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燃料重整制氢技术研究进展

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摘要: 对燃料制氢技术的各种制氢方法和典型设备进行综述,明确随车燃料重整制氢技术的应用前景,并指出各种燃料重整方法的局限性.对蒸汽重整、部分氧化、自热重整、裂解、等离子裂解等技术进行归纳分析,针对随车燃料重整提出基于尾气的分类方法.

关键词: 氢气; 燃料; 重整; 车载; 制氢

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Progress of the Reformation of Fuel to Produce Hydrogen

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Abstract: This paper reviewed hydrogen production methods and typical equipment of fuel reforming technology, analyzed the application prospect of onboard hydrogen generation technology with fuel reformation, and pointed out the limitation of onboard hydrogen generation technology. The methods of steam reforming, partial oxidation, auto thermal reforming, decomposition and plasma were analyzed, and the classification method based on tail gas was proposed.

Keywords: hydrogen; fuel; reforming progress; onboard; hydrogen generation

氢能是一种清洁的能源,氢在自然界中广泛存在.水、生物质、石油、天然气等物质中都含有氢,氢气具有优良的燃烧特性^[1-6].与传统化石燃料相比,氢气具有很多方面的优势:氢气相比传统燃料几乎没有废气排放;氢气是一种可再生资源;氢气有很广泛的获得渠道,如天然气、煤炭、生物质、风能、太阳能等.如果将天然气、煤炭等燃料先转换为氢气,所产生的二氧化碳可以在制氢过程中去除,从而减少二氧化碳的排放.余热制氢技术利用发动机余热使醇类燃料发生重整反应,余热重整制氢一方面回收了发动机余热,提供了发动机的热效率;另一方面,制取的重整气因为富含氢气,可以通过重整气提高发动机的燃烧效率. Houseman 等^[7]将汽油和水混合,经过催化重整制备富氢气体. Cohn 等^[8-9]进行了等离子随车重整制氢的相关研究. Li 等^[10]在汽油机上进行了随车乙醇重整的实验研究. 徐元利等^[11]进行了发动机废气余热催化甲醇的相关实验. Song 等^[12]进行了二甲醚随车重整的相关研究. 随车重整燃料制氢主要采用技术可以分为 3 大类:1) 发动机废气不参与重整反应也不提供重整反应所需热量,燃料重整制氢设备独立进行工作;2) 发动机废气不参与重整反应,但提供重整所需热量;3) 发动机部分废气和重整燃料一起进行重整反应. 随车重整技术对于重整气有小型化的要求,本文主要分析各种可能被随车重整制氢所采用的技术.

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1 燃料重整制氢技术

1.1 蒸汽重整

很多燃料可以用来进行蒸汽重整,如甲醇、乙醇、甲烷等^[13-17]. 目前,甲烷蒸汽重整技术是最为成熟的制氢技术,已经成熟地用于工业化生产,在美国有超过 90% 的氢气来自甲烷蒸汽重整. 甲烷蒸汽重整的基本原理是:将甲烷和水蒸汽按照一定比例混合,通入催化重整器,重整压力为 3~25 个大气压,温度为 700~850 ℃,甲烷水蒸汽重整反应为 $\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2$ ($\Delta H = +206.16 \text{ kJ} \cdot \text{mol}^{-1}$). 水汽转换反应 $\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2$ ($\Delta H = -41.14 \text{ kJ} \cdot \text{mol}^{-1}$),水汽变换反应可以去除重整气中的 CO,并将其转换为氢气,水汽变换反应的发生温度大约为 600 ℃.

总体来说,甲烷蒸汽重整为吸热反应,需要外部提高热源. 甲烷蒸汽重整有一种较为实用的结构,即氢气筛选膜重整反应器^[18-19],该重整器的结构,如图 1 所示. 该反应器整合了甲烷蒸汽重整、水汽变换反应、氢气提纯这 3 个工序,甲烷蒸汽在重整的时候,产生的氢气通过氢气筛选膜过滤出来,促使重整反应更强烈地往产生氢气的方向反应.

相比管式重整器,平板重整器具有更加紧凑的设计结构^[20-27],重整器内换热更充分、流体的流动更平稳,比表面积更大. 因此,平板式重整器更加适合小型重整制氢设备,其结构如图 2 所示. 平板重整器具有类似三明治的结构,加热燃料在流动过程中进入空气层,空气层中含有燃烧催化剂,燃料与空气混合并在燃烧催化剂的作用下燃烧,燃烧所产生的热量通过隔板传递给重整层,加热燃料的流动方向和重整燃料的流动方向相反.

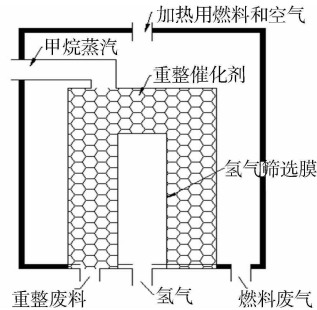


图 1 氢气筛选膜重整器结构图

Fig. 1 Membrane reactor for methane stem reforming

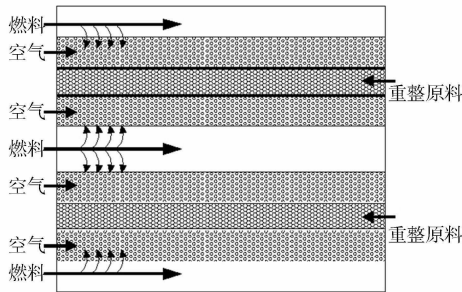


图 2 平板重整器

Fig. 2 Plate type reactor

甲醇蒸汽重整与甲烷蒸汽重整相比,在很多方面具有优势^[28-30]. 甲醇是液体,方便储存和运输,甲醇的重整温度在 250~350 ℃,反应温度适中,且作为化工产品,现阶段其产量大、价格低. 对于很多发动机,甲醇已经作为燃料供给发动机使用. 利用甲醇进行重整,重整产生的氢气改善发动机的性能. 很多汽车公司也已经开展了随车重整制氢方面的工作,如尼桑、丰田和克莱斯勒公司等^[11-16].

1.2 部分氧化

甲烷部分氧化的化学反应为 $\text{CH}_4 + (1/2)\text{O}_2 \rightarrow \text{CO} + 2\text{H}_2$ ($\Delta H = -36 \text{ kJ} \cdot \text{mol}^{-1}$). 该反应为放热反应,无需外部供热. 由于反应放出大量热量,部分氧化反应无需使用催化剂,利用该方法获得氢气一般需要 3 步工序,即部分氧化反应、水汽变换反应、氢气提纯. 现在也有很多大型设备利用部分氧化原理制氢,所用的原料除了甲烷外,还有乙醇、甲醇、汽油等,还包括各种使用价值较低的废油等.

相比于燃料蒸汽重整反应器,部分氧化重整器由于无需外部供热,可以设计得更紧凑. 由于该反应是放热反应,产生大量热量,使得反应器的温度很高,一般可以达到 1 000 ℃ 以上,为了降低温度减少能源浪费,可以采用催化剂. 另外,该反应的尾气温度较高,需采用复杂的设施来回收余热,这些因素都进一步增加部分氧化反应制氢的成本^[31-38]. 含有膜结构的部分氧化反应器,如图 3 所示. 部分氧化反应器采用氢气筛选膜,可以有效提高甲烷的转换率,采用吹扫气和催化剂,可以降低反应温度.

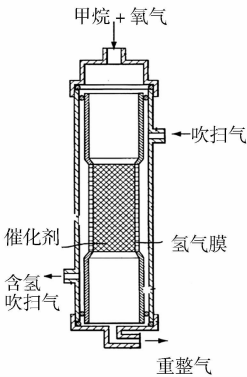


图 3 氧化反应器
Fig. 3 Oxidation reactor

1.3 自热重整

自热重整结合部分氧化重整和蒸汽重整, 反应器结构和部分氧化反应器的结构类似, 只是在进料中加入水, 自热重整原理可以适用于甲烷和很多液体类燃料^[39-49]. 自热重整利用部分氧化所产生的热量进行蒸汽重整反应, 合理调节燃料、空气和水的比例, 可以让自热重整的反应持续进行. 与蒸汽重整比较, 自热重整无需外部热源加热, 可以使反应器的结构简单化; 与部分氧化比较, 自热重整由于含水, 使得尾气中的烟尘大大降低.

1.4 氨裂解

图4为氨裂解的装置. 我国氨产量大, 价格较为便宜, 大部分氨都被用来生成化肥, 氨的运输和储存条件相对氢气安全很多, 氨经过裂解可以产生氢气. 因此, 氨成为制氢的较好原料. 氨裂解制氢的方程为 $2\text{NH}_3 \rightarrow \text{N}_2 + 3\text{H}_2$ ($\Delta H = 46.2 \text{ kJ} \cdot \text{mol}^{-1}$). 该反应是吸热反应, 需要外部供热, 随着温度升高, 反应速率变快, 较合适的反应温度在 $700\text{ }^{\circ}\text{C}$ 左右. 由于无需水汽变换反应, 氨裂解设备相比燃料蒸汽重整设备, 可设计得更加紧凑. 氨裂解在燃料电池中具有特别优势, 因为产生的氮气是惰性气体不参加反应, 不需要提纯氢气, 使得费用大大降低, 对于需要提纯的设备, 可以利用合理的膜结构分离氮气和氢气^[50-51].

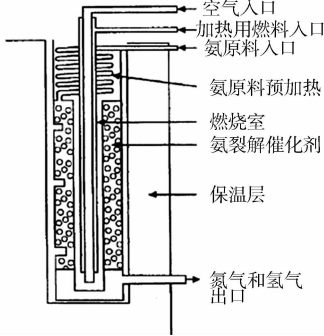


图4 氨裂解反应器
Fig. 4 Ammonia cracking reactor

1.5 甲烷裂解

甲烷在高温($850\sim 1\,200\text{ }^{\circ}\text{C}$)、催化剂辅助下发生裂解, 其裂解反应方程为 $\text{CH}_4 \rightarrow \text{C} + 2\text{H}_2$ ($\Delta H = 75 \text{ kJ} \cdot \text{mol}^{-1}$). 该反应是吸热反应. 如果甲烷同时作为裂解原料和提供热源燃料, 需要有 10% 左右的甲烷燃烧来保证反应进行, 然而该反应产生的碳会使催化剂中毒, 该装置现在还处于试验研究阶段^[52-55].

1.6 等离子重整器

图5为等离子重整反应器. 它可以利用甲烷、柴油等燃料制氢^[56-59], 且反应器的体积可以做得很小, 制氢效率很高, 利用甲烷蒸汽作为原料可以实现 95% 的原料利用率, 等离子重整器具有所产生的电弧具有很高的能量, 一般可以达到 $3\,000\sim 10\,000\text{ }^{\circ}\text{C}$. 因此, 反应物的反应速度很快, 无需再加入催化剂.

1.7 微通道反应器

微通道反应器的典型结构, 如图6所示. 微通道反应器是利用特殊加工工艺生产出来的微型催化反应装置^[60-61]. 该反应器供流体流通的通道一般小于 $500\text{ }\mu\text{m}$, 反应器由多层结构单元叠加而成, 每个结构单元的表面均有催化剂, 该结构使得流体更容易在催化剂的作用下发生反应, 流动稳定, 热传导较好. 因此, 微通道反应器能有效加快反应速度. 微通道反应器较传统反应器, 尺寸大为缩小. 因此, 该反应器可以较好地适用有体积限制的场合, 如燃料电池和随车重整等.

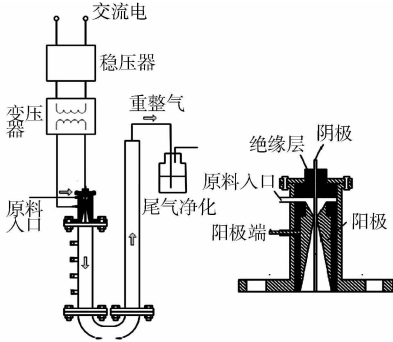


图5 等离子反应器
Fig. 5 Plasma reactor

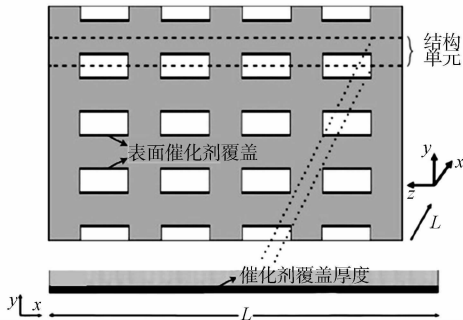


图6 微通道反应器结构示意图
Fig. 6 Micro channel reactor

2 研究展望

燃料制氢技术有很广的应用, 随车重整燃料制氢可以很好地促进发动机性能提高, 甲烷蒸汽重整、甲醇蒸汽重整、自热重整、氨裂解、甲烷裂解等各项技术都有各自的优缺点, 部分氧化和自热重整在燃料

重整阶段较为简单,但是后续的净化较为困难。

管式重整器、板式重整器、微通道重整器、膜技术、等离子重整技术等各项技术的进步推动着重整器设计的发展,对于随车重整燃料制氢,板式重整器、膜技术、等离子重整技术等都非常适合小型化的要求。随着催化剂研究的发展和反应器结构设计的发展,随车重整燃料制氢技术也会有更进一步的发展。

在汽油、柴油等燃料中掺加富氢气体可以有效提高发动机性能,但是氢气的运输和储存都较为困难。随车重整制氢技术可以较好地符合掺氢燃烧的要求,随着环保法规的要求越来越严格,已经有越来越多的发动机厂商在研究随车重整制氢技术,对各种随车重整技术的研究也将会成为未来发动机节能减排技术的一个重要内容。

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