

# 国VI GDI汽油车颗粒物排放特性与微观形貌特征

胡志远, 赵心语, 夏孝程, 宋博

(同济大学汽车学院, 上海市 201804)

**摘要:** 试验研究了国VI GDI汽油车WLTC(world light vehicle test cycle)循环的颗粒物质量和固态颗粒物数量排放特性以及颗粒物微观形貌特征。结果表明,该车颗粒物质量与固态颗粒数量排放均低于国VI法规限值;固态颗粒数量累积呈3段变化形态,0~300s、600~800s与1500~1800s的累积量显著大于其他时间段。冷机起动时颗粒数量较多,仅有0~300s的累积量已大于热机起动时颗粒物数量总累积量。颗粒物主要由基本碳粒子堆积形成,形状不规则。基本碳粒子由大量微晶碳层组成,内核无序而外壳有序;重叠时有的形成大颗粒物,有的形成大碳粒子,基本碳粒子直径、碳晶长度等特征参数呈单峰分布;与GDI(gasoline direct injection)汽油机台架试验结果比较,整车试验基本碳粒子直径、碳晶长度和曲率分布相对集中且数值相对较小。

**关键词:** 颗粒物;微观形貌;缸内直喷汽油车;国VI法规

中图分类号: U467.1+9

文献标志码: A

## Emission Characteristics and Microscopic Morphology of Particles from a Gasoline Direct Injection Vehicle based on China VI Emission Regulation

HU Zhizhan, ZHAO Xinyu, XIA Xiaocheng, SONG Bo

(School of Automotive Studies, Tongji University, Shanghai 201804, China)

**Abstract:** The mass, cumulative quantity and micro-morphology of particulate matters from a China VI gasoline direct injection (GDI) vehicle during Worldwide Harmonized Light Vehicles Test Cycles (WLTC) have been researched experimentally. The results showed emission factors of particulate mass (PM) and particulate number (PN) were both lower than the regulation limits; The cumulative amount of particulate matters varied in three stages, which was significantly higher in the first 300s, the last 300s and 600~800s. The PN was much more when

engine was started under cold condition; the accumulation from first 300s had been over the total amount of hot-engine operation. Particulate matters were mainly formed by basic carbon particles whose shapes were irregular. The basic carbon particles were composed of many microcrystalline carbon layers, the inner core was disordered while the outer shell was ordered; When basic carbon particles overlapped, some formed large particles and others formed large carbon particles. The characteristic parameters of basic carbon particles were all unimodal distribution, similar to the GDI engine; The distribution of basic carbon particles diameter, carbon crystal length and carbon crystal curvature were relatively concentrated and the values were all lower than test results of the engine bench.

**Key words:** particulate matters; microscopic morphology; gasoline direct injection vehicles; China VI regulation

颗粒物作为大气污染的主要影响因素<sup>[1]</sup>对人类健康有着巨大的危害<sup>[2]</sup>,环保法规对它的限制也日益严苛:国VI(a)增加对汽油车颗粒物质量和固态颗粒物数量(particulate number, PN)的限值要求,试验工况也由新欧洲驾驶循环(new European driving cycle, NEDC)改为全球统一轻型车测试循环(worldwide harmonized light vehicles test cycle, WLTC),而国VI(b)的颗粒物质量限值比国VI(a)进一步降低33%<sup>[3]</sup>。研究表明缸内直喷(gasoline direct injection, GDI)汽油车的PN高于进气道喷射(port fuel injection, PFI)汽油车与加装柴油颗粒物捕集器(diesel particulate filter, DPF)的柴油车<sup>[4-7]</sup>,对应WLTC循环的颗粒物质量(particulate mass, PM)与PN比NEDC循环大幅升高<sup>[7-9]</sup>。目前的国

收稿日期: 2019-04-30

基金项目:国家重点研发计划(2017YFB0103405)

第一作者:胡志远(1970—),男,副教授,主要研究方向为汽车能源与低排放设计技术。

E-mail:huzhiyuan@tongji.edu.cn



论文拓展介绍

V GDI汽油车WLTC循环的PM可能低于国VI法规限值,但其PN排放较高<sup>[10]</sup>。

颗粒物数量已成为GDI汽油车设计与开发的重要挑战,可行的应对方案之一是为车辆加装汽油机颗粒物捕集器(gasoline particulate filter, GPF)。研究表明GPF可有效减少GDI汽油车的颗粒物数量<sup>[11]</sup>,使PN低于法规限值<sup>[12-13]</sup>,但是也面临和DPF(diesel particulate filter)类似的再生问题。

颗粒物再生与其本身的氧化性能密切相关,其微观结构对氧化性能有决定性影响<sup>[14-16]</sup>。目前对移动源颗粒物微观形貌的研究以柴油机为主<sup>[17-20]</sup>,其目的是为后处理装置的氧化机理提供研究基础。有关GDI汽油机颗粒物的研究相对较少且主要集中于颗粒物质量、数量及组分分析<sup>[21-22]</sup>。例如:夏晨<sup>[23]</sup>研究了甲苯及乙醇添加剂和GDI汽油机颗粒物数量浓度的关系;郑荣<sup>[24]</sup>比较了GDI汽油机与PFI汽油机颗粒物数量、有机碳、元素碳组分的差异。关于颗粒物微观结构的研究很少,且研究对象为汽油机。例

如:Gaddam等<sup>[25]</sup>研究了空燃比、点火角和GDI汽油机颗粒物理化特性的相关性;潘锁柱<sup>[26]</sup>研究了空燃比、点火时刻、喷油时刻以及EGR率等对GDI汽油机尾气颗粒物基本碳粒子微观结构的影响。

本文以某缸内直喷汽油车为研究对象,试验研究其整车测试工况的颗粒物质量、固态颗粒数量排放因子和瞬态特性以及颗粒物微观形貌特征,为GDI汽油车颗粒物氧化机理与GPF再生机理的研究提供基础。

## 1 试验方案

### 1.1 试验车辆与燃油

试验车辆为一辆面向国VI开发的缸内直喷汽油车,整车整备质量为1520kg;发动机主要技术参数见表1;试验燃油符合国VI标准,具体理化参数如表2所示。

**表1 试验车辆主要技术参数**  
**Tab. 1 Main technical parameters of test vehicle**

排量/L	缸径/mm	行程/mm	额定功/kW	额定转速/(r·min <sup>-1</sup> )	额定转矩/(N·m)	额定转速/(r·min <sup>-1</sup> )	压缩比	进气方式	后处理装置
1.4	74.5	80	110	5000	250	1500~3500	10:1	涡轮增压	三效催化器

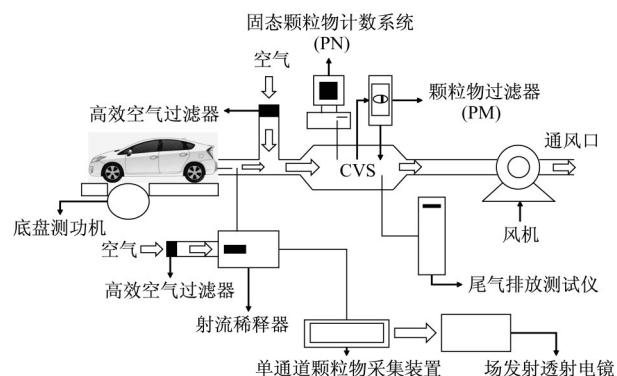
**表2 试验燃油主要理化指标**  
**Tab. 2 Main physicochemical indicators of test fuel**

理化参数	研究法辛烷值(RON)	20℃密/kg·m <sup>-3</sup>	硫含量/mg·kg <sup>-1</sup>	氧含量体积分数/%	烯烃含量体积分数/%	芳香烃含量体积分数/%	苯含量体积分数/%
测试指标	93	745.7	6.1	0.26	12.2	29.9	<0.1
法规限值	92~94	720~775	≤10	≤2.7	10~15	27~32	≤0.8

### 1.2 试验设备与循环

试验设备由德国Weiss整车试验环境舱、德国Schenk底盘测功机、日本HORIBA全流稀释定容采样系统(constant volume dilution system, CVS)、日本HORIBA MEXA-2000固态颗粒计数系统(solid particulate counting system, SPCS)、芬兰DEKATI射流稀释器(fine particle sampler, FPS)、美国FEI TecnaiG2F20S-Twin场发射透射电子显微镜(field transmission electron microscope, TEM)组成,如图1所示。

试验循环采用WLTC循环,试验时长1800s、行驶里程23.27 km,由低速、中速、高速和超高速4个工况段组成,循环的速度、加速度与时间的关系如图2所示。相比NEDC循环,WLTC循环的工况覆盖范围更广(最大车速升高约9.4%,最大加速度升高



**图1 试验设备示意图**

**Fig. 1 Schematic diagram of test equipment**

51.3%),以加、减速为代表的过渡工况变化更为频繁,具有较强的瞬态特性<sup>[27-28]</sup>。

### 1.3 试验方案

试验开始前在环境舱(温度298K、压力

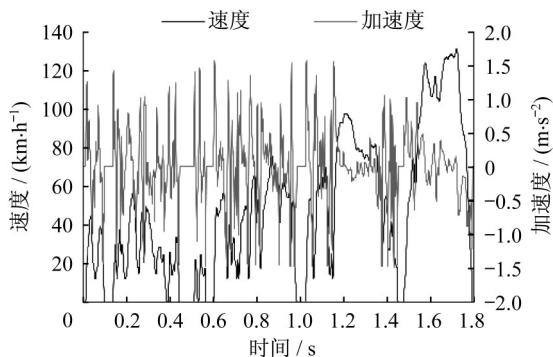


图2 WLTC循环时间与速度、加速度关系曲线

Fig. 2 Velocity and acceleration curves of WLTC cycle

0.1 MPa)中浸车24h,使用底盘测功机开展WLTC测试试验。试验时利用CVS以 $8.4 \text{ m}^3 \cdot \text{min}^{-1}$ 的流量稀释车辆尾气,采用滤纸称重法测量尾气中的颗粒物质量,并使用SPCS测量尾气中 $23 \text{ nm} \sim 2.5 \mu\text{m}$ 的固态颗粒数量。同时,采用FPS抽取部分(体积分数约3%)尾气,借助单通道颗粒物采集装置,使用47mm的石英滤膜采集尾气中的颗粒物作为分析样本,稀释温度为180°C,采样流量 $21 \text{ L} \cdot \text{min}^{-1}$ 。对采集的样本使用超声震荡法预处理,然后将含颗粒物的无水乙醇滴至碳膜铜网,置于白炽灯下烘干,使用场发射透射电子显微镜观察颗粒物的微观形貌特征。

为确保分析样本数量充足和准确性,分别采集3次冷机起动与3次热机起动车辆WLTC循环颗粒物排放,冷机试验完成30min后开始热机试验。

## 2 试验结果与分析

### 2.1 颗粒物的质量与数量

表3为试验车辆颗粒物质量及固态颗粒数量排放因子。可见,该试验车辆PM与PN均符合国VI法規限值要求。

表3 颗粒物排放因子

Tab. 3 Particulate emission factor

排放因子	PM/mg·km <sup>-1</sup>	PN/ $10^{11}$ 个·km <sup>-1</sup>
试验值	1.77	2.24
国VI(a)	4.5	6.0
国VI(b)	3.0	6.0

试验车辆WLTC循环颗粒物累积数量如图3所示,可见该车颗粒物数量排放累积呈现3段变化形态:0~300s、600~800s与1500~1800s累积量明显大于其他时间段(表4)。颗粒物累积量数幅度升高即表明颗粒物生成量增加,研究表明空间浓混合气

不完全燃烧与壁面油膜池火燃烧是GDI汽油机产生颗粒物的主要诱因<sup>[29-31]</sup>。前300s为起动工况与低速工况,一方面为确保车辆起动而多喷油,另一方面缸内气流强度较弱,两方面效应叠加使得空间浓区变大、壁面油膜变多,颗粒物生成增加。600~800s虽然转速较高、气流强度变大,但行驶速度及加速度频繁变化使得转速与负荷亦相应变化,节气门与废气旁通阀开度波动较大,涡轮增压器动态响应具有滞后性,使实际空燃比与当量比间有不同程度的误差<sup>[32]</sup>,尤其是从减速转到加速工况时易形成较浓的空间混合气,不完全燃烧比例升高,颗粒物排放增加。1500~1800s以高速工况与大加速工况为主,常加浓混合气以实现车辆的动力需求,虽然高转速使气流强度更大,但是燃油雾化、蒸发、混合以及颗粒物滞留缸内的绝对时间相应减少,前两者使燃烧室中浓混合气的质量分数变多,后者削弱燃气对颗粒物的高温氧化作用;有研究显示喷油器头部残余油量变多使积碳变多,也是颗粒物数量升高的原因之一<sup>[33]</sup>。

同时,由图3可见,冷机起动工况的颗粒物累积数量约为热机起动工况的2.5倍,主要原因是0~300s的冷起动、起燃与暖机工况产生的颗粒物数量极多,已大于热机起动时循环总累积量(图3)。这是因为,相比热机状态,冷机起动时因燃烧室温度较低、壁面油膜的蒸发量小,池火燃烧加剧<sup>[34]</sup>;其次由于负荷较低而传热量较大使燃气温度下降,对颗粒物的氧化能力减弱。车辆起动后,为降低循环的污染物总量,常推迟点火以缩短三效催化器的升温时间,使其尽快起燃;此时为确保燃烧的稳定性,常采用二次喷射的供油策略:第1次喷油(预喷)位于进气冲程,使点火前缸内形成部分均质混合气;第2次喷油(主喷)位于压缩冲程,使点火时火花塞周围能形成略浓的混合气,防止空气充量的冷熄或吹熄作用而发生失火。这种工况下,导致颗粒物累积数量高于热机的原因有三点:一是主喷时喷油器与活塞头部的距离较小,使燃油的湿壁现象较为严重,加之燃烧室表面温度较低使燃油蒸发量减少,较多的壁面油膜发生池火燃烧;二是二次喷油导致混合气分层,预混火焰于浓区燃烧时生成的碳烟;三是缸内燃气温度不高,难以氧化已生成的颗粒物。同样,冷机起动后发动机暖机时为减少冷却水的升温时间亦常采用混合气分层二次喷油策略,颗粒物累积数量较多<sup>[35-36]</sup>。

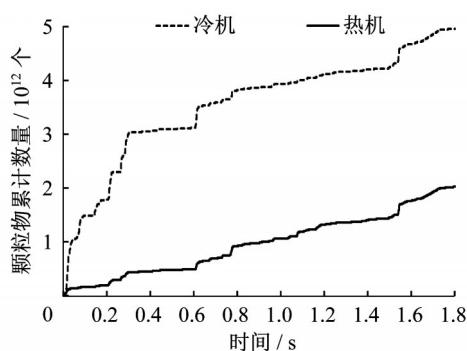


图3 颗粒物累积排放数量

Fig. 3 Cumulative emissions of PN

## 2.2 颗粒物微观形貌

一般认为,在采样滤膜上选取20处及以上位置制取样本,拍摄TEM图像,得到的结果具有代表性<sup>[25]</sup>。选取30处位置取样,参考文献[26]对GDI汽油机颗粒物与基本碳粒子微观形貌的分类方法,从

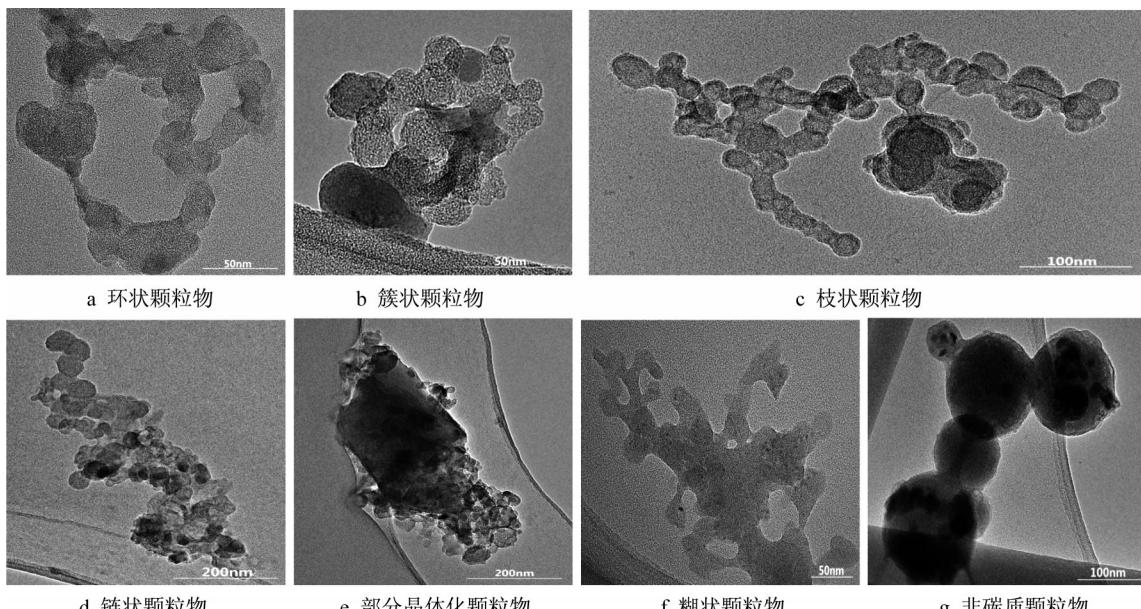


图4 颗粒物典型微观形貌

Fig. 4 Typical micro-morphology of particulate matter

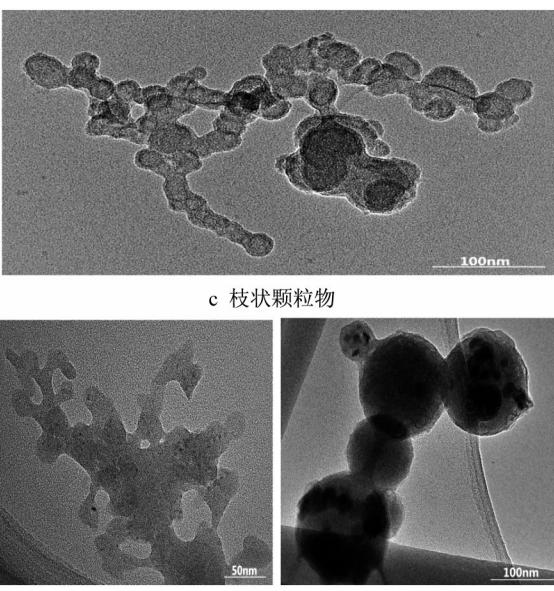
由图5可见,缸内直喷汽油车尾气颗粒物基本碳粒子是由大量微晶碳层组成的类圆形结构,呈明显的内核-外壳不定形结构,内核碳晶排列极为杂乱,无规律可循;外壳碳晶结构比较规则,表现为向心排列的有序结构。尺寸较小的碳粒子(图5 a~5d)表面粗糙度明显大于尺寸较大者(图5 e~5g),且其内核与外壳的碳晶排列更加疏松,对应的有序程度更低。对于存在重叠部分的相邻碳粒子,有的重叠区域中尚能显现出较为清晰的外壳边界(图5c),有的则基本呈融合状态,无法明确分辨两者外

表4 不同时间段内颗粒物累积数量占比  
Tab. 4 The proportion of accumulated PN in different time periods

时间段/s	颗粒物数量占比/%	
	冷机起动	热机起动
0~300	60.5	21.4
400~600	14.1	21.4
1500~1800	14.3	28.0

TEM图像中归纳得到GDI汽油车尾气颗粒物的典型微观形貌如图4所示,基本碳粒子典型微观形貌如图5所示。

由图4可见,缸内直喷汽油车颗粒物由几个、几十个甚至几百个基本碳粒子堆积而成,颗粒物的分形结构极不规则,有环状(图4a)、簇状(图4b)、枝状(图4c)、链状(图4d)等,部分颗粒物呈糊状(图4f),且有一定数量的颗粒物已部分晶体化(图4e),也有少量颗粒物以非碳质(图4g)的形式存在。



壳的界限(图5h)。当多个基本碳粒子相互重叠时,它们既有可能完全融合形成一个不规则的大颗粒物,导致原先基本碳粒子的典型结构几乎完全消失(图5i);又有可能2个或多个基本碳粒子的外壳融合形成一个内核中空的大碳粒子(图5j),此时颗粒物中碳晶的有序程度较高,排列比较规整,石墨化程度也较高。另外,观察到的颗粒物中也有少量具有无定型结构的基本碳粒子(图5k,5l),其碳化程度较低,不存在清晰可见的碳晶结构。

取基本碳粒子直径、碳晶长度、碳晶曲率和碳晶

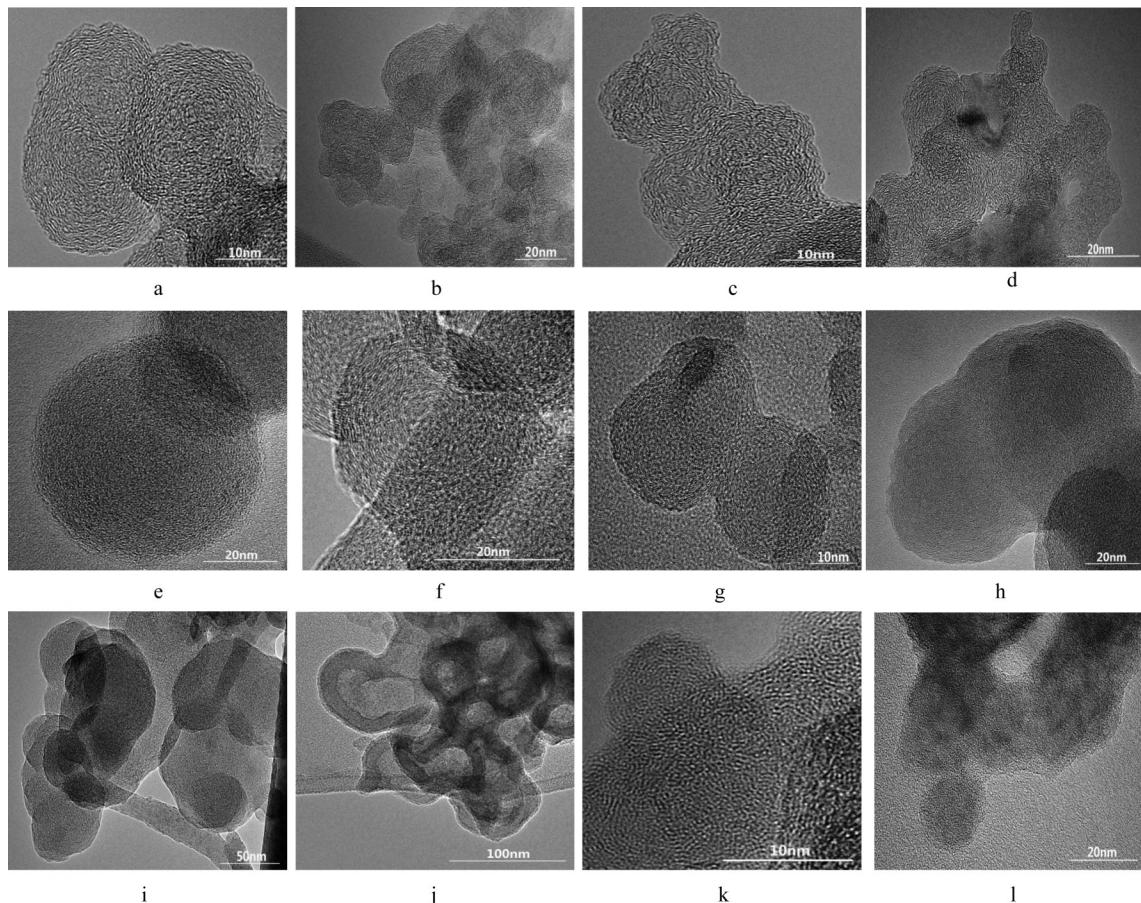


图5 基本碳粒子微观结构

Fig. 5 Microstructure of basic carbon particles

层间距作为特征参数<sup>[37-38]</sup>,分析颗粒物微观结构特征,如图6所示。

由图6a可见,该车尾气颗粒物基本碳粒子直径范围为3.8~53.6 nm,呈单峰分布形式,峰值直径约为20~22 nm,平均直径为23.2 nm,平均方差等于9.05,直径为10~32 nm碳粒子数量最多,约占总数量的81.8%。与宋博等<sup>[10]</sup>利用颗粒物粒径谱仪(engine exhaust particle seizer, EEPS)测量得到的颗粒物粒径分布基本相同,但峰值粒径从15 nm升高至20~22 nm。这是因为,EEPS测量的是颗粒物空气动力学直径,包含一些半挥发性组分,与基本碳粒子直径的定义有显著差异<sup>[37]</sup>。相比Liati欧VI缸内直喷汽油车WLTC循环颗粒物基本碳粒子直径分析结果,该国VI GDI汽油车颗粒物中小于10 nm基本碳粒子数量占比减小,40 nm以上碳粒子数量占比升高,基本碳粒子平均直径从18 nm升高至23.2 nm,升高约29%。相比潘锁柱<sup>[26]</sup> GDI汽油机研究结果,该车的基本碳粒子直径较小。

由图6b可见,该车尾气颗粒物基本粒子碳晶长度呈单峰分布,约88.9%碳晶长度小于1 nm。峰值

碳晶长度约0.2~0.4 nm,平均碳晶长度约0.53 nm,平均方差0.45。与潘锁柱<sup>[26]</sup>和Gaddam等<sup>[25]</sup> GDI汽油机结果比较,该GDI汽油车颗粒物基本碳粒子碳晶长度分布特性基本相同,碳晶长度峰值减少至0.2~0.4 nm,平均值减小至0.53 nm左右。相比Liati欧VI GDI汽油车WLTC循环研究结果<sup>[38]</sup>,该GDI汽油车尾气颗粒物基本粒子碳晶长度分布趋势总体相似,碳晶长度峰值较小。

由图6c可见,该GDI汽油车颗粒物基本粒子碳晶曲率为1.1~1.4的碳粒子数量最多,约占总数量的72.9%;峰值曲率出现于1.2~1.3处,数值略大于Gaddam等<sup>[25]</sup>的研究结果(1.1~1.2),小于潘锁柱<sup>[26]</sup>的研究结果(1.2~1.4);碳晶曲率平均值约为1.36,介于Gaddam等<sup>[25]</sup>(1.175~1.210)与潘锁柱<sup>[26]</sup>(1.45~1.52)的研究结果之间。

由图6d可见,该GDI汽油车颗粒物基本粒子碳晶层间距呈单峰分布,最大层间距约为0.38~0.40 nm,平均层间距约为0.40 nm,平均方差0.06;对比文献<sup>[26]</sup>的研究结果,碳晶层间距的峰值与平均值基本相似,但该GDI汽油车颗粒物基本碳粒子碳晶

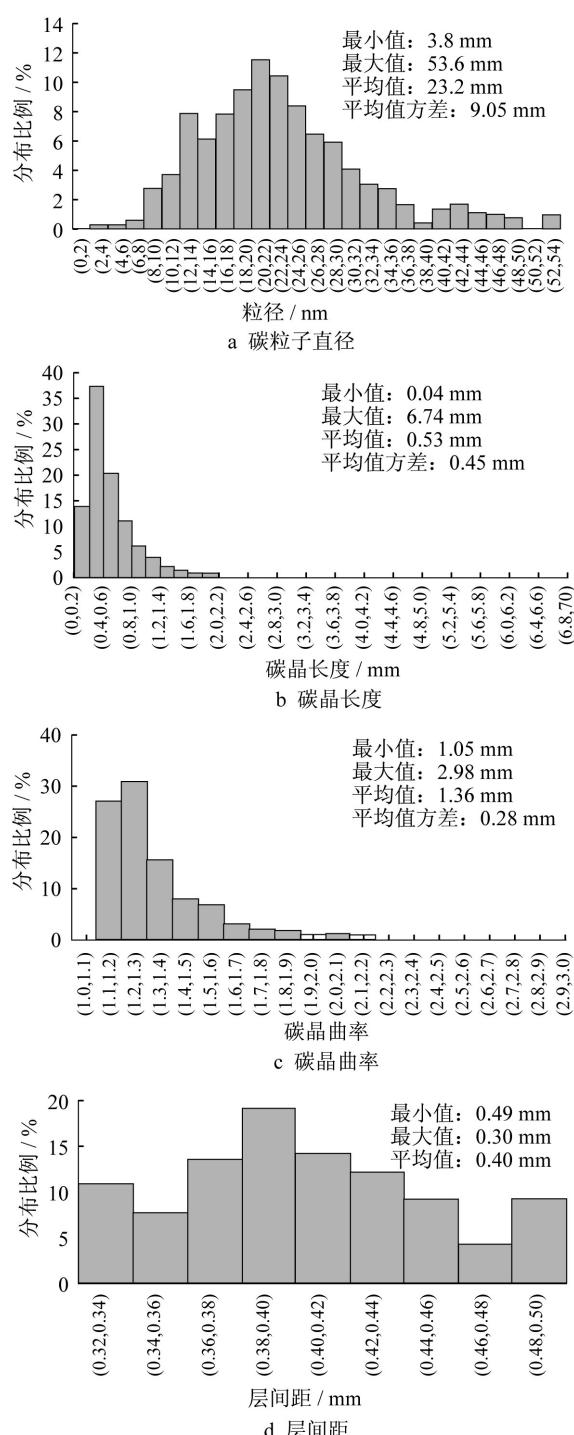


图6 基本碳粒子特征参数

Fig. 6 Characteristic parameters of basic carbon particles

层间距小于0.34nm和大于0.46nm的比例增大,基本碳粒子碳晶层间距分布相对更均匀。

### 3 结论

(1) GDI汽油车WLTC循环颗粒物数量累积呈

3段变化形态,0~300s、600~800s与1500~1800s的累积量明显大于其他时间段,起动与低速工况、加、减速过渡工况以及高速与高负荷工况的颗粒物累积量较多。

(2) GDI汽油车排放的尾气颗粒物由数量不等的基本粒子堆积形成,包括环状、簇状、枝状、链状等不规则结构,以及少数糊状颗粒物或部分晶体化颗粒物形态。

(3) GDI汽油车尾气颗粒物基本碳粒子呈类圆形的内核-外壳不定形结构,内核无序而外壳有序;尺寸较小的颗粒表面粗糙度较大,有序度低;相邻碳粒子重叠时有的保留外壳界线,有的基本融合;多个碳粒子重叠时有的完全融合形成大颗粒物,有的外壳融合形成内核中空、石墨化程度较高的大直径碳粒子。

(4) GDI汽油车尾气颗粒物基本碳粒子直径、碳晶长度、碳晶曲率和层间距均呈单峰分布,峰值点分别出现在20~22 nm、0.2~0.4 nm、1.2~1.3和0.38~0.40 nm,平均值分别为23.2 nm,0.53 nm,1.36和0.40 nm。基本碳粒子直径、碳晶长度及碳晶曲率的分布较集中且数值小于汽油机台架试验结果。

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