

韧性体应力腐蚀机理模型

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摘要: 工业上常见的应力腐蚀 (SCC) 是一种具有低应力脆性解理特征的失效模式, 同时 SCC 机理一直是近百年来的科学难点和研究热点。结合最新研究进展, 综述了韧性体 SCC 机理模型。宏观上, SCC 通常由敏感材料、特定环境和应力三因子构成, 其失效准则遵循 Griffith 理论。微观上, 重点分析了近几十年提出的 SCC 机理的构型、适用性和局限性。大体上, SCC 微观机理主要包括滑移溶解模型、择优溶解模型、膜致解理模型、腐蚀促进塑性变形模型和环境断裂一致性模型。尽管各模型对 SCC 的物理化学本质尚存争议, 但从阳极溶解到膜破裂、位错滑移到脆性解理、氢的促进作用到电负性粒子的耦合作用, 各模型对 SCC 的认识已逐步深化, 已深入至原子尺度, 具有重要理论意义和应用价值。此外, 还展望了 SCC 前沿科学与应用前景。

关键词: 应力腐蚀; 机理模型; Griffith 理论; 脆性解理; 韧性体; 低应力; 特定环境

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Mechanism Models of Stress Corrosion Cracking in Ductile Solids

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ABSTRACT: Stress corrosion cracking (SCC), often occurring in industries, is a failure mode with the feature of brittle cleavage at low stress levels. Meanwhile, the SCC mechanism is always a tough and focused scientific issue during about one hundred years. Based on recent research progress, SCC mechanism models specific in ductile solids are reviewed in this paper. Macroscopically, the three factors on SCC consist of susceptible materials, specific environment and stress as usual. The failure criterion of SCC accords with the Griffith's theory. Microscopically, this work primarily analyzes models, suitability and limit of SCC mechanisms proposed in the past several decades. In principle, the microscopic SCC mechanisms are composed of the slip dissolution model, the selective dissolution model, the film induced cleavage model, the corrosion enhanced plasticity model and the uniformed environment assisted cracking model. It is obvious that the physical and chemical SCC essence is controversial until now. However, from anodic dissolution to film rupture, from dislocation slipping to brittle cleavage and from the hydrogen-enhanced effect to the synergistic effects of electronegative particles, the development of the models gradually tents to atomic scales. As a consequence, the SCC models are fairly valuable in the aspects of theory and application. Besides,

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the prospects of advanced SCC science and application are also done at the final.

KEY WORDS: stress corrosion cracking; mechanism model; Griffith's theory; brittle cleavage; ductile solid; low stress; specific environment

应力腐蚀 (SCC) 在武器装备、航空航天、核能核力、深潜器、舰艇船舶、海洋设施等领域时有发生。科学上较早记载的 SCC 事故, 是 20 世纪初英国殖民军在印度贮存的黄铜弹体的季节性开裂^[1]。尽管已延宕百年, 至今 SCC 问题依然严峻, 很具挑战。例如, 近几年印度与俄罗斯研制的 6000 m 深水潜航器链绳控制系统所用的 SS316L 奥氏体不锈钢螺钉, 在海水中浸泡约 200 h、暴露空气中 6 年后, 发生了穿晶解理型应力腐蚀断裂; 承压舱用 AISI204 奥氏体不锈钢螺钉在海水中浸泡 7~8 h、暴露空气中 2 个月, 首先发生点蚀, 而后发生穿晶解理型应力腐蚀断裂^[2]。2002 年印度运载火箭辅助燃料箱波纹管、2003 年美国核反应堆压力容器等也出现了 SCC 现象。同时, 我国舰载与临海武器装备值班寿命较短, 文昌卫星发射架腐蚀多发, 其中也存在 SCC 现象。尽管一些系统多次涂覆防护涂层, 或更换材料或部件, 但 SCC 问题并没有根本解决。

应力腐蚀一直困扰至今难以解决的原因, 就在于人们不清楚引起 SCC 的机理, 特别是韧性体脆性解理的微观机制。研究表明 SCC 是一种低应力失效模式, 即在特定环境下受许用应力的材料发生滞后脆性开裂或断裂的现象, 也称之为“敏感材料、特定环境和应力”三因子理论 (如图 1 所示), 如 NaCl 水溶液+奥氏体不锈钢+恒载荷的经典 SCC 体系^[3-4]。整体上看, SCC 具有外加应力低与韧性体脆性开裂的特点。同时, 基于二维与三维 SCC 实验及模拟结果, 提出了“局部腐蚀、膜破裂形成蚀孔和裂纹纵深扩展”的三阶段理论以及阳极溶解、氢致开裂两型 SCC 宏观机制^[1,3-4], 但目前 SCC 微观机制仍存在争议。

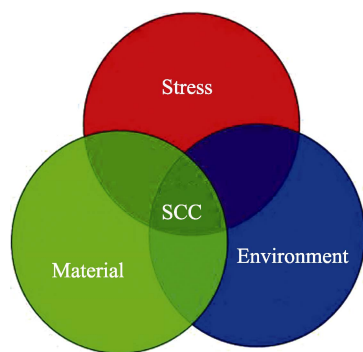


图 1 应力腐蚀三因子
Fig.1 Three factors on SCC

1 Griffith 理论

针对脆性开裂与断裂的现象, Griffith 认为脆性

裂纹形核扩展的临界应力强度因子 K_{IC} 取决于新裂纹面的表面能 γ_s ^[5], 也称之为 Griffith 理论, 即:

$$K_{IC} = \sqrt{\frac{2\gamma_s E}{1-\nu^2}} \quad (1)$$

其中: E 为材料的弹性模量, ν 为材料的泊松比。基于 Schmidt 定律, 在真空和自然环境条件下, 韧性材料的临界分切应力一般都低于解理应力门槛值。因此在外应力作用下, 其首先发生以位错滑移为元过程的塑性变形, 而后裂纹形核扩展以致断裂。据此, Irwin 和 Orowan 认为裂纹扩展的阻力 R_C 不仅是新裂纹面表面能, 还包括塑性变形功 γ_p , 那么公式(1)推广至韧性体裂纹形核扩展的现象即变为:

$$K_{IC} = \sqrt{\frac{(2\gamma_s + \gamma_p) E}{1-\nu^2}} \quad (2)$$

近期的研究表明, 极低外加应力 (约 $0.1\sigma_s$) 条件下, 具有优异韧塑性的奥氏体不锈钢单晶在高浓度 Cl⁻ 水溶液中首先发生点蚀, 而后发生低应力 SCC 裂纹形核扩展, 却不发生位错滑移塑性变形^[6-10], 如图 2 所示, 表明强腐蚀环境粒子使韧性体脆化, 以致发生无位错发射条件下的低应力解理。这与经典的 Griffith 理论和多数工程失效不发生宏观塑性变形的

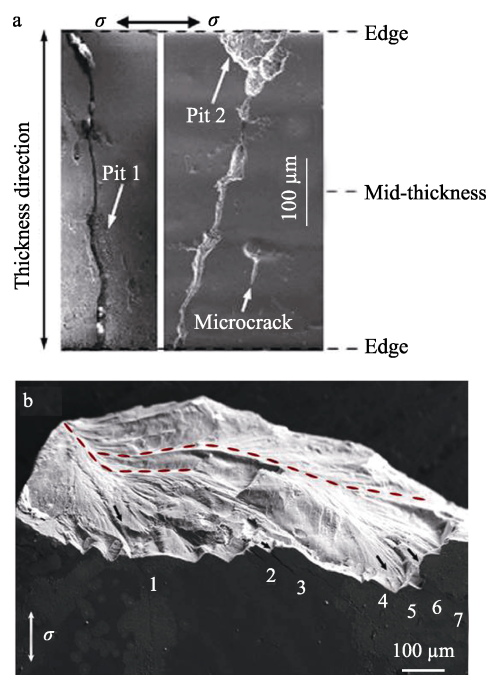


图 2 恒载荷 (约 $0.1\sigma_s$) 作用下 316L 不锈钢单晶在高浓度 Cl⁻ 溶液中的点蚀致 SCC 与微观断口形貌^[7-8]

Fig.2 Pit-to-SCC morphology and fractography of 316L single-crystal stainless steels subjected to a constant load (approximately $0.1\sigma_s$) in a high-density chloride solution^[7-8]

实例相吻合, 但与 Irwin 和 Orowan 以塑性变形功 γ_p 弥合高应力下 SCC 裂尖出现塑性区的观点并不一致, 即韧性体 SCC 的门槛应力位于远低于 σ_s 的量值区间。由此可知, 目前 SCC 研究多采用慢应变速率拉伸、U 型弯曲、C 型环加载、楔形开口恒位移载荷、高应力疲劳、大振幅振动、高载荷冲击等实验方法, 其实验应力通常高于屈服强度 σ_s , 与远低于 σ_s 的实际服役应力条件下构件 SCC 脆性断裂的失效机理并不自洽, 因此有必要深入开展低载荷条件下韧性材料试样、局部结构与复杂系统的 SCC 加速评价方法研究。

2 SCC 机理模型

基于 SCC 三因子理论, 受应力的敏感金属、合金、复合材料及其结构系统等韧性体在特定环境下会发生 SCC 脆性开裂或断裂。工业上常见 SCC 敏感体系包括: Cl^- +钢基构件、潮湿大气+铝镁基构件、热盐+钛基构件、氨水+铜基构件、超临界水+镍基构件、低熔点焊料+高温合金构件等。通常, 无预裂纹构件 SCC 发源于点蚀或缺陷。研究表明, 只有在点蚀微孔生长至一定尺寸, 微孔边缘或底部应力达到 SCC 临界值后, 才发生点蚀致 SCC 转化^[12-16]。研究 316L 奥氏体不锈钢单晶 SCC 的大量微尺度结果表明, 极低弹性应力作用下 SCC 裂纹形核于点蚀肩部^[7], 同时发现强酸性裂尖前缘优先发生阳极溶解, 微裂纹形核于溶解缺陷^[8], 即择优形核的半圆形或 U 形点蚀微孔生长至临界尺寸后, SCC 裂纹在微孔边缘形核, 其扩展速率高于点蚀生长速率, 导致 SCC 裂纹贯穿整个点蚀微孔, 如图 3 所示。除点蚀致 SCC 转化机理, 针对 SCC 裂纹形核与扩展的微观机理, 目前还提出了滑移溶解模型、择优溶解模型、膜致解理模型、腐蚀促进塑性变形模型和环境断裂一致性模型^[1,4,11]。本

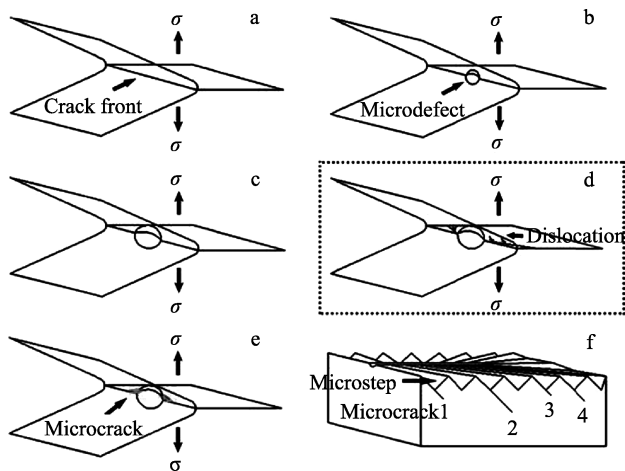


图 3 点蚀致 SCC 转化机理示意图

(虚线表示可略去微剪切过程)^[10]

Fig.3 Schematics of the pit-to-SCC mechanism (the dashed line shows micro-shear is a selective process)^[10]

部分主要结合工业上常见的 Cl^- +韧性体晶体(如不锈钢)这一 SCC 典型体系的近期研究进展, 阐述韧性体 SCC 的微观机理。

2.1 滑移溶解和择优溶解模型

较早提出的滑移溶解模型和择优溶解模型认为, SCC 裂尖新鲜金属阳极溶解形成氧化物、脱合金层等钝化膜, 脆性的钝化膜在外加应力或金属基体滑移剪切作用下发生断裂, 导致了 SCC 裂纹形核扩展^[17-18], 如图 4 所示, 即认为 SCC 裂纹形核扩展的根本原因是本征脆性的钝化膜发生低应力脆性开裂。TEM 原位观察表明, Cl^- 可诱发金属基体纳米厚度钝化膜破裂^[19]。Newman 等^[20]认为滑移溶解模型较好地揭示了 SCC 的动力学特征, 是腐蚀科学的里程碑。依此模型, SCC 应沿滑移面形核扩展。近期的研究首先表明, SCC 微裂纹并不与滑移带平行, 而是与滑移带成一定夹角(如图 5 所示), 一些裂纹在两条滑移带相交处, 并不沿滑移带形核^[9,21]。其次, Magnin 等^[22-23]发现 117 °C 沸腾 MgCl_2 溶液中, 316L 不锈钢 SCC 裂纹不一定沿 $\{111\}$ 滑移面扩展, 也可发生在 $\{110\}$ 解理面, 利用双面痕迹法从不同角度分析所得的 SCC 裂纹面发生在 $\{100\}$ 晶面^[9-10]。这是滑移溶解模型在揭示 SCC 本质特征中不具实证性的根源。同时, 滑移溶解模型和择优溶解模型认为 SCC 会形成钝化膜, 但依据闭

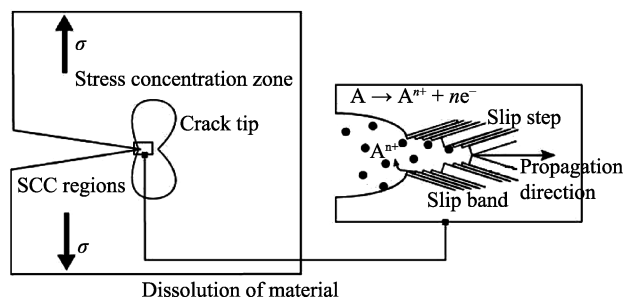


图 4 滑移溶解模型作用下 SCC 裂纹形核与扩展示意图^[17]

Fig.4 Schematics of slip dissolution induced SCC crack initiation and propagation^[17]

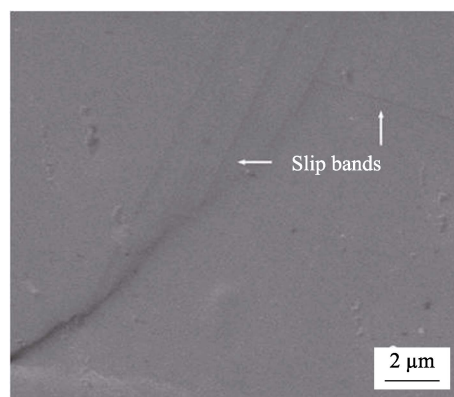


图 5 316L 不锈钢单晶表面 SCC 裂尖与滑移带形貌^[9]

Fig.5 Morphology of the SCC crack tip and the slip bands on the 316L single-crystal stainless steel surface^[9]

塞电池的自催化理论,奥氏体不锈钢应力腐蚀时,裂尖为强酸性环境,SCC扩展过程中很难形成化学稳定的钝化膜,这一事实甚至更早就被发现,即奥氏体不锈钢存在无钝化膜 SCC 体系^[24-25]。因此,将阳极溶解形成的本征脆性钝化膜发生低应力脆性开裂,定义为韧性奥氏体不锈钢 SCC 的根本原因,但目前并没有充分的实验结果和基础理论相佐证。

2.2 腐蚀促进塑性变形模型

基于 316L 不锈钢+沸腾 $MgCl_2$ 溶液+慢应变速率拉伸 SCC 体系, Magnin 等^[22-23]提出了腐蚀促进塑性变形模型^[22-23],即裂尖活化局部溶解,氢、空位沿晶面扩散,集中应力促进位错发射形成塞积群,局部应力和氢的软化作用使微裂纹在位错塞积处不连续形核,滑移面与解理面张开,以 Zigzag 模式扩展,如图 6 所示。从模型可知,空位、位错等微缺陷和氢的交互作用,是裂纹尖端奥氏体不锈钢基体脆化开裂的根源。此模型认识到氢对 SCC 的作用,与 TEM 原位观察所发现的实验现象(氢促进位错运动、降低层错能及沉淀硬化奥氏体不锈钢断裂强度降低)相吻合^[26-27],而且与氢增加阳极溶解速率、降低 K_{ISCC} 、缩短断裂失效时间等宏观现象相一致^[28-29]。可是,腐蚀促进塑性变形模型把 SCC 等同于氢脆,认为氢起控制作用是行不通的,因为面心立方晶体结构的 316L 不锈钢较大的八面体间隙半径为 0.019 nm,而氢原子半径为 0.053 nm^[30],原子氢很难大量扩散进入不锈钢基体,达到氢脆的临界氢浓度。此外,奥氏体不锈钢在低应力作用下充氢时,并不发生氢脆。因此在奥氏体不锈钢 SCC 过程中,氢起促进作用,而不是决定性作用。另一方面,此模型认为 SCC 裂纹胚在位错塞积处萌生。Guo 等^[31-32]利用高分辨 EBSD 和先进光源微束 X 射线衍射显微镜,分析了位错塞积区的弹性应力分布,发现位错塞积处应力强度因子明显高于滑移带上的应力强度因子,同时也发现一些位错塞积的滑移带并无应力集中。近期的研究表明,极低的外加应力作用下,316L 奥氏体不锈钢单晶 {111} 滑移面上沿<110>滑移方向的切应力为 21.4 MPa,达不到滑移系开动的临界分切应力 ($\tau_K=23$ MPa),不发生位错滑移变形,但依然发生 SCC 裂纹形核扩展^[6-10,33]。因此,SCC 可在无位错滑移条件下发生。由于略高应力(高于 τ_K 且低于 σ_S)作用时,裂纹形核扩展更快,同时河流状断口花样上出现撕裂岭,如图 7 所示,因此韧性体 SCC 脆裂中微剪切韧性机制与微解理脆性机制的协同作用有待完善。

2.3 膜致解理模型

从无位错发射的脆性角度出发,Newman 与合作者提出了膜致解理模型,即腐蚀产物膜阻碍金属基体位错运动或通过镜像力将位错吸附在膜内,微裂纹在腐蚀产物膜中形核并脆性解理扩展至金属基体^[34]。原

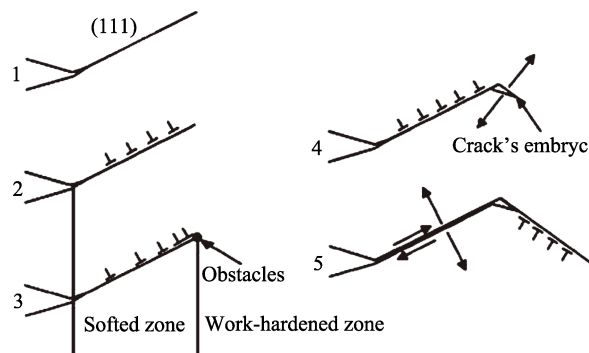
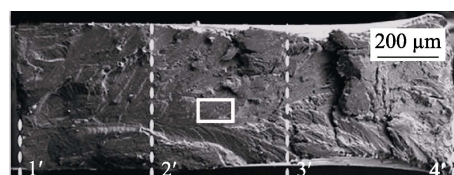
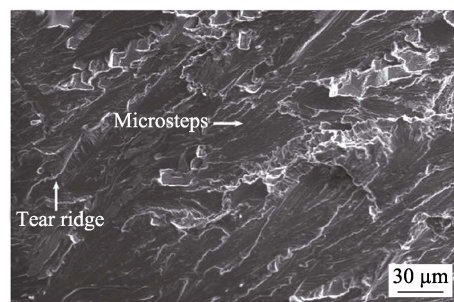


图 6 腐蚀促进塑性变形模型作用下 SCC 裂纹形核与扩展示意图^[22]

Fig.6 Schematics of corrosion enhanced plasticity induced SCC crack initiation and propagation^[22]



a 河流状花样



b 韧性撕裂岭(b图为a图矩形标记部分)

图 7 316L 不锈钢单晶微观断口形貌^[9]

Fig.7 Fractography of 316L single-crystal stainless steel^[9]: a) river-like morphology, b) ductile tear ridges and brittle cleavage microsteps (b is the part marked by the rectangle in a)

子力显微镜研究表明,316L 不锈钢表面生成的氧化膜具有半导体特性^[35]。进一步研究表明,在三点弯曲应力作用下,形核于脆性膜的微裂纹扩展至韧性基体,并使韧性体发生脆性开裂^[36-37]。研究黄铜在 Mattassion 溶液中的 SCC 时也发现,将拉应力施加于已形成钝化膜的缺口时,微裂纹形核于钝化膜,并扩展至金属基体,裂尖前沿无宏观塑性变形^[38-39]。更进一步地,腐蚀产物膜不仅阻碍位错运动,还犹如一个楔子,通过裂尖金属基体的氧化疏松、体积膨胀,对裂尖两侧面产生压应力,整体上看,为拉应力 σ_p 促进 SCC 裂纹形核扩展^[40-47]。但是,此模型对裂尖酸化的 SCC 不一定适用,因为如上所述,动态扩展的强酸性裂尖不一定形成固态的腐蚀产物膜^[24-25],TEM 原位观察确实未发现在动态扩展过程中,裂尖前沿有钝化膜生成^[48],故不应将强酸性条件下 SCC 的脆化本征原因完全归结于腐蚀产物膜阻碍金属基体位错发射或吸附位错。

2.4 环境断裂一致性模型

本质上, SCC 裂纹形核扩展阻力是裂尖原子键断裂所需的作用力^[49-50], 微观上表现为新形核裂纹面的表面能。环境断裂一致性模型试图从原子角度解析 SCC、氢脆和液态金属诱发脆性开裂的本征原因, 认为价电子、导电电子与裂尖正负电势区发生交互作用, 降低了裂纹形核扩展阻力^[11,51]。这与 SCC 无论是阳极溶解形核, 亦或是脆性解理扩展, 都离不开电化学反应的观点是一致的^[3-4,7-8,52-55], 更清晰地说, 裂尖前缘存在微观及原子尺度的电偶对, 在外加载荷条件下, 受引力与斥力协同作用的多个微米纳米尺度电偶对有序分离张开, 即为韧性体 SCC 脆裂的原因。Lu 等^[56]研究高温水溶液中镍合金应力腐蚀的结果表明, Pb 能促进 SCC, 同时, Ca^{2+} 、 Mg^{2+} 也可诱发 SCC, 表明含有价电子、导电电子的电负性粒子是 SCC 的作用因素。以此角度, 环境断裂一致性模型似乎找到了 SCC 的发生源, 因为正是带负电的电子与带正电的原子核相互吸引以及原子间电子云图的相互作用, 才构成金属晶体, 一旦这种作用力消失, 则表现为晶体分离张开。Pu 等^[52]认为可将 SCC 形核扩展的阻力从表面能转化为界面电势, 即:

$$K_{\text{IC}} = (1 - \mu c) \sqrt{\frac{E}{(1 - \nu^2)}} \varphi_n \quad (3)$$

式中, c 为腐蚀性介质浓度, μ 为浓度系数, φ_n 为界面电势。可是, 此模型仍然认为 SCC 裂纹由位错滑移而形核扩展, 这与 316L 不锈钢单晶无滑移条件下的 SCC 实验现象 (如图 8 所示) 不相符^[6-10]。此外, Lu 等^[56]还发现, Ca^{2+} 、 Mg^{2+} 可抑制 Pb 致镍合金 SCC, 因此多种电负性粒子协同作用下的 SCC 机理有待进一步研究。

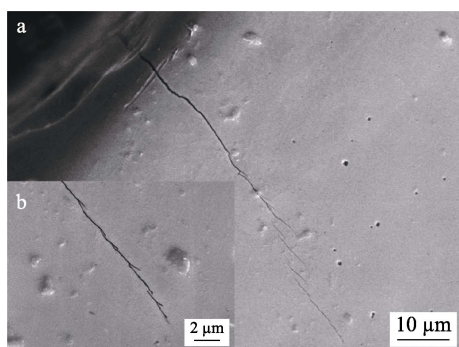


图 8 316L 不锈钢单晶无滑移条件下的 SCC 形貌^[9-10]

Fig.8 Non-slipping SCC morphology of 316L single-crystal stainless steel^[9-10]

2.5 模型评述

工程失效分析表明, SCC 构件并无明显的宏观塑性变形发生^[57], 相应实验结果也表明无位错滑移条件下 SCC 裂纹可形核扩展^[6-10], 且易于沿低表面能晶

面生长 (如奥氏体不锈钢 {100} 晶面, 而非 {111} 滑移面)^[9]。这与滑移溶解模型、腐蚀促进塑性变形模型和环境断裂一致性模型认为 SCC 裂纹沿滑移面形核扩展并不相同, 表明在此情况下上述模型并不适用。尽管膜致解理模型认识到 SCC 的无位错解理特征, 但不适用于具有强酸性裂尖的无钝化膜体系, 也鲜有实验证实钝化膜吸附基体位错的现象。此外, 点蚀致 SCC 转化机理揭示了 SCC 萌生机制, 但并未阐述 SCC 脆性解理的本征特征。因此, 目前 SCC 裂纹形核扩展的微观机理依然不完善。进一步研究发现, 即使相同的 SCC 体系, 不同应力作用下也表现出不同的 SCC 特征, 如 SCC 不同阶段裂纹扩展速率和微观断口形貌存在明显差异, 这表明同一 SCC 体系可能具有不同机理。其中, 高分辨动态原位表征方法 (如同步辐射 X 射线层析成像技术 (如图 9 所示)), 是厘清 SCC 微观机理的可行路径之一^[8,58-62]。

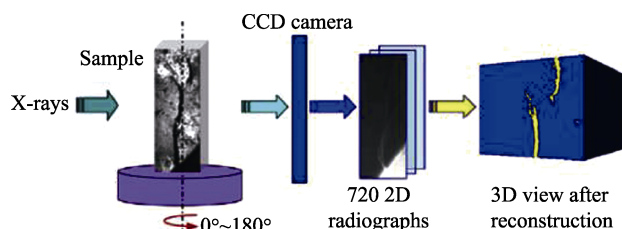


图 9 同步辐射 X 射线层析成像示意图^[8]

Fig.9 Schematic of synchrotron radiation X-ray computed tomography^[8]

近期的研究表明, 局部微电偶对的阳极溶解与正应力作用的低表面能晶面微解理协同, 导致了 SCC 裂纹形核扩展, 同时当滑移面切应力高于临界分切应力时, 位错发射形成微剪切, 新剪切面微溶解促进 SCC 脆性开裂, 形成微溶解-微解理-微剪切耦合作用机制。316L 奥氏体不锈钢单晶 SCC 的大量微尺度结果表明, 无位错滑移条件下, SCC 裂纹可形核扩展^[6-10], 强酸性裂尖前缘优先发生阳极溶解^[8], 微裂纹形核于溶解缺陷^[8], 且易于沿最低表面能的 {100} 晶面而非 {111} 滑移面生长^[9], 这些现象与上述机制相吻合。此外, 发生位错滑移且未缩颈时, 应力弛豫的滑移带会发生阳极溶解^[9], 导致局部弹性应力集中, 产生电负性粒子、氢和空位等, 即微剪切可促进 SCC 脆性开裂。同时, 研究表明, 由于毛细效应和金属离子水解, 局部腐蚀或裂尖内部 pH 值远低于外部溶液的 pH 值^[63], 多数呈强酸性, 因此这一新机制不仅适用于 Cl^- 不锈钢 SCC 体系, 也适用于局部腐蚀致 SCC 裂纹转化和含有强酸性裂尖的多种 SCC 体系。

3 结论与展望

SCC 微观机理一直是国内外研究的热点, 目前已提出的滑移溶解模型、择优溶解模型、腐蚀促进塑性

变形模型、膜致解理模型和环境断裂一致性模型具有重要理论意义和应用价值。但由于先前的研究多采用高应力实验方法,一些韧性假象屏蔽了 SCC 的脆性解理本征,因此基于韧性机制提出的 SCC 微尺度模型在相应体系中已不适用,需要修正。

由于 SCC 环境的强腐蚀性及表征手段的局限性,SCC 微观机理依然是很具挑战性的科学难点。其中,多离子及原子作用下纳米尺度电偶对脆化与低应力解理基因是一个亟待回答的科学问题。此外,由于 SCC 事故通常在经过材料与结构选型的复杂系统服役过程中无征兆发生,因此应在相同失效机理和消除尺寸效应误差的基础上,开展多层次系统装备 SCC 高加速高通量评价技术研究,实现复杂系统多环境耐久兼容和安全可靠服役。

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