

· 综述 ·

一体化快堆燃料包壳用铁素体/马氏体钢研究进展



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摘要: 一体化快堆采用金属燃料, 燃料包壳工作温度介于 350~630 °C, 工作周期将最长达到 50 000 h 以上。随着工作周期延长, 包壳承受的快中子辐照剂量将由目前的 MOX 燃料的 80 dpa 增加到 150~300 dpa, 因此, 发展承温能力高、抗辐照性能优异的新型包壳材料, 成为一体化快堆发展的重要组成部分。文章概述了铁素体/马氏体钢(铁马钢)材料的发展背景, 分析了不同牌号铁马钢力学性能、辐照性能特点及不同合金元素对钢的性能的影响规律, 进而提出了适用于一体化快堆堆芯组件用铁马钢材料的合金优化策略。利用优化策略对 HT9 钢进行初步改进, 改进后的 HT9G 进行室温拉伸测试和 700 °C/100 MPa 持久寿命测试。结果表明, 改进后的合金具有良好的拉伸强度和持久寿命, 其室温屈服强度可达 880 MPa, 比 T91 钢提高约 310 MPa, 比 HT9 和 T92 钢提高约 80~120 MPa; 700 °C、100 MPa 持久寿命达 372~385 h, 远高于 HT9 同条件下的 70~82 h。显示出强韧化设计的有效性, 为组件结构材料的进一步优化设计和长时持久强度提升奠定了基础。

关键词: 一体化快堆; 燃料包壳材料; 铁素体/马氏体钢; 辐照性能; 持久强度

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Research Progress on Ferritic/Martensitic Steels for Integrated Fast Reactor Fuel Cladding

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Abstract: The integrated fast reactors use metallic fuel, with fuel cladding operating at temperatures ranging from 350 °C to 630 °C and a service lifespan exceeding 50 000 hours. As the operational cycle extends, the fast neutron irradiation dose of the cladding will increase from the current 80 dpa of MOX fuel to 150 dpa-300 dpa. Therefore, the developing new cladding materials with high thermal resistance and excellent irradiation performance has become an important part of the integrated fast reactor development. This paper summarized the background of ferritic/martensitic steels (FM steels) development, analyzed the mechanical properties and irradiation performance of various FM steels, and investigated the influence of different alloying elements on these properties. Based on these analyses, alloy optimization strategies for FM steels suitable for integrated fast reactor fuel cladding were proposed. The optimization strategy was initially modified for HT9 steel, and the modified HT9G was tested for room temperature tensile test and 700 °C/100 MPa creep rupture life. The results show that the modified alloy exhibits excellent tensile strength and creep rupture life, its room temperature yield strength reaches 880 MPa, which is approximately 310 MPa higher than that of T91 steel and 80 MPa-120 MPa higher than that of HT9 and T92 steels. Under conditions of 700 °C and 100 MPa, the creep rupture life is 372 hours-385 hours, significantly exceeding the 70 hours-82 hours of HT9 under the same conditions. It shows the effectiveness of toughening design, laying the foundation for further optimization of component structural materials and enhancement of long-term durability and strength improvement.

Key Words: Integrated Fast Reactor; Fuel cladding; Ferritic/martensitic steel; Irradiation performance; Creep property

一体化闭式循环快堆核能系统(以下简称“一体化快堆”)是在同一地址建设快堆、干法后处理厂和燃料生产线,通过高质量闭式燃料循环,实现铀

资源的高效利用和废物最小化是我国核能“三步走”战略的重要一环^[1]。一体化快堆的建设基于钠冷快堆^[2],但与钠冷快堆相比,一体化快堆燃料形

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式、换料周期、燃耗等发生明显变化。一体化快堆采用金属合金燃料,其燃料燃耗由快堆的 10at.%^[3] (at.%为燃耗单位,指核燃料中裂变的原子数占总重金属原子数百分比)提高至 15~20at.%^[4],燃料工作周期由近 20 000 h 增至 50 000 h 以上,相应地堆芯材料所承受的辐照剂量由 80dpa 增加至 150~300 dpa。燃料包壳是燃料组件中服役环境最为苛刻的部件,不仅承受高辐照剂量,其工作温度和寿命末承受的应力分别达到 600~630 °C 和 35~55 MPa。“长寿命、深燃耗、高应力”的服役工况对包壳材料的耐辐照性、高温长时力学性能及组织稳定性都提出了更高的要求。

奥氏体耐热钢^[5-6]、9%~12%Cr 铁素体/马氏体耐热钢(以下简称“FM 钢”)^[7-8]和纳米氧化物强化的铁素体或铁素体/马氏体钢(以下简称“ODS 钢”)^[9-10]是一体化快堆包壳材料的主要候选材料。15Cr-15Ni 体系奥氏体耐热钢是钠冷快堆包壳材料的主要候选材料,承温能力可以达到 650~700 °C,其不足之处在于快中子辐照剂量达到 80~100 dpa 后易于发生肿胀,严重时可能引起包壳破裂^[5-6, 11]。ODS 钢具有良好的高温强度和抗辐照肿胀性能,被认为是一体化快堆长远发展的包壳候选材料。该材料目前研究主要集中在材料体系的发展^[9-10, 12-14],中试或近工程化级别的制备技术研究等方面^[15]。FM 钢具有良好的工程制造和应用经验,抗辐照肿胀性能得到验证^[16-17],是一体化快堆包壳材料极具潜力的候选材料,其主要不足在于高温强度偏低而使得承温能力通常限制在 620 °C 以下^[17]。自 1980 年代以来,世界范围内针对核用 FM 钢的研发取得了长足进步,提出了系列强韧化技术手段,使得 FM 钢的承温能力由早期 HT9 钢的 550~580 °C 提升到 T92 钢的 600~620 °C^[18]。尽管新近发展的 SAVE12AD 钢^[19]、MARBEN^[20]和 G115^[21]使用温度达到了 630~650 °C,然而其采用的 $w[\text{Co}]3\%$ 、 $w[\text{B}]0.015\%$ 合金化策略不适宜于核能应用。因此,突破核用 FM 钢强韧化技术瓶颈,发展承温能力满足高燃耗燃料包壳发展

需求的 FM 钢,成为我国一体化快堆技术发展的重要部分。

本文简要回顾了 FM 钢发展历史,汇总分析了系列 FM 钢性能以及关键合金元素对 FM 钢性能的影响,提出了提升 HT9 钢承温能力的强韧化策略。对现有实验研究结果进行了汇总,验证了强韧化的有效性,显示出改进型 FM 钢满足一体化快堆包壳应用的强度需求,具备高温应用的前景。

1 FM 钢发展及其在钠冷快堆中的应用

FM 钢的发展起源于火电站热效率提升的技术需求,促进了火电机组实现了亚临界向超临界、超超临界的跨越^[21]。按照 600 °C、10 万小时持久强度进行划分,火电用 FM 钢主要经历了 5 个发展阶段^[22],见表 1。

核用 FM 钢继承了表 1 所示的发展脉络,并基于抗辐照性能、与冷却剂相容性、服役温度、承受的应力等的差异化而有所不同,其主要钢种及成分见表 2。

20 世纪 70 年代美国在发展钠冷快堆技术过程中选用 12%Cr 型 HT9 钢制作燃料组件(前期用于外套管和包壳管,后期包壳改用 15Cr-15Ni 钢),是辐照性能数据最多、研究最为全面的钢种^[23]。蒸汽发生器回路选用 2.25Cr-1Mo 体系耐热钢,并沿用至今^[24]。同时期,欧洲钠冷快堆燃料组件材料选用 FV448 钢、1.4914 或 EM10 钢,并发展出了 EM12 钢^[24]。为了解决 2.25Cr-1Mo 体系耐热钢在高温钠中的脱碳问题和高温强度不足的问题,美国发展的快堆蒸汽发生器改选 9Cr 型 FM 钢,发展出改进型 9Cr-1Mo-V-Nb 钢^[25],即在超超临界火电机组中获得广泛应用的 T/P91 钢。通过添加 B 和采用 W 和 Mo 协同强化的合金化措施,在 T/P91 钢基础上发展出 T/P92 钢^[26]。日本原子能机构为满足钠冷快堆技术发展需求,开展了 PNC-FM 钢的研制,并应用于 JSFR (Japan Sodium-cooled Fast Reactor)^[27]。俄罗斯

表 1 FM 钢发展代系强韧化策略及代表钢种

代系	强韧化策略	600 °C、10 万小时持久强度/MPa	代表钢种
初始	采用 Mo 进行强化	40	T22、T9、EM10
第 1 代	优化 Mo、添加 V 或 Nb	60	HT9、EM12
第 2 代	降低 C, 优化 Nb 和 V, 添加 N	100	T91
第 3 代	以 W 代 Mo, 添加 B 或 Cu	140	T92、E911、T122
第 4 代	高 W、B, 添加 Co、Cu	180	SAVE12AD、MARBEN、G115

主导的钠冷快堆燃料组件用FM钢牌号为EP450,其与美欧日材料体系和组织控制差异显著,独树一帜^[28]。近期,美国泰拉能源公司提出了改型HT9钢用于支撑行波堆(钠冷快堆的一种)技术的发展^[29],俄罗斯推出了EK181、ChS139和EP450-ODS钢以满足新型钠冷快堆技术发展需求^[30],韩国采用优化成分的措施发展出9%Cr体系的FC92钢用于制造包壳管^[31]。

为满足聚变堆和加速器驱动次临界系统(Accelerator Driven Sub-critical System, ADS)技术发展需求,低活化FM钢应运而生,先后发展出9Cr-2WVTa、F82H、JLF-1、Eurofer97、CLAM等钢种,其高温强度与T/P91钢相当,承温能力通常控制在600℃左右^[32]。2000年以来发展的SAVE12AD、MARBEN和G115主体成分为Fe-9Cr-3W-3Co,采用Cu、B、N、V、Nb进行复合强化^[19-21],使用温度可以达到650℃。不足之处在于,典型钢种中的 $w[\text{Co}]$ 通常高达3%, $w[\text{B}]$ 大多控制在0.01%~0.015%,添加一定量的Cu(如G115钢),在快中子辐照环境中引起嬗变反应而生成⁶⁰Co、He或Cu团簇,使其不适合应用在承受高辐照剂量的燃料组件上。

通过机械合金化的方法在FM钢中添加一定量的纳米Y₂O₃颗粒^[30,33-34],可以有效提高其高温强度,由此发展出ODS钢满足了高承温能力的技术需求,代表牌号有ODS-Eurofer97,EP450-ODS,12YWT等。为了满足规模化应用需求,研发适用于钠冷快堆和一体化快堆包壳服役环境的材料体系、发展近工程化级别的制备技术、建立材料性能数据库(集)是ODS钢发展的主要趋势^[15]。

2 FM钢典型性能

2.1 辐照性能

2.1.1 辐照肿胀

燃料组件中的包壳和外套管经受150~300 dpa剂量辐照,组织中形成孔洞而产生肿胀^[35],使得组件发生一定程度的变形,严重时引起破损,对反应堆安全运行造成不利影响。FM钢抗辐照性能总体偏优,但辐照行为与钢中的Cr含量密切相关。图1^[35-38]显示,随着快中子辐照剂量由100 dpa增加到200 dpa,12%Cr型的HT9钢辐照肿胀率不足1.5%,而9%Cr型T91钢的肿胀率可达到2.5%以上,即12%Cr钢在高辐照剂量条件下的抗辐照肿胀性能优于9%Cr钢。Dvoriashin A M等^[39]的研究显示,在90 dpa辐照剂量下,EP-450钢最大肿胀率不超过1%。与HT9钢相比,EP-450钢表现出较大的肿胀率,这可能与该钢中较高含量 δ -铁素体,促进了辐照肿胀有关。几种典型的低活化钢在多离子束辐照下的肿胀率如图2^[40-41]所示。随辐照剂量的增加,均出现了低稳态膨胀向高稳态膨胀转变的过渡剂量。在多离子束辐照剂量小于40 dpa之前,JLF-1钢表现出良好抗辐照肿胀性,肿胀率仅为0.2%左右,辐照超过过渡剂量时,肿胀率迅速上升。与JLF-1相比,F82H钢过渡剂量相对较低。几种典型ODS钢的辐照肿胀行为如图3所示^[7,40]。ODS-MA957与ODS-YWT钢辐照肿胀率表现出明显的辐照强度相关性,显示出良好的抗辐照肿胀性。其中,ODS-MA957在辐照剂量400 dpa时,肿胀率低于1%。

2.1.2 辐照硬化

辐照硬化表现为FMS钢辐照前后屈服强度和

表2 各代表钢种主要成分范围
Table 2 The main chemical composition range of each representative steel grade

钢种	成分范围(质量分数)/%
HT9	Fe-12Cr-1Mo-0.6Mn-0.6Ni-0.52W-0.38Si-0.3V-0.2C ^[23]
T/P9	(8~9.5)Cr-(0.85~1.05)Mo-(0.3~0.6)Mn-(0.2~0.5)Si-(0.18~0.25)V-(0.08~0.12)C-(0.06~0.1)Nb-(0.03~0.07)N-(≤ 0.4)Ni ^[25]
T/P92	(0.07~0.13)C-(≤ 0.05)Si-(0.3~0.6)Mn-(8.5~9.5)Cr-(≤ 0.4)Ni-(0.3~0.6)Mo-(0.15~0.25)V-(0.04~0.09)Nb-(1.5~2)W-(0.05~0.07)N-(0.001~0.006)B ^[26]
PNC-FM	(0.09~0.15)C-(0.1~0.5)Si-(0.2~0.6)Mn-(≤ 0.02)P-(≤ 0.1)S-(10.5~11.5)Cr-(0.3~0.6)Mo-(0.1~0.3)V-(0.02~0.06)Nb-(0.2~0.5)Ni-(≤ 0.2)Cu-(0.1~0.3)Co-(0.1~0.3)W-(0.001~0.003)B ^[27]
EP-450	(11~13.5)Cr-(1.2~1.8)Mo-0.8Mn-0.5Si-(0.3~0.6)Nb-(0.05~0.3)Ni-(0.1~0.3)V-(0.1~0.15)C-0.004B ^[28]
G115	(0.06~0.1)C-(8~9.5)Cr-(2.5~3.5)W-(2.5~3.5)Co-(0.2~0.8)Mn-(0.1~0.3)V-(0.1~0.5)Si-(0.8~1.2)Cu-(0.03~0.07)Nb-(0.006~0.01)N-(0.01~0.016)B-(≤ 0.03)Ni ^[21]
ODS-Eurofer97	(9~10)Cr-1W-0.2V-0.1Ta-0.1C-0.2Si-(0.3~0.5)Y ₂ O ₃ ^[33]
EP450-ODS	13Cr-2Mo-0.12C-0.5Y ₂ O ₃ ^[30]
12YWT	12.3Cr-3Mo-0.4Ti-0.25Y ₂ O ₃ ^[34]

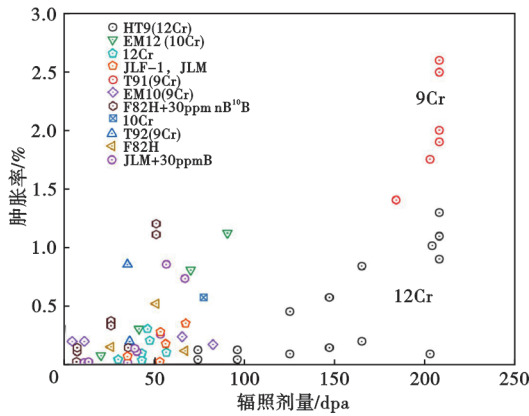


图1 快中子辐照剂量与FM钢肿胀率关系

Fig. 1 Relationship between fast neutron irradiation dose and swelling rate of FM steels

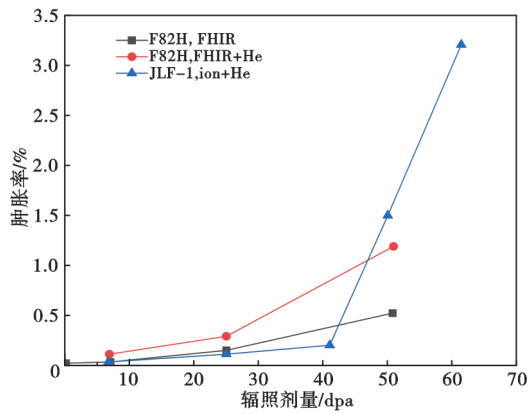


图2 多离子束辐照下辐照剂量与低活化钢肿胀率关系

Fig. 2 Relationship between irradiation dose and swelling rate of low activation steel under multi-ion beam irradiation

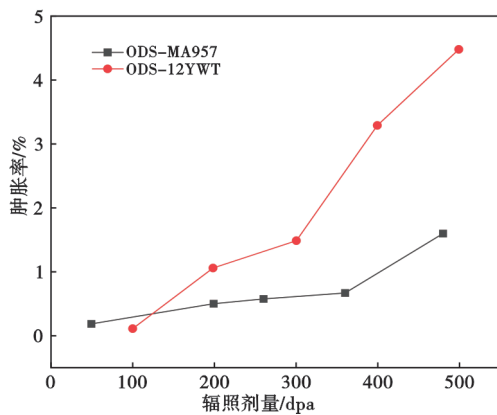


图3 快中子辐照剂量与ODS钢肿胀率关系

Fig. 3 Relationship between fast neutron irradiation dose and swelling rate of ODS steels

抗拉强度明显增大,这与辐照温度和辐照剂量密切相关。图4(a)^[42]指出在300~350℃范围内FMS钢表现出明显的辐照硬化现象,相同辐照剂量条件下

9%Cr型EM10和T91钢屈服强度增量($\Delta\sigma_y$)高于12%Cr型的HT9和PNC-FMS钢。在365℃辐照6.4~7.6 dpa后,T91钢的 $\Delta\sigma_y$ 介于149~156 MPa,而HT9钢的 $\Delta\sigma_y$ 介于345~382 MPa。在390℃辐照12 dpa后,T91钢的 $\Delta\sigma_y$ 为307 MPa,而HT9钢的 $\Delta\sigma_y$ 随热处理制度变化在125~231 MPa范围内变化。随辐照温度升高到400℃,不同Cr含量的FMS钢辐照硬化显著减小。图4(b)^[43]显示,辐照硬化随辐照剂量增大而增大,在70~80 dpa左右变化趋于平衡。与FM钢类似,低活化钢与ODS钢均表现出明显的辐照硬化,辐照导致钢材屈服强度显著提高。图5^[44-46]表明,F82H,JLF-1与EUROFER钢屈服强度增量($\Delta\sigma_y$)随辐照剂量增加,增长趋势相似。辐照剂量30~40 dpa时,辐照硬化逐渐趋于稳态,屈服强度增量均趋近于500 MPa。如图6所示^[47],ODS钢辐照条件下表现出明显的辐照硬化行为,在相同辐照温度范围内,不同种类的ODS钢屈服强度增量($\Delta\sigma_y$)随辐照强度的增加呈现出上升后趋于稳定的规律变化,辐照剂量40~50 dpa时,辐照硬化逐渐趋于稳态。

2.1.3 辐照脆化

辐照脆化表现为拉伸塑性(伸长率)降低、冲击吸收能量减小和韧脆转变温度(DBTT)升高。随着辐照硬化增加,拉伸塑性(伸长率)降低^[48]。通常地,9%~12%Cr钢辐照后均发生冲击吸收能量减小和韧脆转变温度升高的现象,其中12%Cr钢变化幅度约为9%Cr钢的2倍,如图7所示^[49]。关于HT9钢的研究显示,在365℃辐照10~17 dpa后,DBTT变化趋于饱和,如图8所示^[49]。9Cr-2W低活化钢辐照后DBTT变化幅度低于9Cr-1Mo,显示出W合金化的有益之处^[50]。如图9所示^[50]低活化钢表现出与FM钢类似的辐照脆化趋势,在辐照条件下,辐照剂量的上升导致韧脆转变温度(DBTT)的显著上升。EUROFER97钢相较于F82H钢,其DBTT变化趋势更快,300℃辐照0.3~2.4 dpa后,EUROFER97钢DBTT从7℃上升至78℃。0-5 dpa辐照后F82H钢DBTT由20℃上升至107℃。

2.2 蠕变性能

图10^[51-57]汇总了9~12%Cr型FM钢蠕变性能。根据持久应力-寿命关系和热强参数方程开展了不同FM钢在600~650℃长时持久强度预测,结果列入表3。可以看出,在高温条件下,T92、PNC-FM和FC92钢持久强度优于HT9钢和T91钢,这是由于采用以W代Mo、通过添加N促进MX相析出或添加B

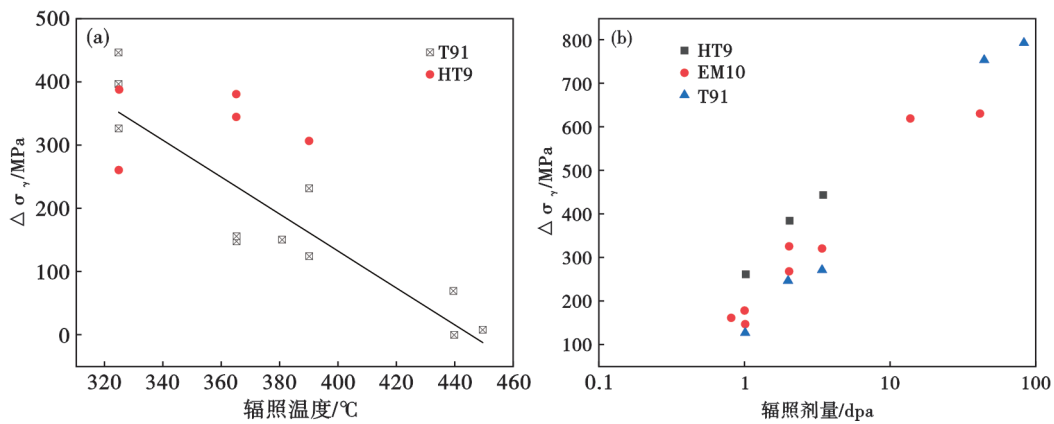


图4 9%~12%Cr钢辐照硬化行为:(a)辐照温度与 $\Delta\sigma_y$ 关系,(b)300 °C辐照剂量与 $\Delta\sigma_y$ 关系

Fig. 4 Irradiation hardening behavior of 9%~12% Cr steels: (a) relationship between irradiation temperature and $\Delta\sigma_y$, (b) relationship between irradiation dose at 300 °C and $\Delta\sigma_y$

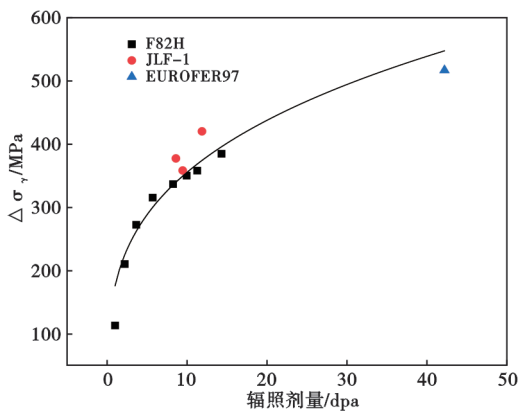


图5 低活化钢辐照硬化行为:300~325 °C辐照剂量与 $\Delta\sigma_y$ 关系

Fig. 5 Irradiation hardening behavior of low activation steel: (a) relationship between irradiation temperature and $\Delta\sigma_y$, (b) relationship between irradiation dose and $\Delta\sigma_y$ at 300 °C~325 °C

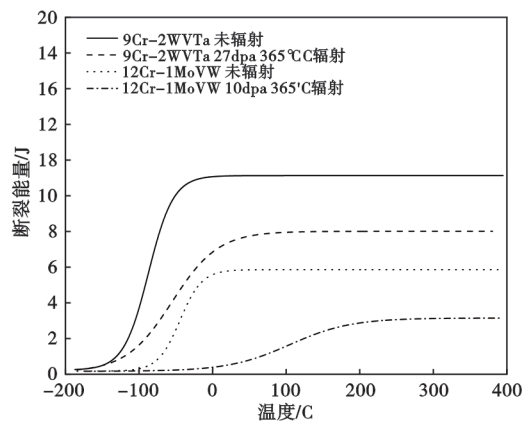


图7 365 °C辐照对 HT9 (12Cr-1MoVW) 和 T91 (9Cr-2 WVTa)钢冲击性能的影响

Fig. 7 Impact of 365 °C irradiation on the impact toughness of HT9 (12Cr-1MoVW) and T91 (9Cr-2 WVTa) steels

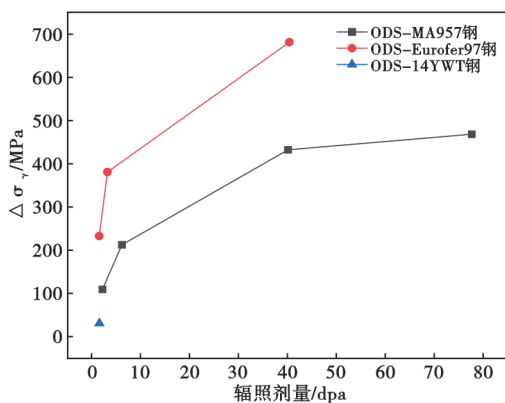


图6 ODS钢300~340 °C辐照剂量与 $\Delta\sigma_y$ 关系

Fig. 6 The relationship between irradiation dose and $\Delta\sigma_y$ of ODS steel at 300 °C~340 °C

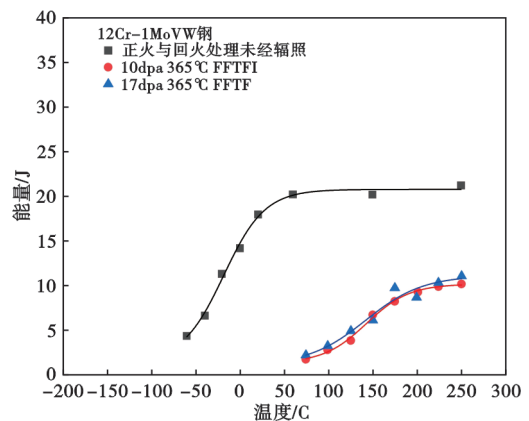


图8 辐照剂量对 HT9(12Cr-1MoVW)钢冲击性能的影响

Fig. 8 Effect of irradiation dose on the impact toughness of HT9(12Cr-1MoVW) steel

的结果。通过进一步优化成分和改善合金化程度,是核用FM钢强度提升、承温能力提高的关键所

在^[58]。图11^[40,59-60]汇总了不同ODS钢蠕变性能。其中650 °C下ODS-12Cr钢表现出最为优异的持久强

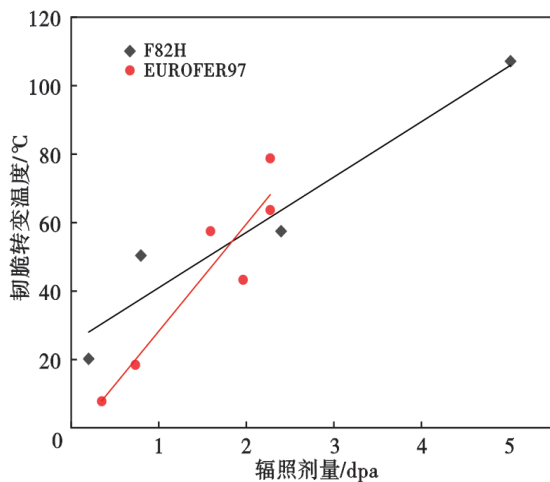


图9 300 °C下辐照对F82H与EUROFER钢韧脆转变温度影响

Fig. 9 Effect of irradiation at 300 °C on DBTT of F82H and EUROFER steels

度。这主要由于在 ODS 钢中大量弥散存在的氧化物颗粒与小角度高强度晶界^[59],使得 ODS 钢的蠕变强度普遍高于 FM 钢。

Jeong E H 等^[61]的研究指出,适量添加 B 元素使 FC92 在 650 °C、100 MPa 下的断裂时间达到 7 000 h,而不含 B 的钢蠕变断裂时间仅有 2 500 h 左右。科研人员对 EP450 的蠕变性能做了研究^[62],发现在 60 MPa 载荷,700 °C 蠕变环境下合金的蠕变断裂寿命达到 1 000 h。当处于 140 MPa,650 °C 蠕变环境下合金的蠕变速率为 $1.2 \times 10^{-2} \text{ h}^{-1}$ 。120 MPa,700 °C 蠕变环境下,EP450 蠕变速率为 $8 \times 10^{-2} \text{ h}^{-1}$ 。Uehira A 等^[63]探究了 PNC-FM 钢的辐照蠕变状况,发现 PNC-FM 钢的抗辐照肿胀性能优异,应力引起的肿胀几乎可以忽略不计。

3 FM 钢性能退化机制及合金化策略

3.1 FM 钢性能退化机制

9%~12%Cr 型 FM 钢典型组织如图 12、图 13 所示^[64],按照介观和微观次序细分为原奥氏体晶粒(PAG)、马氏体板条、析出相和位错。高温和应力作用下,PAG 和马氏体板条界面上的碳化物易于粗化,导致其对板条、位错钉扎能力下降,板条内位错合并反应加速,促进了板条宽化和亚晶形成,是 FM 钢高温力学性能下降的主要原因^[65]。如何通过合金设计优化等手段降低碳化物粗化速率,使析出相更为弥散、细小,在服役过程中可以继续有效钉扎位错,延缓亚晶形成,是提高该材料的高温持久强度,进而研发出承温能力更为优异的、适用于一体化快堆深燃耗燃料组件使用的铁马钢所首先要解决的问题。

3.2 燃料包壳用 FM 钢成分优化策略

FM 钢高温强度与板条马氏体、强化相的稳定性密切相关。Cr 是典型的铁素体形成元素,也是 $M_{23}C_6$ 形成元素,能够促进碳化物的生成^[66],过度添加会使蠕变强度降低。Cr 含量过高会导致固溶处理后形成 δ -铁素体相,并促进 FeCr 化合物析出,使强度和韧性降低^[67]。在 FM 钢中,Cr 含量的增加也会增大 Z 相的析出驱动力^[68],促使 Z 相析出。Z 相为有害相,对 FM 钢力学性能产生不利影响。Cr 含量与 FM 钢的辐照行为也存在较大的关系,随着 Cr 含量的升高,铁马钢的辐照硬化程度增大^[69],但 Cr 含量升高可以提高 Cr 的扩散率,减少钢中的空位型缺陷,提高铁马钢的抗辐照肿胀性能^[42]。此外,关于 FM 与高温钠液相容性研究显示, $w[\text{Cr}]$ 含量低于 10.5% 的钢易于发生脱碳现象^[70]。

V、Nb 和 Ta 为 MX 形成元素,通过与 C 或 N 结合

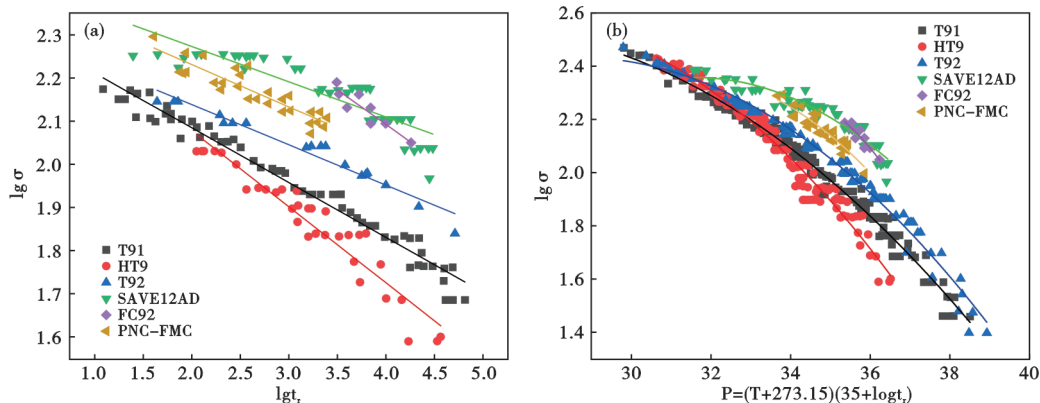


图 10 FM 钢(a)650 °C 持久应力-寿命关系曲线和(b)热强参数曲线

Fig. 10 (a) Stress-rupture life relationship curve of FM steel at 650 °C, and (b) Larson-Miller analysis of long-term strength of FM steels

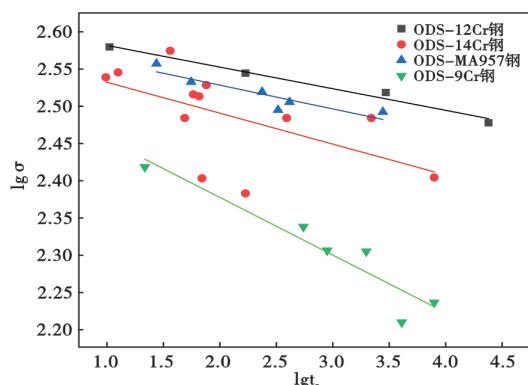


图 11 ODS 钢 650 °C 持久应力-寿命关系曲线

Fig. 11 Stress-rupture life relationship curve of FM steel at 650 °C

表 3 不同牌号高铬铁素体/马氏体钢 600~650 °C 蠕变应力预测值

Table 3 Predicted creep stress of various grades of high-chromium FM steels at 600 °C~650 °C MPa

牌号	600 °C/ 50 000 h	630 °C/ 50 000 h	650 °C/ 50 000 h	
	热强参数 方程	热强参数 方程	热强参数 方程	持久应力- 寿命关系
T91	103	72	55	55
HT9	90	56	39	40
T92	123	87	64	77
SAVE12AD	173	133	106	113
FC92	187	133	92	63
PNC-FMC	152	108	78	91

形成致密分布的碳化物和碳氮化物,对铁素体基体及晶界起到钉扎作用,加强析出相强化作用^[71]。Nb 的添加,也可以增加钢的抗拉强度、屈服强度和持久强度,但会降低合金的冲击韧性^[72-74]。Onizawa T 指出,在高 Cr 铁马钢中,考虑强度和塑韧性的平衡,认为 $w[\text{Nb}]$ 对钢的影响在 0.01% 时趋于饱和。作为碳化物形成元素, Nb 的添加会影响碳化物的析出,

对 MX 相退化行为具有不利影响,促进 Z 相形成和粗化^[72]。Ta 能够细化马氏体组织,使沉淀相均匀化^[75],且 Ta 能够细化碳化物^[76]。在低 C-10Cr-2Mo 钢中, $w[\text{V}]$ 和 $w[\text{Nb}]$ 的含量分别为 0.1%~0.18% 和 0.05% 时,强化效果最优^[77]。Nb(C, N) 主要在短时间内起析出强化作用,而 V(C, N) 的析出强化作用可以延续到长时间^[78]。此外, V 和 Nb, Ta 联合添加的强化作用效果更加明显,说明 V 和 Nb, Ta 组成的沉淀相是相互关联的关系^[78-80]。

Mo 和 W 是促进合金固溶强化的主要元素,能够固溶进入 Fe 基体中提高蠕变强度。Mo 能够提高钢的热强性及淬透性,并且提高钢的高温强度和蠕变断裂强度^[81]。W 的加入降低了 M_s 点,形成了精细的亚晶结构,减小了初始亚晶粒尺寸,延缓了 $M_{23}C_6$ 颗粒的粗化从而提高了蠕变强度^[82]。将 9Cr 钢中 $w[\text{W}]$ 从 2% 提升到 3%, MC、MX 和 Z 相的粗化速率显著降低,且 W 抑制了 MX 相向 Z 相转化^[83]。Abe F.^[84] 对比了相同 1.50Mo 当量下 W、Mo 元素对 9Cr 钢 Laves 相析出和持久强度的影响, Mo 当量的定义为 $0.5 W / (0.5 W + Mo)$, 平衡 Laves 相分数随着 W 含量的提升而增大,持久强度则随着 W 含量的提高而增强,认为 W 元素的添加相比于 Mo 元素而具有更好的强化效果。

C 和 N 是奥氏体的组成元素,能够抑制 δ -铁素体的形成。C 和 N 含量直接影响 FM 钢中碳化物和碳氮化物的析出与粗化,随着 C 含量减少和 N 含量增加, MX 碳氮化物显著增多^[85]。9%~12%Cr 钢中 $w[\text{C}]$ 应控制在 0.08%~0.16% 或 0.09%~0.14% 之间效果较好^[86]。N 是提高 9%Cr 耐热钢的必需元素,其主要作用是和 V、Nb 生成 MX 相,更好钉扎晶界,从而提高高温强度和蠕变性能。N 含量过高时,会导致

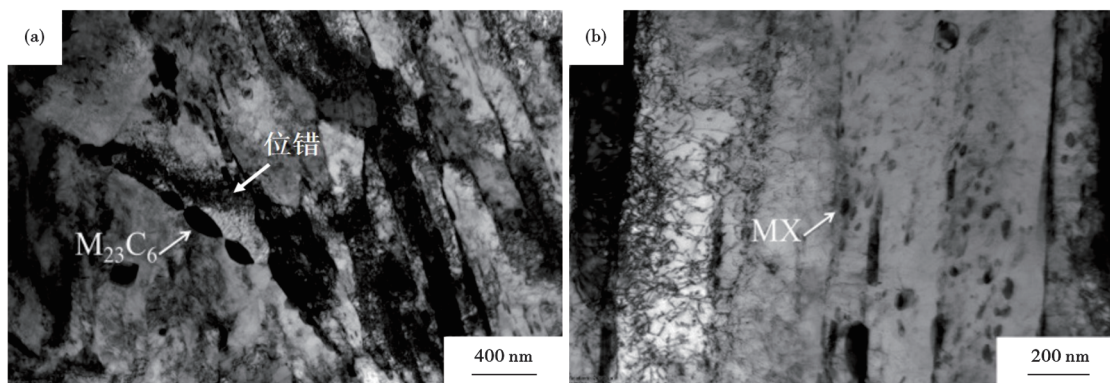


图 12 HT9 钢热处理后的显微组织:(a)位错,(b)析出相

Fig. 12 Microstructure of HT9 steel after heat treatment : (a) dislocation, (b) precipitated phase

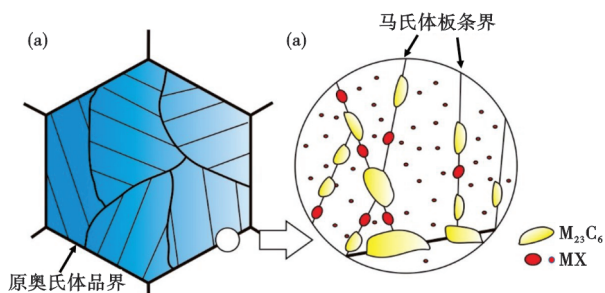


图 13 HT9 钢热处理后马氏体精细结构示意图: (a) 奥氏体晶粒, (b) 马氏体板条和 $M_{23}C_6$

Fig. 13 Martensitic fine structure schematic of HT9 steel after heat treatment: (a) austenitic grains, (b) martensitic slats and $M_{23}C_6$

蠕变阶段 Z 相的生成^[87], Z 相析出时尺寸较大, 同时析出伴随着 MX 相的消耗, 从而降低了 MX 相弥散强化的效果, 但不会促进沉淀强化, 且使 9%~12%Cr 铁素体耐热钢的蠕变寿命下降, 对蠕变性能产生不利影响, 所以应避免 N 含量过高生成 Z 相。

B 属于间隙固溶元素, 特别适合在晶界处的空位中存在, 稳定晶界提高晶界强度。B 能够明显提高 12%Cr 钢在 650~750 °C 时的蠕变断裂强度, 而当 $w[B]$ 超过 0.1% 时, 蠕变强度反而下降^[88-90]。Hättestrand 等指出, B 会在晶界处偏析, 而且主要聚集在 $M_{23}C_6$ 沉淀相中, 能提高 $M_{23}C_6$ 相的稳定性, 降低其粗化的速率^[91-92]。B 扩散到 $M_{23}C_6$ /基体界面, 阻碍元素扩散, 降低碳化物粗化速率, 从而提高材料的强度^[93]。蠕变过程中, B 抑制 $M_{23}C_6$ 相颗粒粗化而延缓板条宽化, 同时促进细小 MX 析出^[94], 提高了长期蠕变强度。Abe F.^[94]通过对不同 N 含量 FM 钢的研究发现, B 与 N 生成了粗大的 BN 化合物, 降低高温持久强度。因此, 在不生成 BN 的条件下, B 含量越高对持久性能越有利。

3.3 新型 HT9G 钢研制及长时性能预测

基于元素作用的理论分析, 通过添加 Nb、Ta 促进 MX 相析出, 调节 W/Mo 和 B/N 比等手段, 发展出成分改性的 HT9 钢(以下简称“HT9G 钢”)。

真空感应熔炼 HT9G 钢锭, 经锻造和轧制加工成板材, 经 1 100 °C 正火 30 min + 720 °C 回火 90 min 后开展拉伸和持久寿命测试, 结果如图 14 和图 15 所示。从图 14 可以看出, HT9G 钢的室温屈服强度与抗拉强度显著高于 HT9、T91 和 T92 钢。其屈服强度可达 880 MPa, 比 T91 钢提高约 310 MPa, 比 HT9 和 T92 钢提高约 80~120 MPa。抗拉强度也高于上述三种核用 FM 钢, 达到 1 046 MPa。由图 15 可知, HT9

钢在 700 °C、100 MPa 条件下的持久断裂时间仅为 70~82 h, 而 HT9G 钢持久断裂时间延长到 372~385 h, 持久寿命得到明显提升, 显示出成分优化的有效性, 为后续长时持久强度测试奠定了基础。

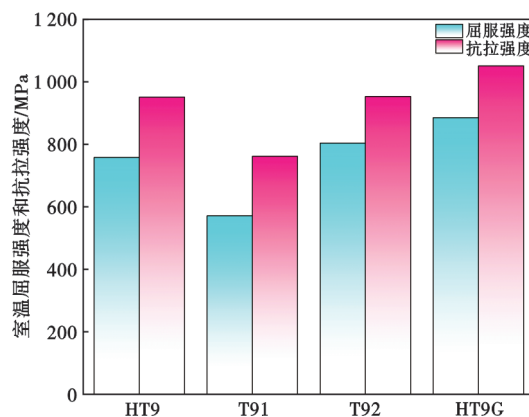


图 14 HT9G 合金室温拉伸性能及与 HT9、T91 和 T92 合金性能比对

Fig. 14 Comparison of room temperature tensile properties of HT9G alloy, HT9 alloy, T91 alloy, and T92 alloy

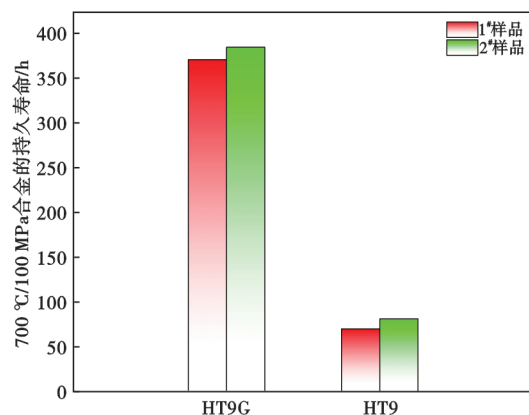


图 15 HT9G 合金 700 °C/100 MPa 下的持久寿命及与 HT9 合金比对, 1*样品和 2*样品为同种合金的平行试样。

Fig. 15 Creep rupture life of HT9G and HT9 alloys at 700 °C/100 MPa, Samples 1* and 2* are parallel specimens of the same alloy

4 结论

针对一体化快堆堆芯组件的材料性能要求, 综述了核电体系内铁马钢牌号的发展背景以及力学性能水平。通过对铁马钢中可添加合金元素的系统分析, 提出了在 HT9 合金的基础上, 通过添加 Nb 相形元素, 调节 W/Mo 比, 调控适宜 N、B 含量, 添加部分稀土元素等手段, 初步优化出一种用于一体化快堆堆芯组件的改性 HT9 合金。该合金在室温拉伸性能测试和 700 °C/100 MPa 持久寿命测试中, 表现出了

远高于HT9合金的性能水平,通过后续的迭代优化,

预计有望满足一体化快堆包壳材料服役指标要求。

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