文章编号:0253-4339(2024) 03-0023-15
doi:10.3969/j.issn.0253-4339.2024.03.023

新型低 GWP 制冷剂在-100~200 ℃的应用研究进展

张 迪　郭智恺　于万金　卢 朋　刘瑶瑶　刘武灿　张建君

(浙江省化工研究院有限公司 含氟温室气体替代及控制处理国家重点实验室　杭州　310023)

摘　要　制冷剂是蒸气压缩循环中必不可少的工作流体,氢氟烷烃类化合物是目前制冷、空调和热泵设备中的主要制冷剂,但该类制冷剂会引起全球变暖等问题,因此亟需开发应用性能相似、环境性能友好的新型制冷剂。介绍了不同制冷剂在-100~200 ℃蒸气压缩循环应用场景中的使用现状,并综述了具有前景的低 GWP 制冷剂替代方案研究进展;在此基础上,对 HFO-1234yf、HFO-1234ze(E)、HFO-1234ze(Z)、HCFO-1233zd(E)、HFO-1336mzz(Z) 5 种低 GWP 制冷剂在实际应用系统中的最新研究及关键问题进行总结。

关键词　制冷剂;空调;热泵;有机朗肯循环;GWP

中图分类号:TB61$^+$2;TB64　　**文献标识码**: A

Research Progress on the Application of Novel Low GWP Refrigerants in the Temperature Range from −100 ℃ to 200 ℃

Zhang Di　Guo Zhikai　Yu Wanjin　Lu Peng　Liu Yaoyao　Liu Wucan　Zhang Jianjun

(State Key Laboratory of Fluorinated Greenhouse Gases Replacement and Control Treatment, Zhejiang Research Institute of Chemical Industry Co., Ltd., Hangzhou, 310023, China)

Abstract　A refrigerant is an essential working fluid in a vapor-compression refrigeration cycle. Hydrofluorocarbons are currently used as refrigerants in refrigeration, air-conditioning, and heat pump systems and are potent greenhouse gases. Thus, replacement refrigerants with high thermal performances and low environmental impacts must be identified. This paper presents a comprehensive review of the refrigerants in different vapor-compression cycle applications covering the temperature range from −100 ℃ to 200 ℃. The most promising low-GWP alternative is proposed. Moreover, five refrigerants, including HFO-1234yf, HFO-1234ze(E), HFO-1234ze(Z), HCFO-1233zd(E), and HFO-1336mzz(Z), for application in vapor compression equipment, are summarized.

Keywords　refrigerant; air conditioning; heat pump; organic Rankine cycle; GWP

气候变化已成为全球关注的焦点,采暖、通风、空调和制冷行业是导致全球变暖的主要原因之一[1]。制冷剂作为制冷、空调、热泵等设备的工作流体,因设备充注、维修、事故损坏等原因被大量聚集在大气环境中,导致温室效应,引起环境的破坏。

制冷剂大致分为卤代烃、碳氢化合物、有机化合物和无机化合物。其中,卤代烃是指除了碳、氢元素外,还含有氟和/或氯元素的制冷剂。按其发展历程可分为四代:第一代为氯氟烷烃(CFCs),第二代为氢氯氟烷烃(HCFCs),第三代为氢氟烷烃(HFCs),第四代为氢氟烯烃(HFOs)、氢氯氟烯烃(HCFOs)。CFCs 和 HCFCs 由于含有氯原子,具有较高的臭氧损耗潜值(ozone depletion potential, ODP),已在全球范围内逐步停产淘汰。HFCs 的 ODP 为 0,是目前应用最为广泛的第三代制冷剂,但其大气寿命长、温室效应强,受到国际社会的密切关注。欧洲议会和理事会(EU) No 517/2014 号法规基于 100 年全球变暖潜值(Global Warming Potential, GWP),禁止在最广泛使用的制冷、空调和热泵的设备中使用 HFCs,禁止日期则根据更环保的替代品可获得性确定[2]。《〈关于消耗臭氧层物质的蒙特利尔议定书〉基加利修正案》已于 2019 年 1 月 1 日生效,中国于 2021 年 9 月 15 日正式宣布加入,至 2024 年开始对 HFCs 的生产和消费进行冻结,2045 年将削减 80%[3]。零 ODP 和低 GWP 成为第四代制冷剂的重要发展趋势与指标。

为寻找适合制冷系统的环境友好型替代品,M. O. Mclinden 等[4-7]对 56 000 个小分子进行筛选研究,并通过应用进化算法确定了 1 200 种低 GWP 制冷剂。以 GWP 低于 200、不可燃或低可燃性、化学性质稳定、无毒、合适的临界温度作为筛选参数,进一步

收稿日期:2023-03-24;修回日期:2023-06-30; 录用日期:2023-08-03

筛选出 62 种候选制冷剂,也可以考虑用于有机朗肯循环(organic Rankine cycle,ORC)[8]。在不同应用场景和设备工况中,制冷剂的使用性能均是评估候选制冷剂是否为合适的 HFCs 替代品的关键指标[9]。在超低温冷冻干燥、低温制冷、空调、热泵、高温热泵、ORC 等领域,由于所需制冷或制热的温度不同,选用制冷剂也不尽相同。本文将制冷剂应用场景根据所需温度进行划分如图 1 所示,分别总结了不同场景下制冷剂的研究进展,列举了目前正在使用的制冷剂及替代方案。此外,还针对 2,3,3,3-四氟丙烯(HFO-1234yf)、反式-1,3,3,3-四氟丙烯(HFO-1234ze(E))、顺式-1,3,3,3-四氟丙烯(HFO-1234ze(Z))、反式-1-氯-3,3,3-三氟丙烯(HCFO-1233zd(E))、顺式-1,1,1,4,4,4-六氟-2-丁烯(HFO-1336mzz(Z))5 种新型低 GWP 制冷剂在相关应用领域替代技术的研究进展进行总结。

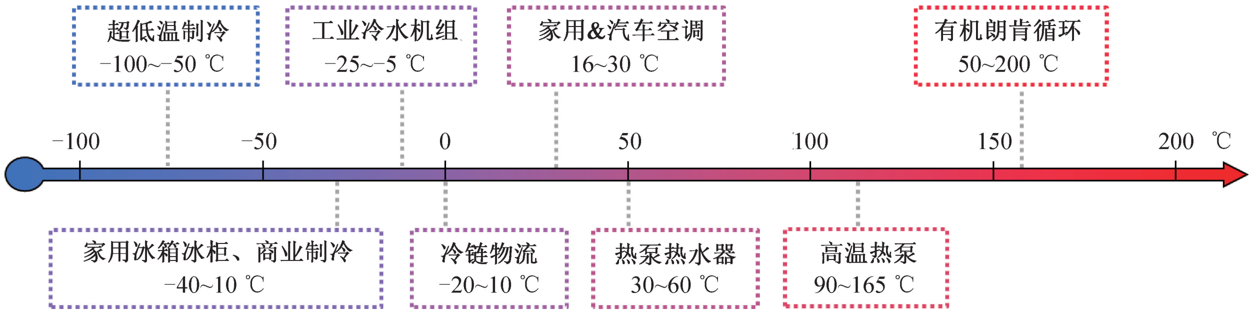

图 1 制冷剂应用场景及使用温度范围

Fig.1 Applications and indicative temperature range of the refrigerant

1 应用场景及制冷剂方案

1.1 超低温制冷

温度低于-50 ℃的设备可被定义为超低温制冷,其蒸发温度范围可达-100 ℃,广泛应用于生物医药、食品加工、国防军事、现代工业(特殊合金钢的沉淀硬化、液体储存、石油蒸气液化)、科学研究(挥发性有机物的冷却)等领域[10-11]。与其他应用相比,超低温制冷运行的设备数量较少,目前商业化的制冷剂为三氟甲烷(HFC-23)和 R508B,理化性质如表 1 所示[12-13]。

表 1 超低温制冷用制冷剂的应用性能[11-15]

Tab.1 Properties of refrigerants for ultra-low temperature refrigeration[11-15]

制冷剂	组成(质量分数/%)	ODP	GWP_{100}	摩尔质量/(g/mol)	临界温度/℃	临界压力/MPa	汽化潜热/(kJ/kg)*
HFC-23	三氟甲烷	0	12 400	70. 01	26. 14	4. 83	237. 71
R508B	PFC-116/HFC-23(54. 0/46. 0)	0	13 396	95. 39	11. 20	3. 77	160. 36
HC-170	乙烷	0	6	30. 07	32. 17	4. 87	444. 77
HC-1150	乙烯	0	4	28. 05	9. 25	5. 04	443. 18
HFO-1132a	1,1-二氟乙烯	0	<1	64. 00	29. 66	4. 46	—
HFC-41	一氟甲烷	0	116	34. 03	44. 13	5. 90	490. 19

注:* 在蒸发温度为-80 ℃进行评估。

由于法规(EU) No 517/2014 未对运行在-50 ℃以下的制冷系统所使用的制冷剂限定 GWP,相关替代品研究较少。A. Mota-Babiloni 等[14]综述了近年来低温冷冻系统和工质的研究进展,认为最具有应用前景的低 GWP、超低温制冷替代工质包括乙烷(HC-170)、乙烯(HC-1150)、1,1-二氟乙烯(HFO-1132a)、一氟甲烷(HFC-41),但仍存在一定局限性,还需通过进一步实验研究来完善替代方案。Sun Zhili 等[11,15]研究了三级级联制冷剂系统(蒸发温度低于-80 ℃)所用制冷剂,使用 HC-1150 替代四氟化碳(PFC-14),用 HFC-41、HC-170 替代 HFC-23。

1.2 家用/商用低温制冷

1.2.1 家用冰箱冰柜

异丁烷(HC-600a)和 1,1,1,2-四氟乙烷(HFC-134a)是目前主流的家用冰箱制冷剂。HC-600a 具有低 GWP、总体能效较高的优势,已被证明是一种可

靠、高效的选择。日本预测在 2030 年使用 HC-600a 替代 HFC-134a,可减少 72 kt CO_2 当量引起的全球变暖影响[16]。出于对易燃性的考虑,一些国家和地区对碳氢化合物(HCs)的充注量进行限制,安全法规严格的国家更倾向于使用不易燃的制冷剂来取代 HFC-134a。J. M. Belman-Flores 等[17]综述了 1,1-二氟乙烷(HFC-152a)、HFO-1234yf 及几种 HCs 单工质或混合物作为 HFC-134a 在家用制冷中的替代品,但强调缺乏关于新合成替代品的研究。GWP 小于 150 的限制减少了家用冰箱冰柜中替换 HFC-134a 的选项,目前研究主要集中在新氟化物的选择。已有研究显示,HFO-1234yf[18-20]、HFO-1234ze(E)[21-22]等制冷剂应用于家用冰箱时性能较好,可替代 HFC-134a。综上所述,HC-600a 应用于家用冰箱的技术已较成熟,且由于家用冰箱全封闭的系统结构以及较低的充注量,其他工质替代 HC-600a 的概率较小。

1.2.2 商业制冷

目前商业制冷应用最广泛的制冷剂是 R404A、HFC-134a 和二氟一氯甲烷(HCFC-22)。其中独立单元制冷剂和冷凝机组制冷剂以 R404A、HFC-134a 为主,发达国家逐步用丙烷(HC-290)、二氧化碳(R744)作为替代品。对于超市集中系统,HCFC-22 仍是主要选择。由于超市内部不能使用易燃或有毒的制冷剂,通过提高制冷效率(如采用跨临界和级联系统),R744 逐渐占据较大的市场份额。根据调查显示,欧洲目前安装或使用的大多数商业制冷系统以 HFCs 和天然制冷剂为基础,使用最多的合成制冷剂包括 HFC-134a、R404A、R407 系列(HFC-32/HFC-125/HFC-134a)、R410A 和 R507A,天然制冷剂则是 R744、氨(R717)和 HC-290[23]。由于纯 HFOs 制冷剂与 R404A 和 R507A 存在显著的热力学差异,并非是理想替代品,因此考虑使用 HFCs/HFOs 混配制冷剂,如 R448A、R449A、R452A、R455A 等[24-26]。混配制冷剂的组成及基础物性如表 2 所示。受 GWP 限制,欧洲的制冷系统中仅允许使用 R455A。随着国际环保要求越来越严格,更多环境性能优异的混配制冷剂正在开发并逐步推出中。

表 2 低温制冷用制冷剂的应用性能[23-26,29]

Tab.2 Properties of refrigerants for low temperature refrigeration[23-26,29]

制冷剂	组成(质量分数/%)	ODP	GWP_{100}	摩尔质量/(g/mol)	临界温度/ ℃	临界压力/MPa	汽化潜热/(kJ/kg)*
HC-600a	异丁烷	0	20	58. 12	134. 66	3. 63	372. 31
HC-290	丙烷	0	20	44. 10	96. 74	4. 25	400. 77
R744	二氧化碳	0	1	44. 01	30. 98	7. 38	282. 44
R717	氨	0	0	17. 03	132. 25	11. 33	1 328. 80
HCFC-22	二氟一氯甲烷	0. 03	1 760	86. 47	96. 15	4. 99	220. 02
HFC-134a	1,1,1,2-四氟乙烷	0	1 300	102. 03	101. 06	4. 06	212. 92
HFC-152a	1,1-二氟乙烷	0	138	66. 05	113. 26	4. 52	375. 25
HFO-1234yf	2,3,3,3-四氟丙烯	0	<1	114. 04	94. 70	3. 38	175. 18
HFO-1234ze(E)	反式-1,3,3,3-四氟丙烯	0	<1	114. 04	109. 36	3. 63	196. 21
R404A	HFC-125/HFC-134a/HFC-143a(44. 0/4. 0/52. 0)	0	3 800	97. 60	72. 12	3. 73	182. 18
R407C	HFC-32/HFC-125/HFC-134a(23. 0/25. 0/52. 0)	0	1 378	86. 20	86. 20	4. 63	226. 77
R410A	HFC-32/HFC-125(50. 0/50. 0)	0	2 088	72. 59	71. 34	4. 90	243. 66
R507A	HFC-125/HFC-143a(50. 0/50. 0)	0	4 000	98. 86	70. 61	3. 70	178. 41
R448A	HFC-32/HFC-125/HFO-1234yf/HFC-134a/HFO-1234ze(E)(26. 0/26. 0/20. 0/21. 0/7. 0)	0	1 387	86. 28	82. 68	4. 59	218. 15
R449A	HFC-32/HFC-125/HFO-1234yf/HFC-134a(24. 3/24. 7/25. 3/25. 7)	0	1 282	87. 21	82. 07	4. 50	216. 02
R452A	HFC-32/HFC-125/HFO-1234yf(11. 0/59. 0/30. 0)	0	1 945	103. 51	75. 05	4. 01	176. 10
R455A	R744/HFC-32/HFO-1234yf(3. 0/21. 5/75. 5)	0	146	87. 45	85. 61	4. 65	209. 07

注:* 在蒸发温度为-20 ℃进行评估。

1.2.3 冷链物流

为保障冷藏冷冻类物品的品质,通常要求从生产、贮藏运输、销售到消费前的各环节始终处于规定的低温环境下[27]。目前在运输制冷中,综合考虑制冷能效及安全性问题,低温、中温冷藏车使用的制冷剂分别以 R404A、HFC-134a 为主[28]。未来 R448A、R449A、R452A 等 HFCs/HFOs 混配制冷剂有可能替代 R404A 应用于冷藏运输,但仍存在 GWP 较高的问题[29]。Gao Enyuan 等[30]综述了我国冷链关键设备制冷剂的应用现状与替代技术,认为 R404A 和 R507A 的替代是未来需要关注和解决的突出问题。

1.3 工业冷水机组

大多数大型工业系统使用 R717 作为制冷剂。当 R717 不可接受时,替代方案包括间接系统中的 R744、乙二醇,直接系统中的 HCFCs 或 HFCs[31]。离心式冷水机的选择包括 HCFC-22、HCFC-123 和 HFC-134a,其中 HCFC-123 具有明显的效率优势,约 2/3 的新装置使用 HCFC-123,其余大部分使用 HFC-134a[32]。HFOs 制冷剂 HFO-1234yf、HFO-1234ze(E)及其混配制冷剂(如 R513A、R513B、R515A、R515B、R516A,基础物性如表 3 所示)可替代 HFC-134a 在系统中使用[33-35]。R515A 对环境影响较小,性能系数(coefficient of performance, COP)与 HFC-134a 相近,是潜在的理想替代工质[33,36]。

表 3 用于工业冷水机组的 HFOs 混配制冷剂应用性能[33]

Tab.3 Properties of HFOs blends for chiller applications[33]

混配制冷剂	组成(质量分数/%)	ODP	GWP_{100}	摩尔质量/(g/mol)	临界温度/ ℃	临界压力/MPa	汽化潜热/(kJ/kg)*
R513A	HFO-1234yf/HFC-134a(56.0/44.0)	0	573	108.43	94.91	3.65	173.75
R513B	HFO-1234yf/HFC-134a(58.5/41.5)	0	540	108.73	94.78	3.63	172.91
R515A	HFO-1234ze(E)/HFC-227ea(88.0/12.0)	0	403	118.73	108.71	3.57	175.16
R515B	HFO-1234ze(E)/HFC-227ea(91.1/8.9)	0	299	117.48	108.88	3.58	177.00
R516A	HFO-1234yf/HFC-134a/HFC-152a(77.5/8.5/14.0)	0	131	102.58	96.64	3.62	181.68

注:* 在蒸发温度为 3 ℃进行评估。

1.4 空调

1.4.1 家用空调

目前 R410A 广泛应用于空调系统,是空调制冷剂 HCFC-22 的主要替代品,常见的制冷剂还包括 HC-290、二氟甲烷(HFC-32)。在低 GWP 制冷剂替代方面,单组分制冷剂 HFO-1234yf、HFC-152a 由于单位容积制冷量小,可能无法广泛用于替代 HCFC-22,因此主要考虑 HFCs/HFOs 等混配制冷剂。已开发的 R446A、R447A、R452B、R454B、R463A、R466A 混配制冷剂具有与 HCFC-22 或 R410A 相似的操作压力和使用性能,基础物性如表 4 所示[37-40]。上述混配制冷剂中大部分尚未被大规模商业应用,也无足够的技术数据支持其进入公共领域。目前认为 R454B 混配制冷剂可能具有较大的实际应用潜力,空调制造商正在对其性能进行逐步深入研究。

1.4.2 汽车空调

现代汽车和其他小型车辆的汽车空调(mobile air-conditioning, MAC)系统通常使用 HFC-134a 作为制冷剂,在 2006 年欧盟发布 MAC 指令(禁止使用 GWP 超过 150 的制冷剂)和美国环保局要求减少制冷剂相关温室气体排放的影响下,近年来在开发 GWP 低于 150 的新型替代品方面开展大量研究。对于 MAC 中 HFC-134a 的替代方案有 HFO-1234yf、R444A、R445A 等混配制冷剂[41-42]。M. Mohanraj 等[43]综述了汽车空调用环保型制冷剂的研究进展,总结了 HFO-1234yf 作为 HFC-134a 替代物的性能,认为 HFO-1234yf、HFC-152a、HC-290/HC-600a、R430A、R436A、R444A、R445A、R450A 和 R513A 的沸点与 HFC-134a 相似,适用于汽车空调制冷。虽然替代工质可直接应用于原 HFC-134a 机组中,但在实际应用前仍需对汽车空调系统进行优化,以满足热负荷要求[44]。

1.5 热泵

1.5.1 电动汽车热泵空调

新能源汽车依靠辅助电加热或热泵等方式实现制热。热泵空调具有制冷和制热的双重功能,目前最常见的工质为 HFC-134a、R744,其他包括 HC-290、HC-600a,低 GWP 的 HFC-152a、HFC-161、HFO-

表 4 用于空调系统的混配制冷剂应用性能[39-43]

Tab.4 Properties of mixed refrigerants for air-conditioning system[39-43]

混配制冷剂	组成(质量分数/%)	ODP	GWP_{100}	摩尔质量/(kg/mol)	临界温度/ ℃	临界压力/MPa	汽化潜热/(kJ/kg)*
R446A	HFC-32/HFO-1234ze(E)/HC-600(68.0/29.0/3.0)	0	490	62.00	85.96	5.73	266.01
R447A	HFC-32/HFC-125/HFO-1234ze(E)(68.0/3.5/28.5)	0	580	63.05	85.30	5.22	261.47
R452B	HFC-32/HFC-125/HFO-1234yf(67.0/7.0/26.0)	0	695	63.53	77.10	5.29	249.95
R454B	HFC-32/HFO-1234yf(68.9/31.1)	0	465	62.61	78.10	5.27	254.23
R463A	HFC-32/HFC-125/HFC-134a/HFO-1234yf/R744 (36.0/30.0/14.0/14.0/6.0)	0	1 494	74.72	75.63	5.24	211.15
R466A	HFC-32/HFC-125/CF_3I(49.0/11.5/39.5)	0	733	80.69	73.13	5.28	189.23
R430A	HFC-152a/HC-600a(76.0/24.0)	0	107	63.96	107.02	4.09	294.86
R436A	HC-290/HC-600a(50.0/50.0)	0	<20	49.33	115.89	4.27	362.59
R444A	HFC-32/HFC-152a/HFO-1234ze(E)(12.0/5.0/83.0)	0	92	96.70	106.36	4.47	198.86
R445A	R744/HFC-134a/HFO-1234ze(6.0/9.0/85.0)	0	130	103.11	106.04	4.54	187.82
R450A	HFC-134a/HFO-1234ze(42.0/58.0)	0	547	108.67	104.5	3.82	185.27
R513A	HFC-134a/HFO-1234yf(44.0/56.0)	0	573	108.43	94.91	3.65	172.30

注:* 在蒸发温度为 5 ℃进行评估。

1234yf 及其混配物等[45-47]。R744 在低温加热方面具有显著优势,但在高温环境下制冷效果不佳,当环境温度高于 35 ℃时,CO_2 跨临界系统的冷却性能弱于 HFC-134a 系统[48]。HFO-1234yf 作为替代 HFC-134a 的新一代工质,与 HFC-134a 相比热力学和迁移性质相似或更差,因此在应用之前还需进一步研究[49]。

1.5.2 热泵热水器

研究显示,新一代热泵热水器的整体效率高于传统电热水器,是市场上大量水加热设备的选择之一[50]。常用制冷剂有 HCFC-22、R410A、HFC-134a、R407C、HC-290 等[51]。在新设备中,中小型系统使用 R410A 最具成本效益,而大型系统使用 HFC-134a 最有效。可选的新制冷剂方案包括 R717、R744、HC-290、R1270、HFO-1234ze(E)、HCFO-1233zd(E)、HFO-1336mzz(Z)、HFC-32、R444B、R446A、R447A 和 R450A 等。

1.5.3 高温热泵

高温热泵的"高温"是相对于供水温度在 55 ℃以下的热泵热水器而言。目前市场上的高温热泵产品可提供 90~165 ℃的供暖温度,用于干燥、蒸汽产生、巴氏杀菌、余热回收等工业过程[52]。HFC-245fa 是应用于高温热泵的主要制冷剂,替代品有 HFO-1336mzz(Z)、HFO-1234ze(Z)、HCFO-1233zd(E)、HCFO-1224yd(Z)、R514A(基础物性如表 5 所示),以及碳氢化合物正戊烷(HC-601)和正丁烷(HC-600)[53-54]。Wu Di 等[55]综述了使用低 GWP 制冷剂的蒸气压缩热泵研究现状,并提出应用指南,其中 HC-600、HC-600a、HFO-1234ze(Z)是热源温度为 40~80 ℃、输出温度为 80~120 ℃的高温热泵应用中最合适的制冷剂,HC-601、HFO-1336mzz(Z)、HCFO-1233zd(E)、HCFO-1224yd(Z)可用于热源温度为 60~100 ℃、输出温度为 100~160 ℃的超高温热泵。

1.6 有机朗肯循环

ORC 通过与热源温度相匹配的有机工质相变做功,将低温热能转化为高品位电能[56]。目前 ORC 应用最广泛的制冷剂是 HFC-134a 和 HFC-245fa,HFC-134a 常用于地热发电厂和中低温热利用,HFC-245fa 主要应用于中高热源的余热回收[57]。Chen Huijuan 等[58]综述了 ORC 和超临界朗肯循环相关研究,提出了工质的选择标准。对于特定的应用,Bao Junjiang 等[59-60]总结了不同温度范围内 CFCs、HCFCs、HFCs、碳氢化合物、醇类化合物、水等工质的选择方案,其中 HFC-134a、HCFC-141b、HFC-245fa 分别适用于热源温度为 50~100 ℃、50~150 ℃、50~200 ℃以内的应用,HCFO-1233zd(E)适用于热源温度为 100~110 ℃的应用。朱杰人等[61]根据地热能、太阳能、工业余热及生物质能 4 种热源种类,推荐了相应热源温度下的 ORC 工质。

表 5 用于高温热泵的新型制冷剂应用性能[53-54]

Tab.5 Properties of new refrigerants for high temperature heat pump[53-54]

制冷剂	组成(质量分数/%)	ODP	GWP_{100}	沸点/℃	摩尔质量/(kg/mol)	临界温度/℃	临界压力/MPa	汽化潜热/(kJ/kg)*
HFO-1234ze(Z)	顺式-1,3,3,3-四氟乙烯	0	<1	9.75	114.04	150.12	3.53	165.08
HCFO-1233zd(E)	反式-1-氯-3,3,3-三氟丙烯	0.000 34	1	18.26	130.50	166.45	3.62	157.87
HCFO-1224yd(Z)	顺式-1-氯-2,3,3,3-四氟丙烯	0.000 12	<1	14.62	148.49	155.54	3.38	132.36
HFO-1336mzz(Z)	顺式-1,1,1,4,4,4-六氟-2-丁烯	0	2	33.45	164.06	171.35	2.90	140.05
R514A	HFO-1336mzz(Z)/反式-1,2-二氯乙烯(74.7/25.3)	0	2	—	139.60	178.00	3.52	—

注:* 在蒸发温度为 80 ℃进行评估。

2 HFOs、HCFOs 制冷剂在实际系统中的应用

2.1 HFO-1234yf

HFO-1234yf 的 ODP 为 0,GWP 小于 1,弱可燃,具有制冷能效高、安全性高的优点,其热物理性质与 HFC-134a 相似,是最接近商业化的第四代制冷剂,被广泛认为是汽车应用中 HFC-134a 的替代品,或作为热泵应用中非共沸混合物的组元。

早期研究表明,在 MAC 系统中使用 HFO-1234yf,充注量相比 HFC-134a 减少 10%,但 COP 和制冷量均低于 HFC-134a[62-63]。之后的研究以 HFC-134a 作为基准,从理论上对比了 HC-290、HC-600a、R407C、R410A、R404A、HFC-152a、HFO-1234ze(E)、HFO-1234yf 等不同制冷剂,得出 HFO-1234yf 是移动空调中 HFC-134a 最重要的替代方案,从 HFC-134a 向 HFO-1234yf 过渡无需对 MAC 系统进行大量修改[64-66]。但由于实际存在 HFO-1234yf 的使用性能相比 HFC-134a 有所下降的问题,C. Zilio 等[67]对 HFO-1234yf 的替代技术进行了开创性研究,提出对硬件进行适当修改(如增加冷凝器、蒸发器面积,优化可变排量压缩机控制阀),使 HFO-1234yf 的 COP 高于基准 HFC-134a。关于 MAC 系统及组件优化方面的研究主要包括安装回热器(internal heat exchanger,IHX),优化喷射器、膨胀机、转动膨胀阀等,以提升 HFO-1234yf 替换后的使用性能[68]。

H. Cho 等[69-70]提出安装 IHX 可缩小 HFO-1234yf 与 HFC-134a 的性能差异,压缩机转速为 800~1 800 r/min 时,HFO-1234yf 的 COP 相比 HFC-134a 低 0.3%~2.9%,转速为 2 500 r/min 时,HFO-1234yf 的 COP 相比 HFC-134a 高 0.9%。J. Navarro-Esbri 等[71-73]同样发现使用 IHX 可减少 HFO-1234yf 与 HFC-134a 两种工质的 COP 差异。G. Pottker 等[74]发现冷凝器过冷可使系统 COP 最大提升 18%,使带有 IHX 的系统 COP 提高 9%。D. Mehmet 等[75-76]研究了采用 IHX 的系统在不同压缩机转速、气流温度、冷凝温度和蒸发温度影响下的性能,发现安装 IHX 使 HFO-1234yf 系统 COP 提高 4%~6%,㶲损失降低 13%~16%。Qi Zhaogang[77-78]研究了过热、过冷和压缩机性能等方面的改进潜力,并通过微通道平行流蒸发器使 HFO-1234yf 的制冷量相比 HFC-134a 提高 6.5%。Li Huashan 等[79]通过喷射器补偿标准蒸气压缩制冷循环中的节流损失,发现使用 HFO-1234yf 的喷射器-膨胀制冷循环性能优于标准制冷循环,尤其是在极端工作条件下。F. Molés 等[80]通过使用膨胀机或喷射器,使 HFO-1234yf、HFO-1234ze(E)的系统 COP 相比 HFC-134a 常规制冷循环分别提高 9%~15%、11%~20%。Shi Junye 等[81]通过调节优化膨胀阀,使 HFO-1234yf 的制冷量和 COP 平均提高 11.3%、8%。R. Prabakaran 等[82]结合 IHX 和膨胀机对 MAC 系统性能提升的优势,优化了具有吸入式热交换器的 HFO-1234yf 系统,使系统 COP、制冷量和㶲效率分别比 HFC-134a 系统提高 4.3%~8.6%、6.5%~10.1%和 3.7%~5.1%。

随着电动汽车的快速发展,迫切需要 MAC 系统实现高效率、低能耗的设计与创新。热泵空调可实现电动汽车乘客舱的冷却和加热,已成为替代原有空调制冷+PTC 制热系统的发展趋势之一[83-85]。研究人员通过对比汽车热泵(automotive heat pump,AHP)系统中 HFO-1234yf 和 HFC-134a 的加热和冷却性能参数以及㶲效率,认为 HFO-1234yf 可在 AHP 系统中使用。但与 HFC-134a 相比,加热和冷却性能以及 COP 均降低,系统㶲损失提高[86-88]。Li Wanyong 等[89-90]对 HFO-1234yf 汽车热泵在寒冷气候条件下的性能进行测试,采用强化补气(enhanced vapor injection,EVI)技术可使 HFO-1234yf 在-10 ℃和-20 ℃条件下的加热性能平均提高 27.4%、19.9%,另外通过增大内部冷凝器面积、增大风量、提升压缩机转速等,也能实现 AHP 性能与能效的提升。

2.2 HFO-1234ze(E)

HFO-1234ze(E)的 ODP 为 0,GWP 小于 1,具有比 HFO-1234yf 更低的可燃性。HFO-1234ze(E)的制冷量远低于 HFC-134a,无法在 HFC-134a 系统中直接替代,需要对压缩机、热交换器、膨胀装置等系统部件进行优化,以提升 HFO-1234ze(E)的制冷性能。

在家用制冷领域,K. M. Karber 等[91]发现在两类被测试冰箱中 HFO-1234ze(E)的能耗均低于 HFC-134a,但制冷量偏低,导致压缩机运行时间增加,除霜周期间隔缩短,因此需要在系统优化后使用。C. Aprea 等[18,92]研究了家用冰箱中 HFO-1234yf、HFO-1234ze(E)取代 HFC-134a 的可能性,结果表明,HFO-1234ze(E)的制冷循环性能与 HFC-134a 相似,系统 24 h 的能耗降低 9%,认为 HFO-1234ze(E)相比 HFO-1234yf 更适合用于冰箱制冷剂的替代。

在商业制冷方面,A. Sethi 等[93]估计在自动售货机中 HFO-1234ze(E)的制冷量比 HFC-134a 约低 25%,通过增大压缩机排量、吸气管直径以及增加热交换器内制冷剂的循环次数可提升 HFO-1234ze(E)的制冷性能。A. E. Kabeel 等[94]利用商业蒸气压缩循环系统进行冷库制冷,结果表明,HFO-1234ze(E)比 HFC-134a 的制冷量低 2%～13%,功耗低 9%～15%,HFO-1234ze(E)、HFC-134a 可达到的最低蒸发温度分别为-13 ℃、-30 ℃。

Z. Janković 等[95]研究了 HFO-1234ze(E)、HFO-1234yf 在小容量制冷机组中的性能,发现 HFO-1234ze(E)的制冷量比 HFC-134a 低约 25%,但 COP 高 4%～7%。A. Mota-Babiloni 等[73,80]对比研究了 HFO-1234yf、HFO-1234ze(E)在 HFC-134a 蒸气压缩系统中的性能,与 HFC-134a 相比,HFO-1234yf、HFO-1234ze(E)的制冷量平均减少 9%、30%,COP 分别降低 3%～11%、2%～8%。IHX 的使用有利于提高两种替代制冷剂的 COP,而最有效的配置是使用膨胀器或喷射器作为膨胀装置。A. Mota-Babiloni 等[96]建议引入 IHX 和增大 43%的压缩机转速,以达到 HFO-1234ze(E)与 HFC-134a 在制冷性能上的匹配。

HFO-1234ze(E)在喷射式制冷系统[97-98]、蒸气吸收式制冷系统[99-100]、有机朗肯循环[101-102]、高温热泵[103-104]中的适用性也得到研究。为提高使用性能,需进一步开展工作系统的优化提升。另一方面,可通过在 HFO-1234ze(E)中混入其他制冷剂成分(如 HFC-32 等)有效改善热力学性能[105]。

2.3 HFO-1234ze(Z)

HFO-1234ze(Z)是 HFO-1234ze(E)的顺反式同分异构体,同样具有 ODP 为 0,GWP 低的环保特性。HFO-1234ze(Z)的临界温度为 150.12 ℃,因此能够在高温条件下进行亚临界循环,在制冷剂应用中表现出与 HFO-1234ze(E)(临界温度为 109.36 ℃)明显差异[106]。近年来,HFO-1234ze(Z)主要作为一种潜在的工作流体,在高温热泵、ORC 等领域被广泛研究。

J. S. Brown 等[107]最早评估了 HFO-1234ze(Z)的热力学和热物理性质,认为 HFO-1234ze(Z)是替代 CFC-114 用于高温热泵的潜在工质。S. Fukuda 等[103]分析了制冷剂热力学和循环性能,并结合实验装置主要部件(压缩机、冷凝器、膨胀阀和蒸发器)产生的不可逆损失,提出 HFO-1234ze(Z)适用于高温热泵系统,而非典型的空调或制冷系统。G. A. Longo 等[108-110]通过热力学评估,并考察制冷剂在高温热泵所用商用钎焊板式换热器内的饱和蒸气冷凝传热系数和摩擦压降,发现 HFO-1234ze(Z)在相同质量流量下传热系数高于 HFC-236fa、HFC-134a、HC-600a、HFO-1234ze(E)、HCFO-1233zd(E),相同工况下摩擦压降与 HC-600a 相似,比 HCFO-1233zd(E)低 7%～32%,得出 HFO-1234ze(Z)可作为高温热泵、ORC 制冷剂的结论。

设计先进的热力学循环方式是提高热泵系统效率的关键之一。单级压缩循环配置通常适用于温升低于 50 ℃的高温热泵系统,而对于温升为 60 ℃及以上的热泵系统,采用多级压缩循环对提升 COP 有显著帮助[53]。研究人员对使用 HFO-1234ze(Z)作为工质的不同类型压缩循环系统进行研究。Luo Baojun 等[111]研究了多种单级蒸气压缩循环的性能,发现在冷凝温度为 120 ℃、温升为 40 ℃时,使用 HFC-245fa、HC-600 和 HFO-1234ze(Z)的系统 COP 相近。Zhang Xuedong 等[112]测试了 CFC-114、HFC-134a、HFC-227ea、HFC-236fa、HFC-245fa 和 HFO-1234ze(Z)的蒸气压缩热泵系统循环性能,结果表明,在冷凝温度为 85 ℃、温升为 45 ℃时,HFO-1234ze(Z)的 COP 为 3.55,比 HFC-245fa 的 COP 最大值高 40%,认为 HFO-1234ze(Z)适用于单级蒸气压缩循环的中高温热泵体系。C. Kondou 等[104]对比了使用 R717、HFC-365mfc、HFO-1234ze(E)、HFO-1234ze(Z)的 4 种循环配置 COP,在冷凝温度为 160 ℃、温升为 80 ℃条件下,使用 HFO-1234ze(Z)的三

级分流热泵系统具有最高的 COP,HFO-1234ze(Z)和 HFC-365mfc 的级联循环也具有较高的 COP 及实际效益。Hu Bin 等[113]分析了采用 HFO-1234ze(Z)工质的多级压缩式高温热泵系统,基于余热回收供应出水温度为 120 ℃,两级、三级压缩循环的 COP 分别比单级循环提高 9.1%、14.6%。D. Mikielewicz 等[114]研究发现冷凝温度为 130 ℃、升温为 80 ℃时,HFC-365mfc/HFO-1234ze(Z)和 HC-601/HFO-1234ze(Z)组合的两级级联循环具有较高 COP,高于多数单级体系。Li Shuaiqi 等[115]提出一种以热力车间 20~40 ℃热空气为热源的串级热泵蒸汽系统(cascade air-source heat pump steam system,CASGHP),可直接产生供车间使用的蒸汽。所用制冷剂中,HFO-1234ze(Z)在中低温热源 CASGHP 系统中性能更好。C. Arpagaus 等[52]通过制冷剂热力学性质及两级循环性能对比分析,认为散热器温度比工质临界温度低约 30 ℃时,可获得最佳 COP;HFO-1234ze(Z)、HCFO-1233zd(E)、HCFO-1224yd(Z)在 COP 和容积制热量(volumetric heat capacity,VHC)方面更接近 HFC-245fa,可作为 HFC-245fa 的替代品。

2.4 HCFO-1233zd(E)

HCFO-1233zd(E)的 ODP 不为 0,但大气停留时间短,对臭氧层破坏几乎可忽略,且 GWP 为 1,无毒、不可燃,具有良好的应用前景。HCFO-1233zd(E)作为制冷剂的研究主要集中在 ORC 系统,用于替代 HFC-245fa。此外,HCFO-1233zd(E)还可作为高温热泵的潜在工质[116],以及动力电池直冷板制冷剂[117]。

A. Giuffrida 等[118-119]通过热力学模拟,研究了在微型 ORC 系统中使用含氟烷烃、含氟烯烃制冷剂时的性能差异及热损失,发现 HCFO-1233zd(E)、HFO-1234ze(Z)在所选微型 ORC 系统中的性能略高于 HFC-245fa,是潜在的替代工质。B. V. Datla 等[120]率先使用 75 kW 变速 ORC 系统进行实验研究,结果表明,HCFO-1233zd(E)在同等容量下比 HFC-245fa 的热效率提高 8.7%。F. Molés 等[121]通过商用 ORC 实验台测试发现,HCFO-1233zd(E)的热功率和电功率比 HFC-245fa 低,但可在更高的蒸发温度和更低的过热温度下工作。S. Eyerer 等[122]在 1 kW 的 ORC 实验台研究发现,HCFO-1233zd(E)具有与 HFC-245fa 相似的冷凝传热性能和略低的蒸发传热性能,热效率提高 6.9%,但总输出功率降低 12.2%。L. Guillaume 等[123]在 3.5 kW 的小型 ORC 实验台中对比了 HFC-245fa 与 HCFO-1233zd(E)的性能,结果表明,HCFO-1233zd(E)操作系统的总功率输出较低,但净功率产量相似,在回收长途卡车尾气余热方面具有更好的潜力。

Yang Jingye 等[124]发现 HCFO-1233zd(E)的最大循环热效率比 HFC-245fa 提高约 3.8%,产生的电力比 HFC-245fa 多 4.5%,并提出了使用 HCFO-1233zd(E)的热交换器与膨胀机匹配优化方案。通过实验及综合分析对比,Yang Jingye 等[125]认为 HCFO-1233zd(E)、HFO-1234ze(Z)两者与 HFC-245fa 的循环性能相似,可作为 HFC-245fa 的替代品应用于微型 ORC 系统,而 HFO-1336mzz(E)压力较小,循环效率明显偏低。S. Eyerer 等[126]认为工质饱和蒸气压、蒸发焓、蒸气密度和黏度的差异是造成 ORC 运行参数和系统性能差异的主要原因,并在 ORC 实验台对比了 HFC-245fa、HCFO-1233zd(E)、HCFO-1224yd(Z)的性能差异,结果表明,HFC-245fa 最大输出功率比 HCFO-1233zd(E)高 9%,比 HCFO-1224yd(Z)高 12%;在热效率方面 HFC-245fa 比 HCFO-1233zd(E)低约 2%,与 HCFO-1224yd(Z)相似。

针对 ORC 系统中换热器及传热性能的研究,M. Welzl 等[127]研究了蒸发器核态沸腾传热系数与电力输出的关系,结果表明,在等转数、蒸发器热功率恒定的条件下 HFC-245fa 与 HCFO-1233zd(E)的输出功率相近。D. C. Lee 等[128]研究了钎焊板式换热器(水力直径 D_h=3.9 mm)内工质的蒸发传热特性和压降,在典型的 ORC 应用中,HCFO-1233zd(E)沸腾传热系数和压降与 HFC-245fa 相当。Zhang Ji 等[129]研究了 HFC-134a、HFC-245fa、HFO-1234ze(E)、HCFO-1233zd(E)在板式换热器(水力直径 D_h=3.4 mm)内部的冷凝传热和压降性能,结果表明,冷凝温度为 30~70 ℃、质量流量为 16~90 kg/(m^2·s)时,HCFO-1233zd(E)的传热系数和压降高于 HFC-245fa。

2.5 HFO-1336mzz(Z)

HFO-1336mzz(Z)的 ODP 为 0,GWP 为 2,不易燃,在 HFOs 制冷剂中具有最高的临界温度 171.3 ℃,在 250 ℃高温条件下表现出显著的化学稳定性,被认为是高温热泵、ORC 等应用中潜在的制冷剂[130]。

K. Kontomaris 等[131]在未优化的 ORC 原型系统中对 HFO-1336mzz(Z)进行实验研究,该系统基于容量约为 12 kW 的往复式膨胀机,膨胀机入口温度为 196 ℃,冷凝温度为 60、80 ℃,测得最大净热效率约为 10%。J. Navarro-Esbrí 等[132]在 1.5 kW 低温应用 ORC 实验台上对 HFO-1336mzz(Z)进行实验评估,热源温

度为 140～160 ℃,散热器温度为 25～40 ℃时,HFO-1336mzz(Z)相比 HFC-245fa 和 HCFO-1233zd(E)可获得更高等熵效率,HFO-1336mzz(Z)的净电效率也高于 HFC-245fa。M. Welzl 等[133]在 1 kW 的 ORC 实验台上对比了工质的机械等熵效率,结果表明,膨胀器内部存在较高的泄漏,HFO-1336mzz(Z)的机械等熵效率比 HFC-245fa 低 0.9%～6.2%。

3 总结

为防止全球变暖加速导致极端气候变化,各国政府正不断将气候行动纳入经济发展计划中。基于 HFCs 制冷剂的使用限制及淘汰计划,寻找不同应用场景和设备工况中合适的制冷剂替代方案迫在眉睫,促使全球加快了新型低 GWP 制冷剂的开发与应用研究。

现有研究工作表明,低 GWP 制冷剂的可用性因用途而异,需结合应用场景的性能需求,综合考虑制冷剂的基础物性、安全性(可燃性、毒性)、环境性能、与材料相容性、使用成本等因素。制冷剂的热物理性质决定了其应用场景的适用性,获取准确的物性数据,有助于开展循环分析、传热分析和系统性能评判,对于筛选和验证新型制冷剂必不可少。单一成分的低 GWP 新型制冷剂可选种类较为有限,且对其应用性能的研究也有待完善。目前已商业化的 HFO-1234yf、HFO-1234ze(E)制冷剂存在弱可燃性,在对安全性要求高的应用场景中无法直接使用。此外,HFOs、HCFOs 制冷剂具有碳碳双键,需验证与原有制冷剂接触材料的兼容性,避免系统故障或制冷剂泄漏。可在高温系统中应用的 HFO-1234ze(Z)、HCFO-1233zd(E)、HFO-1336mzz(Z)等制冷剂,其系统及部件仍处于开发阶段,目前仍缺少实际应用性能数据。为满足不同的应用需求,开发以 HFOs、HCFOs 为主体的混合制冷剂,将是一条切实可行的技术路线。但对其性能的评估还需要制冷剂供应商与应用系统开发商加强合作研究,在优化制冷剂配方的同时,不断优化运行系统与部件,加强系统结构及压缩机、冷凝器、蒸发器等关键部件的设计开发,以此协同提高新型制冷剂的应用性能,促进商业化应用。

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通信作者简介

张建君,男,教授级高级工程师,浙江省化工研究院有限公司,(0571)85222268,E-mail:zhangjianjun@sinochem.com。研究方向:消耗臭氧层物质(ODS)替代品以及含氟材料的开发。

About the corresponding author

Zhang Jianjun, male, professor of engineering, Zhejiang Research Institute of Chemical Industry Co., Ltd., 86-571-85222268, E-mail: zhangjianjun@sinochem.com. Research fields: alternatives to ozone depleting substances (ODS), fluorine-containing materials.