transport of hydrogen through carbon steel associated with FAC. The coefficient of hydrogen diffusion in carbon steel was determined at various temperatures and found to obey Arrhenius Law. The effects of surface conditions on hydrogen permeation were studied by treating the surface to form an oxide film and by coating it with platinum in order to elucidate the diffusion barrier of hydrogen atoms and the kinetic barrier at the surface. The surface resistance due to an oxide layer on a carbon steel tube was determined on both the inside and outside of the tube.

## บทคัดย่อ

ชุตินา ลีลาแสงสาย : ชื่อหัวข้อวิทยานิพนธ์ การวัดการแพร่ของไฮโดรเจนผ่านแผ่นเหล็กที่มีและไม่มีเหล็กออกไซด์ (Measurement of The Hydrogen Diffusion Through Various Steels With and Without Oxide Films) อ. ที่ปรึกษา : รศ.ดร. ชีรศักดิ์ ฤกษ์สมบูรณ์, ศ.ดร. แฟรงค์ อาร์ สจีวิต, แอนดรู จัสต์อะซัน และ ศ.ดร. ดีเรก เอช ลิสเตอร์, 132 หน้า

ไฮโดรเจนอะตอมเป็นผลิตภัณฑ์ที่เกิดขึ้นจากการเปลี่ยนแปลงทางเคมีที่เกี่ยวข้องกับอิเล็กตรอนเมื่อพื้นผิวของโลหะเหล็กสัมผัสกับน้ำที่อุณหภูมิสูงโดยอัตราการเกิดขึ้นของไฮโดรเจนอะตอมสัมพันธ์กับอัตราการกัดกร่อนของเหล็กที่เป็นผลมาจากการกัดกร่อนจากความเร็วยังของของไหลที่ไหลมาสัมผัสกับพื้นผิวของโลหะ (Flow Accelerated Corrosion, FAC) ในระบบของน้ำที่ปราศจากออกซิเจน ไฮโดรเจนอะตอมที่เกิดขึ้นจะเคลื่อนที่เข้าสู่ช่องว่างของโลหะและแพร่ผ่านโลหะและจึงรวมตัวกันเป็นก๊าซไฮโดรเจนที่บริเวณพื้นผิวด้านนอกของโลหะนั้น เนื่องจากไฮโดรเจนอะตอมสามารถแพร่ผ่านเหล็กได้อย่างรวดเร็วในช่วงอุณหภูมิที่พิจารณา ดังนั้นอัตราการแพร่ออกมาทั้งหมดของไฮโดรเจนคือการวัดอัตราการกัดกร่อนในขณะนั้น เครื่องมือวัดการแพร่ผ่านของไฮโดรเจน (The Hydrogen Effusion Probe, HEP) ได้ถูกพัฒนาขึ้นเพื่อใช้สำหรับตรวจวัดการกัดกร่อนจากความเร็วยังของของไหลที่ไหลมาสัมผัสกับพื้นผิวของโลหะอย่างเชื่อมตรง โดยการวัดปริมาณไฮโดรเจนที่เกิดขึ้นและแพร่ผ่านออกมายังผนังของโลหะ งานวิจัยนี้ได้ทำการศึกษาหลักการพื้นฐานของการกัดกร่อนและพฤติกรรมเคลื่อนย้ายของไฮโดรเจนผ่านโลหะคาร์บอนที่เกิดจากการกัดกร่อนจากความเร็วยังของของไหลที่ไหลมาสัมผัสกับพื้นผิวของโลหะเพื่อหาสัมประสิทธิ์การแพร่ของไฮโดรเจนในเหล็กคาร์บอน สัมประสิทธิ์การแพร่ของไฮโดรเจนของโลหะคาร์บอนที่อุณหภูมิต่างๆ สัมพันธ์กับกฎของอาร์ฮีเนียส ผลของสภาพพื้นผิวที่มีต่อการแพร่ของไฮโดรเจนถูกศึกษาโดยการทำพื้นผิวเกิดเหล็กออกไซด์และถูกเคลือบด้วยโลหะแพลทินัมเพื่ออธิบายการขัดขวางการแพร่ของไฮโดรเจนอะตอมและการขัดขวางทางจลนศาสตร์ที่พื้นผิว นอกจากนี้ความต้านทานที่พื้นผิวด้านในและด้านนอกบนต่อเหล็กคาร์บอนเนื่องมาจากเหล็กออกไซด์ถูกกำหนด

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**ABBREVIATIONS**

AECL	Atomic Energy of Canada Limited
ASTM	American Society for Testing and Materials
BCC	Body-Centered Cubic
CANDU	Canada Deuterium Uranium
CNER	Center for Nuclear Energy Research
COG	CANDU <sup>®</sup> Owners Group Inc
EDS	Energy Dispersive Spectrometry
EFPY	Effective Full Power Years
FAC	Flow-Accelerated (Assisted) Corrosion
FCC	Face-Centered Cubic
FESEM	Field Emission Scanning Electron Microscope
FOLTM	Feeder On-Line Thickness Monitor
HE	Hydrogen Embrittlement
HEP	Hydrogen Effusion Probe
HMT	Hydrogen Microprint Technique
NTP	Normal temperature and pressure
PHWR	Pressurized Heavy Water Reactor
PLGS	Point Lepreau generating station
RPC	Research and Productivity
STP	Standard temperature and pressure
UNB	University of New Brunswick
XRD	X-Ray Diffraction

## LIST OF SYMBOLS

$\Delta$	Delta operator or difference operator
$\nabla$	Del operator or vector differential operator
$\rho_{Fe}$	Density of iron
$a$	Conversion of days to year (365 days/yr)
$A_i$	Internal area of pipe
$C$	Concentration of the diffusing substance
$C_{H_2}$	Concentration of molecular hydrogen in the metal
$\nabla C$	Concentration gradient
$C$	Concentration of the diffusing substance
$C_r$	Corrosion rate
$D$	Diffusion coefficient or diffusivity
$D_{H_2}$	Hydrogen diffusivity
$D_0$	Maximum diffusion coefficient (at infinite temperature)
$E_A$	Activation energy for diffusion
$E_P$	Activation energy for permeation
$E_S$	Activation energy for solubility
$J$	Diffusion flux
$\sigma k_1$	Surface constants for the adsorption
$\sigma k_2$	Surface constants for the release
$K$	Dissociative adsorption equilibrium constant
$l$	Thickness of the diffusion path
$M$	Relative resistance
$M_1$	Molar mass of gas 1
$M_2$	Molar mass of gas 2
$M_{Fe}$	Molar mass of iron
$\frac{dn}{dt}$	Change of hydrogen molecule with time

N	Relative position
$P$	Permeability
$P_{\infty}$	Hydrogen permeation rate at steady-state
$P_0$	Maximum permeability (at infinite temperature)
$P_{H_2}$	Hydrogen pressure
$P_{H_2,f}$	Feed side partial pressures of hydrogen
$P_{H_2,p}$	Permeate side partial pressures of hydrogen
$Q$	Permeability of hydrogen through the membrane
$R$	Gas constant
$Rate_1$	Rate of effusion of the first gas
$Rate_2$	Rate of effusion of the second gas
$S$	Solubility
$S_0$	Maximum solubility (at infinite temperature)
$T$	Absolute temperature
$\frac{dP}{dt}$	Change of hydrogen pressure with time
$P_1$	Initial hydrogen pressure
$P_2$	Hydrogen pressure at the end
$r_1$	Inside radius of tube
$r_2$	Outside radius of tube
$R_i$	Inside surface resistance
$R_m$	Metal surface resistance
$R_0$	Outside surface resistance
$R_t$	Overall surface resistance
t	Time
$T_{eff}$	Effective temperature
$V$	Total volume of system
$x$	Coordinate chosen perpendicular to the reference surface

$x_1$	Characteristic length
X	Relative time
Y	Unaccomplished concentration change