

中子注量率对低铜 RPV 钢辐照脆化效应的影响

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摘要: 针对低铜 ($Cu \leq 0.08\text{wt}\%$) 反应堆压力容器 (RPV) 钢的辐照脆化效应, 分析了国内压水堆核电站 49 根辐照监督管以及高通量工程试验堆 (HFETR) 和岷江试验堆 (MJTR) 的 18 次辐照试验结果, 研究了不同中子注量率对低铜 RPV 钢辐照脆化效应的影响。结果表明, 研究试验堆高中子注量率 ($> 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$ ($E > 1 \text{ MeV}$, 下同)) 比压水堆核电站辐照监督管低中子注量率 ($\leq 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$) 引起的低铜 RPV 钢韧脆转变温度变化更显著, 并对不同中子注量和不同中子注量率的结果进行归一化处理, 关联了研究试验堆加速辐照与压水堆辐照监督管试验结果, 这对通过研究试验堆加速辐照评估新型国产 RPV 的使用寿命有重要意义。

关键词: 低铜 RPV 钢; 辐照脆化; 中子注量率

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Effect of Neutron Fluence Rate on Irradiation Embrittlement of Low-copper RPV Steel

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Abstract: The reactor pressure vessels (RPVs) of pressurized water reactor (PWR) nuclear power plants currently in operation and under construction in China are all made of low-alloy ferritic steels obtained by quenching and tempering treatments (which meet the specification requirements of 16MND5 or A508-3) with $Cu \leq 0.08\text{wt}\%$. In this paper, the influence of neutron fluence rate difference between reactor surveillance capsules and accelerated irradiation tests in research reactor was studied by the results of the 49 commercial reactor surveillance capsules and the 18 irradiation tests on High Flux Engineering Test Reactor (HFETR) and the Minjiang Test Reactor (MJTR). The results indicate that ductile brittle transition temperature (DBTT) increment of low-copper RPV steels is influenced by both neutron fluence and neutron fluence rate, the DBTT increase caused by higher neutron fluence rates is more significant when the neutron fluences are comparable, and the DBTT increment of higher neutron fluence is observed to be larger when the neutron fluence rates are comparable. It is found that lower neutron fluence rate ($\leq 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$, $E > 1 \text{ MeV}$) has no effect on the ductile-to-brittle transition temperature of low-copper RPV steels. When evaluating low-copper RPV steels, if the neutron fluence

rate is no more than $1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$, it is not necessary to correct the embrittlement caused by the neutron fluence rate difference between reactor surveillance capsules and research reactor data. Meanwhile, there is no uniform international conclusion on the fluence rate effect when neutron fluence rate $> 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$, and high neutron fluence rate ($> 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$) in test reactors is more significant than that caused by low neutron fluence rate ($\leq 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$) in surveillance capsule. As well, the irradiation embrittlement calculation models such as RG1.99 (Rev. 2) in the United States, JEAC4201 in Japan, and FIS in France are not fully applicable to the changing trends of low-copper RPV steels in China. Without considering the influence of chemical composition, a simple exponential fitting was performed to obtain the relationship between ductile brittle transition temperature increment and neutron fluence. Furthermore, the normalized results for different neutron fluence and neutron fluence rate were obtained, and found the correlation with the different neutron fluence rate results of commercial PWRs and research test. It is of great significance to convert the rapid irradiation test of the research test reactor into the transition temperature rise of the pressure vessel under actual service conditions, which can more accurately and reliably verify the design life and assess the remaining service life of the pressure vessel, and at the same time provide reliable data for the design of the pressure vessel of the newly-built nuclear power plant.

Key words: low-copper reactor pressure vessel; irradiation embrittlement; neutron fluence rate

反应堆压力容器(RPV)是核安全一级设备,是核电站全寿命周期内唯一不可更换的大型设备,RPV的寿命直接决定了核电机组是否能长周期安全运行。RPV在服役期间要长时间遭受高温、高压、强烈的中子辐照,而发生辐照硬化和脆化^[1-2]等老化降质的现象。核电站通过实施辐照监督试验^[3-4]来分析评估RPV的辐照脆化效应,其方法是将辐照监督管安装在压力容器内壁合适位置,并按照辐照监督大纲定期提取解剖检查^[5]。辐照监督管空间有限,耗时较长,其在验证新材料的设计制造工艺和寿期性能时具有局限性,因此新材料的开发应用^[6]以及核设备及部件的监督和寿命评估^[7]等常利用研究试验堆进行加速辐照。但压水堆与研究试验堆的运行参数差异较大,尤其是中子注量率对RPV钢辐照脆化的影响十分复杂,直接用高中子注量率的加速辐照试验数据预测RPV钢辐照脆化是不适当的。因此,本文收集整理国内压水堆核电站的辐照监督试验、高通量工程试验堆(HFETR)和岷江试验堆(MJTR)的辐照试验数据,分析中子注量率对低铜($\text{Cu} \leq 0.08\text{wt}\%$)压力容器钢的影响,将研究试验堆的辐照试验数据换算成压力容器运行工况下的韧脆转变温度增量,这对通过加速辐照试验来评估新型国产压力容器的使用寿命有重要意义。

1 辐照脆化的评价方法

RPV钢在一定注量的快中子($E > 1 \text{ MeV}$)辐照下,会发生辐照脆化效应。对于辐照脆化的表征,国内外均通过测定夏比冲击试样的断裂吸收能及其随温度的变化,根据最佳拟合曲线得到冲击吸收功为标记能量(如41、56和68 J)处的标示温度之差DBTT^[8-9]。

相关研究^[10-11]表明,引起RPV钢辐照脆化的因素包括材料自身性质和外部环境因素。RPV钢自身性质^[12-14]包括材料化学成分和微观组织特性等,外部环境主要是指中子注量、中子注量率、中子能谱和辐照温度等^[15-17]。国内压水堆核电站RPV和辐照监督样品所接受的中子注量率一般处于 $3 \times 10^9 \sim 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$ 范围内,而HFETR和MJTR等研究试验堆加速辐照的中子注量率通常高于 $1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$,因此本文重点分析讨论中子注量率差异对RPV辐照脆化的影响。同时,为减少材料因素的影响,本文所指RPV钢是通过淬火和回火处理得到的体心立方结构的低合金铁素体钢(满足16MND5或A508-3的规范要求),且 $\text{Cu} \leq 0.08\text{wt}\%$, $\text{P} < 0.008\text{wt}\%$, $0.5\text{wt}\% < \text{Ni} < 0.8\text{wt}\%$ (质量分数)。国内压水堆核电站辐照监督管的辐照温度为290℃左右,为减轻辐照温度的影响,研究试验堆选择辐照温度在270~292℃之间的试验结果。

2 中子注量率对辐照脆化的影响

辐照监督管试验数据来源于国内核电站共49根辐照监督管的结果,研究试验堆数据来源于HFETR和MJTR开展的18次静态辐照试验结果。图1采用气泡图直观显示了RPV钢辐照前后转变温度增量(Δ DBTT)与中子注量率及中子注量的关系(气泡颜色和尺寸均反映韧脆转变温度(DBTT)增量),可看出, Δ DBTT同时受中子注量率和中子注量的影响,当中子注量率相当时,高注量RPV钢的DBTT增量更大,当中子注量相当时,高中子注量率引起RPV钢的转变温度上升更显著。文献^[18]中认为在288℃下缺陷扩散速率比较慢,研究试验堆中子注量率高,堆内辐照时间较短,退火不充分,而辐照监督管注量率较低,辐照时间长,有足够的时间使缺陷退火,恢复效应大。因此,低中子注量率达到高注量的辐照监督管的转变温度增量,低于高中子注量率的研究试验堆快速辐照的结果。

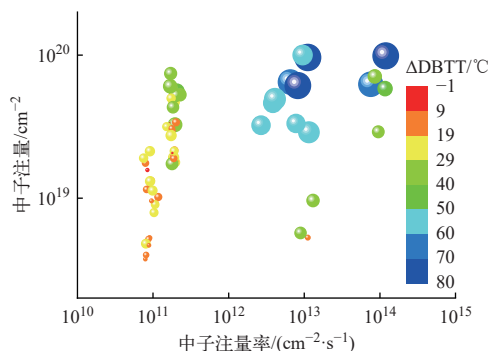


图1 转变温度增量与中子注量率及中子注量的关系气泡图

Fig. 1 DBTT variations with different neutron fluence rates and fluences

国内压水堆核电站辐照监督管的中子注量率 $\leq 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$,先前的研究^[19]表明,当中子注量率 $\leq 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$ 时,未出现明显的辐照脆化中子注量率效应。Dohi等^[20]研究A508-3锻件(Cu: 0.063wt%)发现,不同的中子注量($1 \times 10^{18} \sim 1 \times 10^{19} \text{ cm}^{-2}$)和注量率($7 \times 10^{10} \sim 5 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$),在标准偏差范围内(IECA4201标准差为 $\pm 12 \text{ }^\circ\text{C}$),注量率对低铜RPV钢的DBTT不产生影响。图2为RPV钢转变温度增量与中子注量率的关系。从图2可看出,低注量(10^{18} cm^{-2})辐照监督管的DBTT均在标准偏差范围内,而高注量(10^{19} cm^{-2})

辐照时有7个数据偏离了标准偏差范围。根据辐照监督管试验结果,认为国内压水堆核电站辐照监督管样品的辐照脆化程度基本不受中子注量率的影响。因此,当对低铜RPV进行在役评估时,若采用中子注量率 $\leq 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$ 的辐照监督数据或研究试验堆数据,不需要对中子注量率差异引起的脆化进行修正。

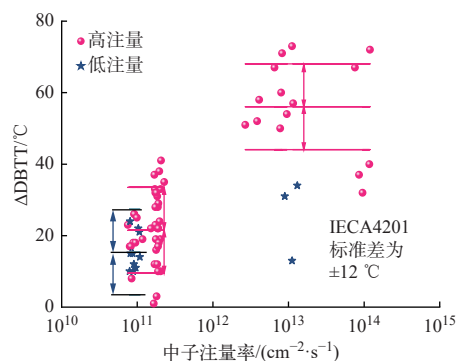


图2 RPV钢转变温度增量与中子注量率的关系
Fig. 2 DBTT variations of RPV steel with different neutron fluence rates

HFETR和MJTR研究试验堆的中子注量率 $> 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$,从图2还可看出,低注量(10^{18} cm^{-2})辐照的数据较少,高注量(10^{19} cm^{-2})试验时有6个数据也偏离了标准偏差范围。Kasada等^[21]研究发现,在 $2 \times 10^{12} \sim 6.3 \times 10^{13} \text{ cm}^{-2} \cdot \text{s}^{-1}$ 范围内,中子注量率对低铜RPV钢的韧脆转变温度增量的影响呈现先减小后增加的现象。Odette^[22]认为低铜RPV钢在中子注量率 $> 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$ 条件下,中子注量率效应不明显,不能直接得出存在注量率效应的结论。至今,国际上中子注量率($> 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$)对低铜RPV的注量率效应没有形成统一的结论。但目前新型国产RPV钢均是通过研究堆加速辐照来评估其耐辐照性能,因此,分析中子注量率差异对辐照脆化效应的影响十分重要,只有有效关联研究堆与压水堆辐照监督的试验结果,才能够准确、可靠地验证新型国产RPV的设计寿命。

3 不同中子注量率试验结果的关联

研究试验堆高中子注量的结果分散性较大,可能与中子注量差异较大有关($3 \times 10^{19} \sim 9 \times 10^{19} \text{ cm}^{-2}$),为减少中子注量的影响,需对不同中子注量的结果进行归一化处理。归一化处理的关

键是选择合适的辐照脆化模型来拟合出我国低铜 RPV 钢的转变温度增量与中子注量的关系曲线。目前美国的 RG1.99(Rev.2)^[23]、日本的 JEAC4201^[24] 和法国的 FIS^[25] 等计算模型均可用来预测 ΔRT_{NDT} 随中子注量增加的变化情况。FIS 是对大量监督数据和试验堆辐照数据上限值的拟合, 明显高于我国 RPV 辐照性能数据。RG1.99-2 是基于美国早中期投运核电站 RPV 辐照监督管的数据库建立的, 对美国核电站 RPV 辐照脆化预计是可以接受的, 但缺少低铜含量数据, 而文中的 RPV 钢铜含量均不超过 0.08%, 应用 RG1.99-2 是不恰当的。日本 JEAC4201 模型的建立利用 1990 年以前的 211 个母材监督数据和 88 个焊缝监督数据, 其预测结果高于我国核电站辐照监督数据。虽然这些经典的模型不完全适用我国低铜 RPV 钢的变化趋势(图 3), 但这些模型均考虑了 RPV 化学成分的有关于中子注量的指数函数。本文不考虑化学成分影响, 因此只做关于中子注量的简单指数拟合。

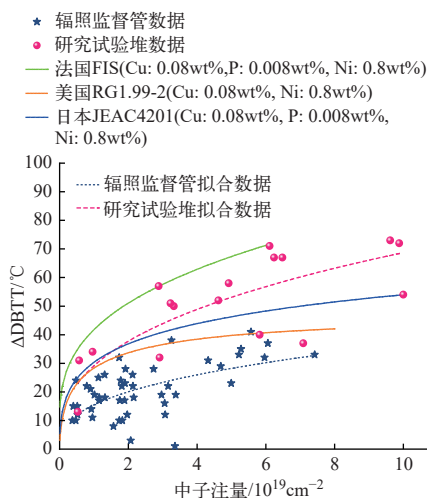


图3 韧脆转变温度增量与中子注量的关系
Fig. 3 DBTT variations with neutron fluence

中子注量归一化过程^[26]: 首先, 分别对辐照监督和试验堆不同中子注量的试验结果进行指数拟合, 得出转变温度增量与中子注量的关系, 见式(1)和式(2)。其次, 根据缺陷湮没理论模型计算得出 1 条转变温度增量-中子注量率关系曲线^[27], 如图 4 所示。最后, 将研究堆和辐照监督数据分别利用式(1)和式(2)将快中子注量归一化至 $2 \times 10^{19} \text{ cm}^{-2}$, 并将数据点标注于图 4。从图 4 可看

出, 辐照监督的 $\Delta DBTT$ 主要分布在计算曲线的上升阶段, 研究堆辐照 $\Delta DBTT$ 主要分布在计算曲线的平台阶段(6 个试验结果低于计算结果), 辐照监督管与研究试验堆所得低铜 RPV 钢的 $\Delta DBTT$ 和计算曲线较为吻合。因此, 利用式(1)和(2)可将研究试验堆快速辐照试验的数据换算成核电站压力容器运行工况下的转变温度增量。

$$\Delta DBTT = 15.47(F \times 10^{-19})^{0.37} \quad (1)$$

$$\Delta DBTT = 29.15(F \times 10^{-19})^{0.37} \quad (2)$$

其中, F 为快中子注量($E > 1 \text{ MeV}$), cm^{-2} 。

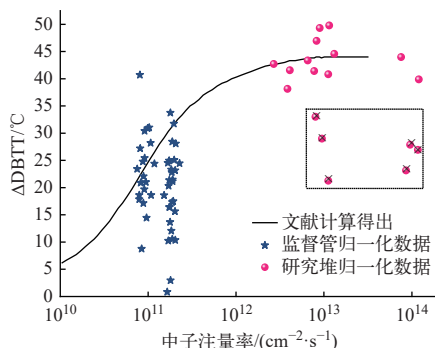


图4 RPV 钢转变温度增量与中子注量率的关系
Fig. 4 DBTT variations of RPV steel with different neutron fluence rates

4 结论

国内 RPV 钢的研究与应用主要是低铜铁素体钢, 常利用 HFETR 和 MJTR 等快速辐照来研究 RPV 钢服役性能, 这与压力容器实际运行工况的主要差异是中子注量率的分布。研究发现, 低铜 RPV 钢的韧脆转变温度增量同时受中子注量率和中子注量的影响, 在同一注量水平下, 研究试验堆的高中子注量率($> 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$)比核电站反应堆压力容器辐照监督管的低中子注量率($\leq 1 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$)引起的转变温度上升更显著。通过对辐照监督管和研究试验堆不同中子注量的试验结果进行指数回归, 将研究试验堆快速辐照试验结果与辐照监督试验结果进行关联, 两种试验结果可以实现相互转换。将研究试验堆的快速辐照试验数据换算成压力容器实际服役工况下的转变温度上升值意义重大, 能够更准确、可靠地验证压力容器设计寿命和评估剩余使用寿命, 同时可为新建核电站压力容器设计提供可靠数据。

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