# Advance in Morphology of Fouling in the Exhaust Gas Recirculation Coolers

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#### ABSTRACT

Morphology, as a science that describes the shape and structure of fouling, is a key component of a comprehensive evaluation of fouling. It also serves as a perspective in deepening the understanding of fouling mechanisms. Nevertheless, a standardized morphological evaluation system of fouling has not yet been established. This study takes exhaust gas recirculation cooler fouling as the research object, for the first time, reviews the progress in morphology over the past 25 years, and then proposes the evaluation system. These findings show that the methods used to evaluate fouling morphology can be classified into visual inspection, optical microscope, digital microscope, surface profiler, scanning electron microscope, and transmission electron microscope. The observation scale decreases from macroscopic to mesoscopic, microscopic, and nanoscopic. The evaluation categories may include attributes such as color, glossiness, wetness, softness, densification, roughness, fragmentation, connection method, orderliness, and more. Defining and categorizing, integrating full scale, developing quantifiable parameters, and reinforcing the full process for fouling should be further deepened.

#### Introduction

Heat exchangers are one of the most important means for exchanging and managing heat [1,2]. However, the accompanying fouling has always been a difficult problem, which leads to the deterioration of the hydraulicthermal performance of the heat exchanger, which not only undermines the accurate management of heat and flows daily but also shortens the service life of the heat exchanger, resulting in additional economic losses and energy consumption [3–5].

Exhaust gas recirculation (EGR) cooler fouling is one of the most representative scenarios among all heat exchanger fouling. The four typical types of EGR coolers inevitably generate a lot of fouling with many diverse and complex components [6–9], because the engine exhaust gas consists of a large number of solid and liquid particulate matter as well as a mixture of organic and inorganic vapors. In addition, as the engine operates under different loads from time to time. So, the flow, temperature, and pressure of the exhaust gas are constantly changing. Therefore, the diverse and complex components of the exhaust gas and the variable temperature and flow fields make the fouling more significant and complex in the EGR cooler.

As Peter Drucker said, "You can't manage what you don't measure." Therefore, if we want to manage fouling scientifically, measuring or evaluating it is one of the prerequisites. Of all the evaluation types, morphology is intuitive and covers both macro and micro scales; it can be evaluated qualitatively as well as quantitatively; therefore, morphology is one of the basic ways of evaluation. In addition, direct evidence of fouling generation and development can be obtained through the measurement of fouling morphology in time and space, rather than indirect reasoning by way of heat transfer efficiency or pressure drop, which is more credible. More importantly, fouling morphology is also one of the most important factors affecting the performance of heat exchangers; for example, fouling skeleton size affects heat exchanger effectiveness [6]. Therefore, establishing the relationship between fouling morphology and performance can help to optimize heat exchanger performance. Overall, fouling

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Nomenclature					
AFM	Atomic force microscopy	SEM	Scanning Electron Microscopy		
CT	Computed tomography	STM	Scanning tunneling microscopy		
EGR	Exhaust gas recirculation	TEM	Transmission electron microscopy		
OM	Optical microscopy	X-ray CT	X-ray computed tomography		
SAXS	Small Angle X-ray Scattering	XRD	X-ray diffraction		

morphology plays an important role in describing fouling, understanding fouling generation and growth processes, and improving heat exchanger performance.

With the high rate of development and application of measurement techniques in recent years, new discoveries on the morphology of fouling have allowed a better comprehension. However, the results of these studies are disseminated in various publications, and no comprehensive review has been conducted. Therefore, to address this problem, the present work systematically reviews the findings of EGR cooler fouling morphology during the period from 1999 to 2023, focusing on the following issues:

- What are the evaluation methods of fouling i. morphology?
- What are the evaluation categories of fouling ii. morphology?
- What are the advances and gaps in fouling iii. morphology?

## Methodology of the proposed evaluation system

To make the proposed EGR cooler fouling morphology evaluation system as comprehensive, hierarchical and consensual as possible. This work follows the methodology as shown in Figure 1. The evaluation system was proposed through three stages: literature search, screening, and summarization. It is worth mentioning that the evaluation system referred to in this work consists of five parts: evaluation object, evaluation method, evaluation scale, evaluation category, and typical descriptive words.

# Morphology determination of fouling

General methods and typical techniques for fouling morphological characterization are given in Table 1 [10-29], along with reference cases. Roughly speaking, the evaluation methods can be classified into probe, visual, and other. Typical techniques in the probe type include atomic force microscopy (AFM), scanning tunneling microscopy (STM), and probe type surface

Table 1. General evaluation methods and techniques for fouling morphology.

Туре	Topic technique	References
Probe	Atomic Force Microscopy (AFM)	[10,11]
× ·	Scanning Tunneling Microscopy (STM)	[12,13]
	Probe Type Surface Profiler	[14,15]
Visual	Optical Microscopy (OM)	[16,17]
	Optical Type Surface Profiler	[18,19]
	X-ray Computed Tomography (X-ray CT)	[20,21]
	Scanning Electron Microscopy (SEM)	[22,23]
	Transmission Electron Microscopy (TEM)	[24,25]
Other	X-ray Diffraction (XRD)	[26,27]
	Small Angle X-ray Scattering (SAXS)	[28,29]





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profiler; while typical techniques in the visual type are optical microscopy (OM), X-ray computed tomography (X-ray CT), ray computed tomography (CT), transmission electron microscopy (TEM), etc. Other types are related to advanced optical diagnostics, such as X-ray diffraction (XRD), and small Angle X-ray Scattering (SAXS), both of which mainly resolve the structural information of the fouling rather than the formal characteristics.

It is worth stating that the shape, size, and connection of the fouling forms are central to this work, and that structural features are beyond the scope of investigation of this work.

#### **Evaluation method for EGR cooler fouling**

The problem of EGR cooler fouling was first raised by Girard et al. [30] in 1999. Since 2004, scientists have begun to pay attention to the morphological characteristics of fouling. Ismail et al. [31] visually observed the fouling of the inlet header at both  $45^{\circ}$  and  $60^{\circ}$ and found that the fouling of the inlet header at  $60^{\circ}$ was more homogeneous, and further measured the thickness of the fouling based on optical microscope [32]. They were the first team to describe the fouling morphology of EGR cooler, and due to the simplicity and efficiency of this evaluation, the experiments could be carried out on-site. Since then, the evaluation of fouling morphology based on visualization and optical microscopy has flourished, and it has received a large number of positive comments from scientists [33-39]. There have been some new changes in how optical microscopy measurements are made in recent years. Several studies have used digital microscope [40-42] or optical surface profiler [18,19] to obtain three-dimensional images of fouling surfaces, which, unlike traditional two-dimensional pictures, have resulted in a more stereoscopic and intuitive presentation of fouling morphology.

Although optical microscope can observe the morphological features of fouling under a certain intensification, they are mainly in millimeters and micrometers, which are still macroscopic morphological features, and it is impossible to analyze the composition of fouling from smaller constituent units [43,44]. Therefore, some researchers began to use equipment with higher magnification to observe fouling, *i.e.*, scanning electron microscope. Lance *et al.* [45] first characterized the microscopic morphology of fouling with scanning electron microscope in 2010 and found the mud-cracking characteristics of fouling and nanoscale pearl-string-like structure of particulate matter, as well as the bridging connection mode. Since then, scanning electron microscope has also begun to be widely applied to evaluate fouling morphology [42,46–52].

With the rapid development of material characterization techniques, transmission electron microscope, which can observe nanostructures more clearly, has also started to be applied to fouling evaluation. Sluder *et al.* [49] employed this technique for the first time in 2013 to observe the smallest unit of fouling formation, *i.e.*, the particulate matter, in its original form. Arnal *et al.* [53] and Paz *et al.* [18] also used this approach to observe the characteristics of foulingforming particulate matter to elucidate the differences in the morphology of fouling exhibited at the macroscopic level at the origin.

To sum up, morphology is a discipline that focuses on visual evaluation, as shown in Figure 2 [30-32,45,49]. In evaluating EGR cooler fouling, six evaluation methods, namely, visual observation, optical microscope, digital microscope, surface profiler, scanning electron microscope, and transmission electron microscope, are mainly used. Among them, visual observation optical microscope, digital microscope, and surface profiler, mainly observe the morphology of fouling at macroscopic and mesoscopic scales, with the size distribution of millimeter to micrometer. Scanning electron microscope mainly observes the morphology of fouling at mesoscopic and microscopic scales, with the size distribution of micrometer to nanometer; and transmission electron microscope mainly observes the morphology of fouling at the nanoscale, with the size of nanometer. Thus, the six evaluation methods completely cover the morphology of fouling at the macroscopic, mesoscopic, microscopic, and nanoscopic scales. Furthermore, it is a process of continuously reducing and analyzing the constituent units of fouling from smaller scales, which makes it possible to interpret the formation mechanism and process of fouling at smaller scales.

Compared to the generally available methods for fouling morphology evaluation, most of the techniques have been well adopted for EGR cooler fouling characterization, but three techniques, AFM, STM, and CT, are still missing.

#### **Evaluation system**

Table 2 [6,7, 9,18, 19,40–42, 45–91] shows the proposed evaluation system of morphology based on the past descriptions of EGR cooler fouling morphology in the literature. It is categorized into four scales, including macroscopic, mesoscopic, microscopic, and nanoscopic, which correspond to five characterization

objects: fouling layer, fouling layering and clustering, particulate matter, and primary carbon particle. The evaluation scales become smaller and smaller and together construct a comprehensive and three-dimensional evaluation system. Each characterization object corresponds to multiple evaluation categories. Taking the fouling layer as an example, it can be described from multiple categories, including color, gloss, dryness, softness, hardness, denseness, roughness, and fragmentation, and the typical terms and evaluation methods that can be used for describing each of these categories are also given in the Table 2. Table 2 also contains a part of the morphology of the fouling that is intrinsically linked to the



String of pearls

[42, 45]

Connection method

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composition and generation process. The differences in the foul's color are due to their composition. Fouling composed of dry carbon particles is usually black in color, whereas those composed of carbon particulates co-mingled with an organic condensate, such as hydrocarbons, are usually brown, yellow, or gold in color, whereas gray and white fouling have not yet been well explained. Cracks and grooves in the fragmentation usually imply that the fouling has experienced condensate ingress or evaporation, triggering structural collapse. In terms of constituent units and connections, the fouling can be divided into two layers, the top and the bottom, which are connected by columns; each layer consists of many clusters of agglomerates or large particles stacked in a mechanically interlocking bridge and the clusters are made up of particles connected by strings of pearls, with each particle in turn consisting of primary carbon particles connected by a chain-like structure.

#### **Research gaps and outlook**

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Past studies have shown fruitful progress in the morphology of fouling in EGR coolers, moving from the macro to the micro-scale and even the nanoscale, from qualitative to somewhat quantitative evaluations, and from a single morphology description to the correlation with the evolution of fouling. However, there are still some gaps, as follows:

- i. Clarify the definition and classification of fouling from the morphological perspective. Fouling is the deposition of unwanted materials on the heat transfer surface [92]. However, this definition is broad, especially since fouling is characterized by diversity and complexity, and there is no consensus on a precise definition as well as a classification for EGR cooler fouling, which hinders the comparison of similar or dissimilar fouling. However, the advancement of morphology maybe can provide the possibility to deal with this issue. Therefore, to define fouling and establish a corresponding basis for its classification from a morphological point of view is a sub-topic that deserves to be expanded in depth.
- ii. Organic integration of full-scale morphology. Past studies have usually analyzed morphology at only one or two of the four scales: macroscopic, mesoscopic, microscopic, and nanoscopic in one study, and no study has examined the full range of all three or four scales simultaneously. However, the epistemological approach, whether based on reductionism or systematics, requires strictly tracing

fouling composition's unit and hierarchical relationships. Therefore, further organic integration of morphology at different study scales is needed to constitute a full-scale morphological theory.

- Development of quantifiable morphology. Although iii. several parameters have been proposed to quantify morphology, such as area ratio and roughness, allowing precise comparisons of morphological differences. However, most of the studies are still based on qualitative descriptions, using words such as porous, fluffy, dense, etc., which are not conducive to quantifying the extent of these differentiations and hamper comparisons between fouling. In addition, some parts of fouling are so complex that qualitative descriptions do not characterize them well, e.g., fluffy or branches fouling at the gasfouling-interface, and clearly describing them is difficult. Therefore, developing operational and quantifiable morphological parameters remains a sub-topic that requires continuous efforts.
  - Reinforce the connection between morphology and the full process of fouling generation mechanism and evolution. Developing the morphology of fouling is important, but more importantly, understanding the mechanism of fouling generation and proposing strategies to mitigate or eliminate it is the ultimate goal. Currently, some studies still focus on the simple comparison of the morphological differentiation of fouling but do not deeply analyze the root causes of these differences, which are detached from the generation mechanism and evolution process. Therefore, the morphology of fouling should be better used to reveal the mechanism of fouling generation, and the evolution of growth, aging, and removal processes should be further strengthened.

#### Conclusions

iv.

The present work provides a comprehensive review of the results of morphological studies of fouling in EGR coolers for the past 25 years, and the following conclusions can be drawn:

i. The evaluation scales are macroscopic, mesoscopic, microscopic, and nanoscopic; the evaluation methods contain visual inspection, optical microscope, digital microscope, surface profiler, scanning electron microscope, and transmission electron microscope, and the evaluation scales are reduced in order. 6 🕢 Y. YAO ET AL.

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- ii. A systematic evaluation system is established, including the evaluation categories of color, glossiness, wetness, softness, densification, roughness, connection method, orderliness etc.
- iii. Defining and categorizing fouling from a morphological point of view, organically integrating full-scale morphology, developing quantifiable morphology, and reinforcing the connection between morphology and the full process of fouling generation mechanism and evolution are four sub-topics that can be further expanded.

## **Disclosure statement**

No potential conflict of interest was reported by the authors.

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tion, targeting applications such as  $CO_2$  capture from flue gases, removal of volatile organic compounds, and elimination of acidic compounds from natural gas and biogas.

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