

Novel Nondestructive Imaging Analysis for Catalyst Washcoat Loading and DPF Soot Distribution Using Terahertz Wave Computed Tomography

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ABSTRACT

This paper describes a three-dimensional (3D) visualization framework using terahertz (THz) waves for quantitative analysis of the distribution of catalyst washcoat loading and diesel particulate filter (DPF) soot deposits. THz waves, which are electromagnetic waves located between infrared and microwave in the electromagnetic spectrum, have good directivity and moderate resolution. Accordingly, a computed tomography (CT) technique has been developed to image the interior of an object using the spectral information of THz waves. This THz wave CT technique can be used to analyze the interior of various target objects, and has good penetrability of ceramic substances used as catalysts in automobiles. Against this background, a method using THz CT for nondestructive analysis of catalyst washcoat loading and the density distribution of soot was developed, and this framework was evaluated. In order to estimate the quantitative measurement capability for nondestructive analysis of catalyst washcoat loading, a DPF sample with a change induced in the density distribution, and with soot that masks the created catalyst washcoat layer, was analyzed using the new approach of THz wave CT technique. In addition, the catalyst washcoat loading distribution was observed using nondestructive 3D visualization. Further, the soot density distribution of the DPF was also visualized and the analysis result satisfied the sample specification that was pre-determined by other methods. These results imply that the 3D imaging analysis method using THz wave CT can be applied to the nondestructive analysis of catalysts and DPF after-treatment systems.

INTRODUCTION

In order to satisfy today's strict emissions standards, diesel vehicles are equipped with automotive catalysts,

and CO and hydrocarbons in the exhaust are oxidized and removed in the presence of oxygen by means of a diesel oxidation catalyst (DOC). In addition, the diesel particulate filter (DPF), which physically traps particles, is a highly effective means of emission control, and their adoption and use is increasing as an after-treatment system component for reducing particulate matter (PM). A typical commercially available DPF can remove over 99% of PM [1]. In addition, an optimized after-treatment system uses the catalyzed DPF (C-DPF) [2], in which DOC material is coated onto the DPF wall, thus enabling oxidation of PM together with filtering.

A major technical subject pertaining to the implementation of on-board DPF systems is the establishment of incineration technology optimized for removal of collected PM (DPF regeneration). The DPF structure enables exhaust filtering and DPF pressure loss increases with the quantity of PM collected, causing increased fuel consumption and difficulty in engine control. One method to remove the PM collected in the DPF is forced regeneration, which burns (=oxidizes) the PM with fuel and oxygen. In that method, the DPF is heated to around 600 °C by the exothermic reaction of fuel at the DOC upper stream, and DPF regeneration is performed [3]. However, an increase of 2~5% in fuel consumption results from this operation [4]. Accordingly, an estimation of accumulated PM quantities and optimized engine control is required for accurate control of the operation. In addition, non-uniform exhaust flow-through the cross-section of the DPF and possible formation of soot deposit can be primary factors in causing large non-uniformities in the density of soot deposited on the DPF while in-use on vehicle. This non-uniformity of soot deposition has been reported to affect the durability of the DPF [5], for instance, by damaging the DPF through excess thermal load, or by increasing thermal stress through heterogeneous combustion. To improve the reliability of after-treatment systems, it is thus necessary to evaluate the soot deposition

process and the distribution of soot deposits in the DPF, as well as to implement appropriate exhaust system design and engine control.

Also, in regard to the catalyst washcoat layer, which may affect back pressure and the distribution of soot loading, the uniformity and amount of washcoat loading are our interests. If the washcoat loading and the washcoat distribution can be analyzed in a non-destructive manner, interpretation of the results and optimization of catalyst design will be facilitated.

To study the above-mentioned subjects and to design emission control systems that work as intended, sample are currently observed by destructive methods that require cutting the DPF brick into slices and performing SEM imaging [6], or by method where the quantity of accumulated PM is measured in separated pieces along the flow direction at intervals of several inches along the DPF brick. As these analyses are time consuming and require destruction of the test object, drawbacks to these existing methods include problems due to the lack of continuity and of reproducibility in measurements. As such, a simple nondestructive analysis technique for determining the soot density distribution and the catalyst washcoat distribution in three dimensions is needed.

In this paper, the analysis system's feasibility and capability are evaluated using THz wave computed tomography (CT) to visualize the catalyst washcoat loading and the density distribution of DPF soot deposition in three dimensions. Up to now, no such analyses have been possible among existing nondestructive inspection measures, and these distribution analysis using THz waves has not been reported.

In this report, attempts to improve the contrast and sensitivity with regards to soot and the catalyst washcoat using THz wave CT are described. Further, the operation of THz wave CT is demonstrated to be a practical nondestructive analysis method that can analyze these distributions.

TERAHERTZ WAVES

As shown in Figure 1, THz waves are located between infrared and microwave in the electromagnetic spectrum [7]. THz waves have features of both radio waves and visible light waves, and can penetrate so-called soft materials such as plastic, ceramics, fats and paper to a certain degree (similar to radio waves), and can be easily manipulated in space using lenses and mirrors (similar to visible light waves). In order to handle THz waves, a technique that differs from existing radio and light wave technologies is necessary. Due to the difficulty of this approach, it has not been proactively applied up to now. However, owing to recent technological advances, extensive research and development of THz wave applications has been carried out in recent years [8], and the effectiveness of these applications has become evident. A notable property of THz waves is that materials have a characteristic absorption spectrum with THz waves, which can be said to be a spectral fingerprint similar to a visible light spectrum. In

addition, in many cases, THz waves can penetrate substances that visible light and infrared cannot due to the absorption of the medium [9]. On the other hand, X-rays are an example of electromagnetic waves that have high penetrability. As the energy of THz waves is lower than that of infrared waves, however, THz waves provide a safe analysis method that does not pose hazards to human health, such as the concerns relating to X-rays. One effective application of THz waves is spectroscopic imaging by which qualities such as the composition, physical phase and quantity can be evaluated by spectroscopic imaging, in addition to spatial structural imaging of the sample. Two-dimensional (2D) imaging of concealed illegal drugs using THz waves has been reported, and the application of THz waves is anticipated as a new nondestructive analysis method [10].

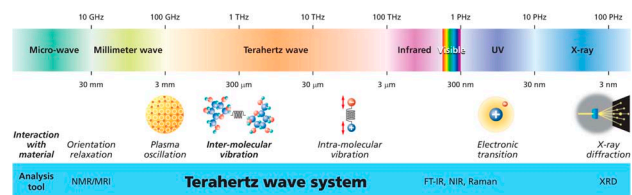


Fig. 1 Frequency and wavelength spectra of electromagnetic waves

THZ WAVE CT

As THz waves have the qualities of penetrability, directivity and practical spatial resolution, they can be used to create a three-dimensional (3D) image. X-ray CT is well-known as a nondestructive dislocation detection method for medical diagnosis. CT is an analysis technique whereby an inverse radon transform is performed on the projection data acquired by measurement, and a reconstructed cross-sectional image is created [11][12]. Ferguson et al. reported one of the first cases of CT measurements using THz waves [13]. In another study, a characteristic of THz wave CT was reported to be that both analysis of the density distribution and 3D imaging of the composition distribution of a specific component in materials is possible [14]. Compared with X-ray CT, which does not possess spectral information, the 3D imaging of the composition distribution is a notable characteristic of THz wave CT. Therefore, the application of THz wave CT to nondestructive analysis of various targets is anticipated. The inner material of ceramics is one potential analysis object for density distribution analysis. Many DPF and catalyst substrates consist of nonconductive ceramic material such as cordierite (Cd), aluminum titanate (AT) or mullite, and as they are highly permeable to THz waves [15], the nondestructive analysis of the inside of the substrate by the CT method is possible.

“TAS7000 3D IMAGING ANALYSIS SYSTEM”

The “TAS7000 3D imaging analysis system” (Advantest, Japan) is being commercialized as a THz wave CT measurement system. Figure 2 shows a

block diagram of the system's internal units and a photograph of its external appearance.

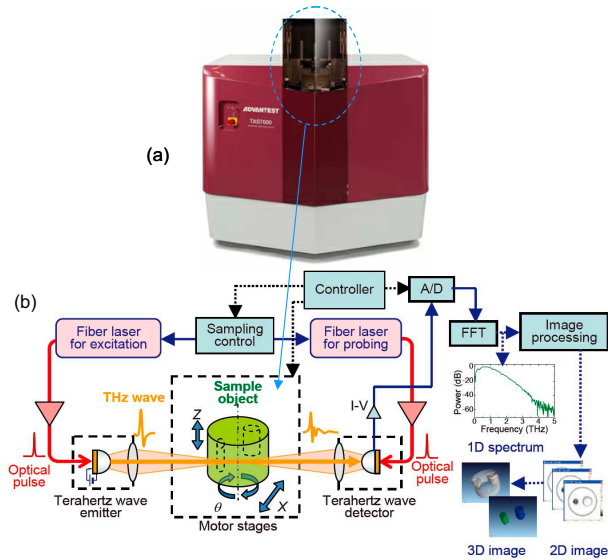


Fig. 2 TAS7000 3D imaging analysis system
(a) external appearance and (b) schematic diagram

The principal feature of the analysis system is that THz wave emitters and detectors are placed on both sides of the stage, and the THz waves that penetrate the sample are detected. To generate THz waves, a sudden change in electric field is produced by driving a photoconductive switch using pulsed light generated by an 80-fs ultra-short pulse optical fiber laser. This generates THz waves with a wide spectral composition in a pulse. As the detection side has almost the same configuration, the variation in electric field of the THz waves at the photoconductive switch is sampled as the time waveform by controlling the pulse timing of the fiber laser. The time waveform is Fourier transformed in order to perform spectral analysis. The sample is spatially scanned in the X(radius)-, θ (rotation)- and Z(height) -directions by use of a motor stage, and at this time, transmission parameters that have a value for each frequency are measured. When the measured values of the scanned points are plotted on the X- θ plane as a cross section that intersects the Z-axis, projection data (a sinogram) are acquired. A slice image of the transmission parameters transformed at the unit transmission length is reconstructed by applying a filtered back projection algorithm as a reverse radon transform of this sinogram. By performing spatial scanning at another Z-coordinate, 3D tomographic information is acquired.

3D DISTRIBUTION ANALYSIS

When THz waves propagate, they interact with the substances in the transmission space, giving rise to an optical absorption spectrum in the THz region. As the strength of the interaction differs according to the substance, the optimum THz wave transmission parameters are selected for each substance in the

target object to be analyzed. There is a correlation between the quantity of a substance and the strength of its interaction with the THz waves [16][17]. Thus, the density distribution of the target substance can be measured in three dimensions by constructing a calibration curve for the 3D tomographic qualities measured. Figure 3 shows reference values and the method of determining fixed quantity coefficients.

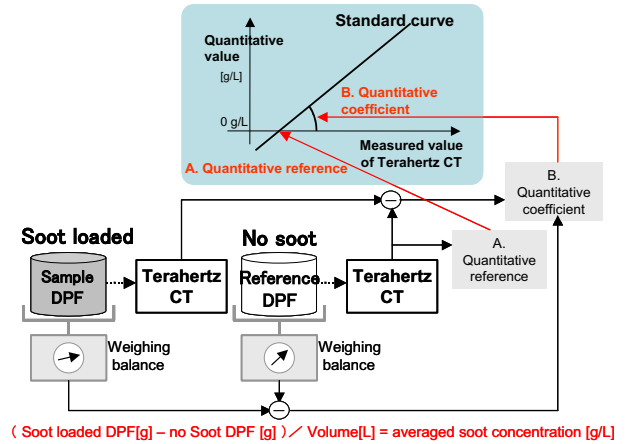


Fig. 3 Quantitative analysis procedure for THz CT

The coefficient of the calibration curve is fitted by comparison with the average measured by THz wave CT and a soot density determined by the gravimetric method. For instance, if the target substance is soot, the total amount of soot in the substrate is found from the difference in weight of the DPF before and after soot loading, and the expected value of average soot density is acquired by dividing the total amount of soot by the substrate's volume. It is also possible to use the specification values of the sample as expected values. Measurements are then carried out by THz wave CT, and the measured values of the section are taken as the reference values of the calibration curve such that the density for the substrate prior to soot loading becomes 0 g/L. Then, for the substrate loaded with soot, the average value of the difference between the THz transmission parameter and the reference value is calculated. A gradient (coefficient) of the calibration curve is determined as the ratio of the average and the expected value. The calibration curve is then applied to all 3D tomographic data. Similarly, a calibration curve is constructed for the catalyst washcoat loading distribution, also by using THz waves to measure the expected values before and after catalyst coating, and then the analysis is performed.

EXPERIMENTAL SETUP

The experiment was performed using the aforementioned “TAS7000 3D Imaging Analysis System” shown in Figure 2. The samples used in the experiments were prepared according to the specifications described in the next section. A sample was fixed onto the system’s micromanipulator stage, and the 3D