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Phase-controlled fractional derivatives for near infrared spectral processing

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This article is dedicated to Prof T Asakura

As an extension of fractional derivatives, phase-controlled fractional derivative is proposed by modifying the phase factor of the filter function in the definition of the fractional differentiation in view of the application to the spectral data analysis in near infrared spectroscopy. This process can control the degree of enhancing tiny peaks and the degree of peak shift independently and continuously. It is therefore possible to obtain, for example, a derivative spectrum having peak shift similar to the first derivative and peak thinning and enhancement similar to the second derivative, which gives better peak separation than conventional and fractional derivatives do. Properties of fractional derivatives and fractional absolute derivatives are also revisited as a base of introducing the phase-controlled fractional derivatives. © Anita Publications. All rights reserved.

Keywords: Fractional derivative, Derivative spectroscopy, Near infrared spectroscopy, Peak enhancement

1 Introduction

An idea of the derivative with a fractional order dates back to Leibniz's note to de l'Hôpital in 1695, in which the meaning of the derivative of order $1/2$ was discussed [1]. Since then, theory of extending the order of calculus including integration to general real number has been developed by famous mathematicians such as Euler, Laplace, Fourier, Abel, Liouville, and Riemann, and several definitions have been given to fractional derivatives and integrals. Among them, the Riemann–Liouville definition is the most well-known and applied to various physical and technological problems in recent years [2,3].

In spectroscopic data analysis, differentiation is one of the fundamental and frequently used pretreatments, and this is also the case with the near infrared (NIR) spectroscopy, which has been developed in recent years as a powerful tool for estimating the concentrations of chemical components and properties in complex materials such as agricultural commodities, foods, pharmaceuticals, chemical products, living bodies and so on [4]. Absorbance spectra in NIR region of such materials consist of overtones and combinations of fundamental absorptions appearing in the mid-infrared region, and hence are very complicated. Therefore, enhancement of tiny peaks and separation of overlapped peaks by differentiation of spectra provide considerable improvement in the performance of NIR estimation of target components and properties of various complex materials. Since such benefits obtained by ordinary derivatives are rather limited practically to the first and second orders, applications of fractional derivatives (FDs) to remove this limitation and to enhance the flexibility of the operation were reported by several researchers [5–10].

In ordinary and fractional derivatives of a single peak, peak position in the derivative shifts to the left with an increase of order, and the peak is inverted in derivatives of even orders. Since the shift of a peak in the ordinary and fractional derivatives make it somewhat difficult to identify the wavelength of an absorption band from the derivative spectra, a modification of FD to suppress the peak shift and inversion in any value of the derivative order has been devised, and is called fractional absolute derivative (FAD).

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It is also noted, however, that the shift of peaks sometimes plays an effective role in separation of overlapped peaks because the amount of the shift is proportional to the width of the peak. Therefore, the first derivative having the largest peak shift may provide a better calibration performance than the second derivative in some cases. On the other hand, when an absorption band that is a key to a target constituent is a very tiny one, higher derivatives than the first one may be suitable for gaining sufficient enhancement.

Therefore, it would be attractive to define a new type of the differential operation that can control the shift and enhancement of peaks separately. In this paper, this is shown to be possible by modifying phase factor of the filter function in the Fourier transform domain and thus by introducing phase-controlled fractional derivative (PCFD) as a further modification of the FD.

2 Theory

There are already some reports discussing applications of the fractional derivative and its modified version, the fractional absolute derivative, to spectroscopy. However, the definitions and properties of those derivatives are revisited in this section as an introduction to the new type of derivative proposed in the present paper. Most of the past reports introduced the fractional derivatives as an extension of the Fourier transform representation of the derivative spectra. In the present paper, therefore, their relation to the Riemann-Liouville integral is made clear.

2.1 Fractional derivative

Let a function $f(x)$ be differentiable in the range of order we concern and consider its fractional derivative (FD) of order ν given by the Riemann-Liouville definition,

$${}_a D_x^\nu f(x) = \frac{d^k}{dx^k} {}_a D_x^{-\alpha} f(x) = \frac{1}{\Gamma(\alpha)} \frac{d^k}{dx^k} \int_a^x (x-x')^{\alpha-1} f(x') dx', \quad (1)$$

where ${}_a D_x^\nu$ is a fractional integro-differential operator, ν is a non-negative real number, $k = \nu + \alpha$ is an integer with $0 < \alpha < 1$, a is a lower limit of the integral and $\Gamma(\cdot)$ is the gamma function [1]. We consider the case of $a \rightarrow -\infty$ and, with the notation of $D_x^\nu = {}_{-\infty} D_x^\nu$ for simplicity, we have

$$D_x^{-\alpha} f(x) = \frac{1}{\Gamma(\alpha)} \int_{-\infty}^x (x-x')^{\alpha-1} f(x') dx' = \frac{1}{\Gamma(\alpha)} \int_0^\infty (x')^{\alpha-1} f(x-x') dx', \quad (2)$$

Therefore, the last integral in Eq (2) is the convolution of $f(x)$ and

$$h(x) = \begin{cases} x^{\alpha-1}/\Gamma(\alpha); & x \geq 0, \\ 0 & ; \quad x < 0. \end{cases} \quad (3)$$

Due to the convolution theorem of the Fourier analysis, the Fourier transform of Eq (2) is given by the product of the Fourier transforms of these two functions,

$$F(\xi) = \mathcal{F}[f(x)] = \int_{-\infty}^\infty f(x) \exp(-i2\pi x\xi) dx, \quad (4)$$

$$H(\xi) = \mathcal{F}[h(x)] = \int_{-\infty}^\infty h(x) \exp(-i2\pi x\xi) dx = \frac{1}{\Gamma(\alpha)} \int_0^\infty x^{\alpha-1} \exp(-i2\pi x\xi) dx, \quad (5)$$

where \mathcal{F} is the Fourier transform operation. Since the last integral in Eq (5) is evaluated to be [11]

$$\int_0^\infty x^{\alpha-1} \exp(-i2\pi x\xi) dx = \Gamma(\alpha) (i2\pi\xi)^{-\alpha}, \quad (6)$$

Equation (2) can be expressed in the inverse Fourier transform representation as

$$D_x^{-\alpha} f(x) = \mathcal{F}^{-1}[(i2\pi\xi)^{-\alpha} F(\xi)] = \int_{-\infty}^\infty (i2\pi\xi)^{-\alpha} F(\xi) \exp(i2\pi x\xi) d\xi, \quad (7)$$

where \mathcal{F}^{-1} stands for the inverse Fourier transform operation. Since

$$f(x) = \int_{-\infty}^\infty F(\xi) \exp(i2\pi x\xi) d\xi, \quad (8)$$

k -th derivative of $f(x)$ can be expressed by differentiating Eq (8) k times as

$$\frac{d^k}{dx^k} f(x) = \frac{d^k}{dx^k} \int_{-\infty}^{\infty} F(\zeta) \exp(-i2\pi x \zeta) d\zeta = \int_{-\infty}^{\infty} (i2\pi \zeta)^k F(\zeta) \exp(-i2\pi x \zeta) d\zeta, \quad (9)$$

Hence, Eq (1) with $a \rightarrow -\infty$ can be expressed as

$$D_x^\nu f(x) = \frac{d^k}{dx^k} \int_{-\infty}^{\infty} (i2\pi \zeta)^{-\alpha} F(\zeta) \exp(i2\pi x \zeta) d\zeta = \int_{-\infty}^{\infty} (i2\pi \zeta)^\nu F(\zeta) \exp(i2\pi x \zeta) d\zeta. \quad (10)$$

Therefore, Eq (10) is a natural generalization of Eq (9) with integer k replaced by real number ν , and this is sometimes regarded to be another way of defining FD.

In the present paper, numerical calculation of fractional derivatives and related derivatives to be introduced later were carried out using Fourier transform representations such as Eq (10) with programs written in MATLAB. It is noted that calculation of FD by means of a polynomial fitting approach has also been proposed [12–14].

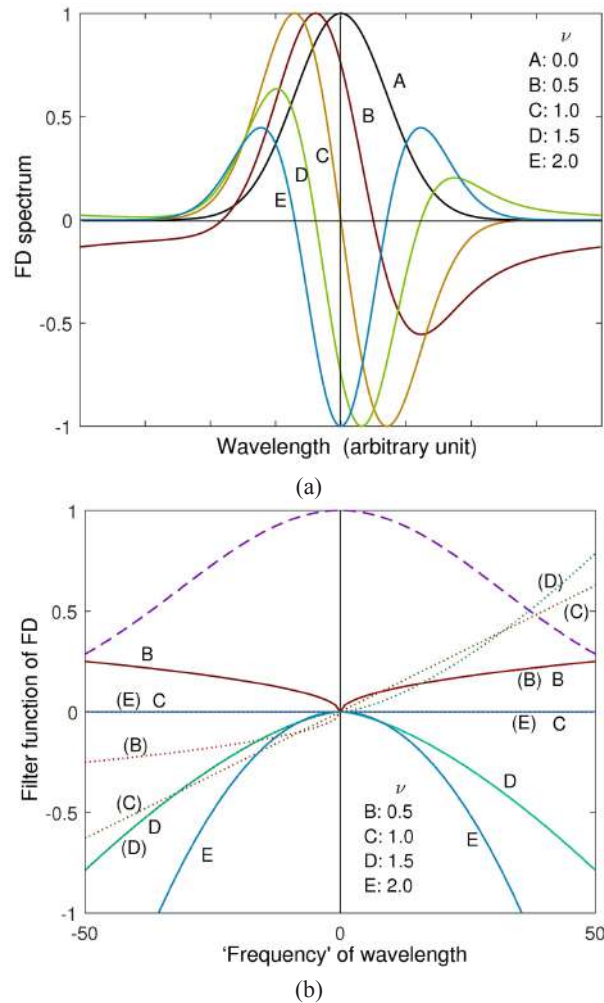


Fig 1 (a) FDs of a single Gaussian peak for every 0.5 order in $0 \leq \nu \leq 2.0$ and (b) the filter function of FD. In (b), solid lines B–E are the real parts of filter functions and dotted lines (B)–(E) stand for their imaginary parts.

In spectroscopic data analysis, differentiation of spectra provides important effects such as enhancement of small and weak peaks, separation of complex peaks, removal of a bias (first or higher derivatives) and a linear trend (second or higher derivatives). To see how fractional derivatives of a single peak look like, results of the operation of Eq (10) for a Gaussian function are shown in Fig 1(a) for every 0.5 order from $\nu = 0.0$ to 2.0. In this figure, derivatives are normalized so that their absolute maximum values are unity. As is shown in Fig 1(a), the peak position in the FD shifts to the left with an increase of order from zero, and the position of a trough appearing from the right end also shifts to the left. These peak and trough form an antisymmetric shape in the first derivative and the trough grows further to become an inverted peak in the second derivative. It is also seen that the width of peaks in derivatives decreases gradually as the order increases.

It is seen from Eq (10) that the fractional derivative is given by a filtering operation in the frequency domain with the filter function

$$T(\xi) = (i2\pi\xi)^\nu, \quad (11)$$

which is shown in Fig 1(b) for $0.5 \leq \nu \leq 2.0$. As is seen from this figure, the magnitude of the filter function rises for higher frequencies, and its degree becomes higher with an increase in ν , though the real and imaginary parts behave in a complicated way as ν changes. This behavior of filter function gives rise to the effects of enhancing small peaks and separating complex peaks. The property of higher frequency enhancement also increases undesirable high frequency noises in practical spectral data analysis. To avoid this, another filter function is used for suppressing very high frequency components while keeping low and moderate frequency components almost intact. As a typical function having such a property, a Gaussian filter function is sometimes used and is also shown in Fig 1(b) by a dashed line. The width of the Gaussian function is to be adjusted depending on the case.

It is noted from Eq (11) and Fig 1(b) that $T(0) = 0$ for $\nu > 0$ and therefore a bias or constant in an original function is removed in the derivative of any positive order. This is why the curve B for $\nu = 0.5$ in Fig 1(a) is shifted down below the baseline.

2.2 Fractional absolute derivative

Fractional derivatives provide us with a flexibility of adjusting the degree of peak enhancement and decomposition of complex peaks by choosing an adequate derivative order in a continuous range of real number. However, the shift of a peak in the ordinary and fractional derivatives make it sometimes difficult to identify the wavelength of an absorption band from the enhanced or separated peak. For such purposes, therefore, it is desirable to suppress the peak shift and inversion in derivative spectra. This is possible by employing a new filter function

$$T_a(\xi) = (2\pi|\xi|)^\nu, \quad (12)$$

which is the modulus of the filter function of Eq (11). Thus a new type of derivative

$$D_{|x|}^\nu f(x) = \mathcal{F}^{-1} [(2\pi|\xi|)^\nu F(\xi)] = \int_{-\infty}^{\infty} (2\pi|\xi|)^\nu F(\xi) \exp(i2\pi x\xi) d\xi \quad (13)$$

is introduced, which may be called fractional absolute derivative (FAD). The symbol $D_{|x|}^\nu$ indicates that the derivative does not depend on whether the function is differentiated in the positive or negative direction of x . The results of FAD of a single Gaussian peak and the filter function are shown in Figs 2(a) and (b), respectively. Figure 2(a) shows that the peak shift and inversion are completely suppressed and that the width of the derivative becomes narrower as the order increases more clearly than the case of FD in Fig 1(a). The filter function in Fig 2(b) also shows that the magnitude of the filter function is monotonically increases with the frequency and ν more clearly than the case of FD shown in Fig 1(b).

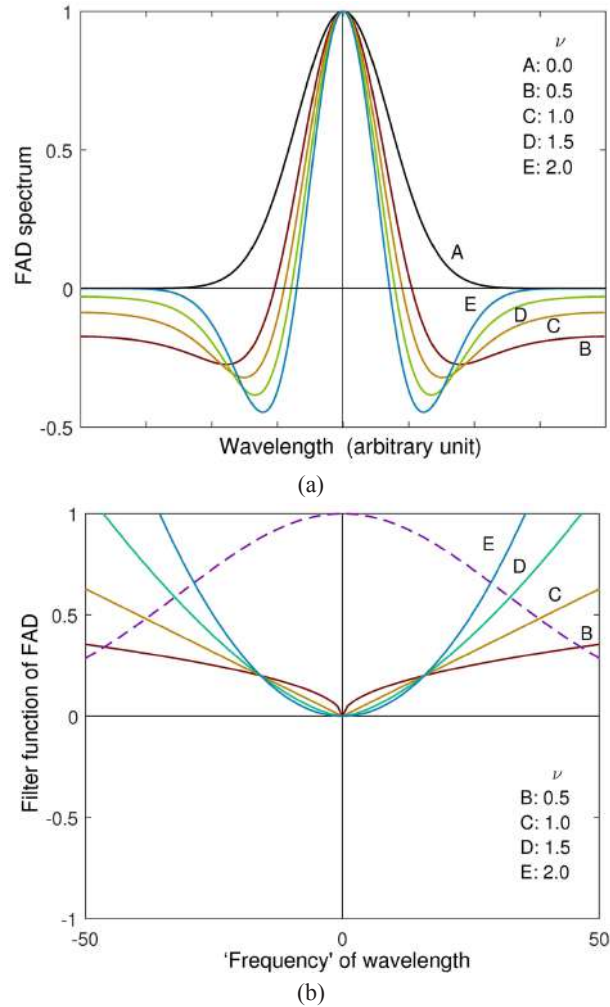


Fig 2. (a) FADs of a single Gaussian peak for every 0.5 order in $0 \leq \nu \leq 2.0$ and (b) the filter function of FAD.

2.3 Phase-controlled fractional derivative

In FAD, the peak shift and inversion are completely suppressed by removing the phase factor in the filter function. However, it would be also attractive to specify the degree of the peak shift independently from the derivative order. To this end, we investigate the filter function in more detail. The filter function of FD in Eq (11) can be written as

$$T(\zeta) = \tau^\nu(\zeta) = (i2\pi\zeta)^\nu, \quad (14)$$

where $\tau(\zeta) = i2\pi\zeta$ is a complex function and can be expressed by

$$\tau(\zeta) = a(\zeta) e^{ib(\zeta)}, \quad (15)$$

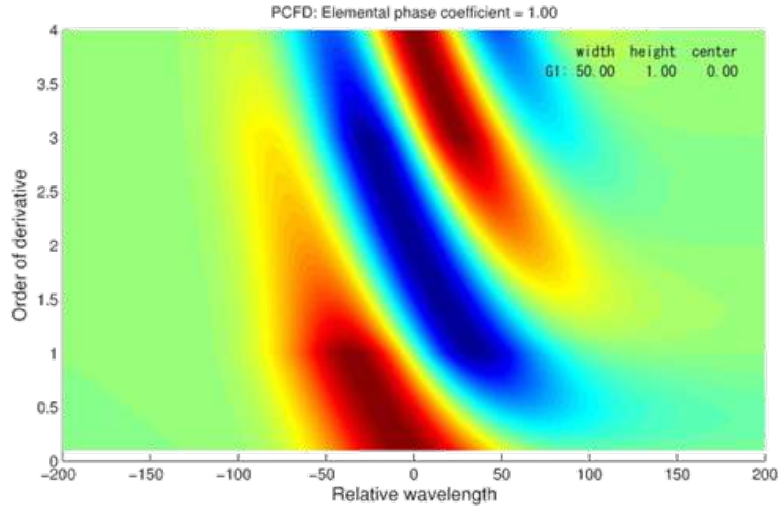
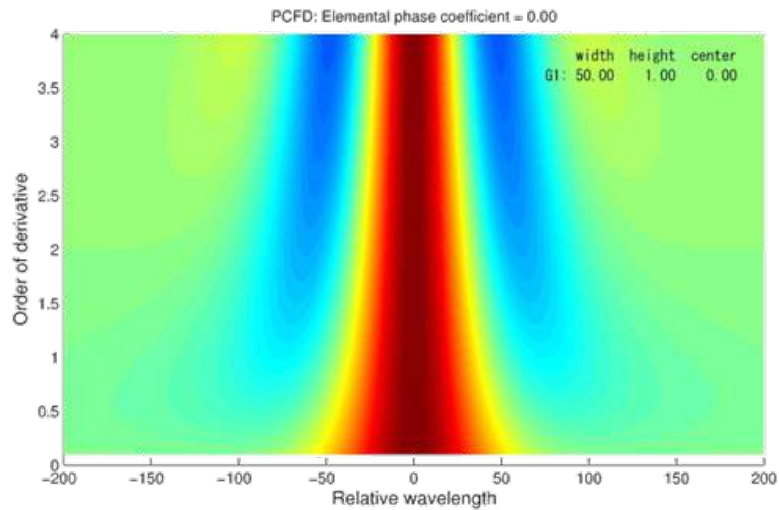
in which $a(\zeta)$ and $b(\zeta)$ are the modulus and phase of $\tau(\zeta)$, respectively.

Since the phase in a Fourier spectrum is strongly related to the lateral shift of its original function, the filter function $T_a(\zeta)$ of FAD was defined by removing the phase $b(\zeta)$ in Eq (15). Now let us introduce a parameter c to modify the phase factor instead of completely removing it;

$$\tau_p(\zeta, c) = a(\zeta) e^{icb(\zeta)}, \quad (16)$$

$$T_p(\xi, c) = \tau_p^\nu(\xi, c) = a^\nu(\xi) e^{icvb(\xi)} = a^\nu(\xi) e^{i\gamma b(\xi)}, \quad (17)$$

where $\gamma = cv$. The parameters c and γ may be called elemental and total phase coefficients, respectively. By selecting an appropriate value for c or γ , it is possible to control the degree of peak shift in the resultant derivative spectrum. This new type of derivative may be called phase-controlled fractional derivative (PCFD). PCFD reduces to FD when $c = 1$, while it becomes FAD when $c = \gamma = 0$, since $T_p(\xi, 1) = \tau^\nu(\xi)$ and $T_p(\xi, 0) = |\tau(\xi)|^\nu$. FCFDs of a single Gaussian peak for these two special cases are shown in Fig 3 for $0 < \nu \leq 4.0$. Namely, Fig 3(a) is the case for $c=1$ corresponding to the FD, and is a continuous version of Fig 1(a), while Fig 3(b) is the case for $c=0$ being equal to the FAD and is a continuous version of Fig 2(a). In both plots, the functions are normalized so that the absolute maximum for every ν is unity, and the colors are mapped so that the deepest blue and red correspond to the values -1 and 1 of the function, respectively.

(a) FD (PCFD for $c = 1$)(b) FAD (PCFD for $c = 0$)Fig 3. (a) FD and (b) FAD of a single Gaussian function for $0 < \nu \leq 4$

The dependence of PCFD of the Gaussian peak on γ is shown in Fig 4 for $\nu =$ (a) 1 and (b) 2. It is shown in Fig 4(a) that the peak of the first derivative of a Gaussian function is located in the center when $\gamma = 0$, moves to the left as γ increases and becomes the ordinary first derivative at $\gamma = 1$. With a further increase in γ , its trough deepens, moves to the left and forms a shape similar to the ordinary second derivative when $\gamma = 2$, which is exactly an inverted version of that in the case of $\gamma = 0$. It follows that coefficient γ controls the locations of peak and trough of the first derivative and that their widths are determined separately by the derivative order ν . Figure 4(b) shows that, when $\nu = 2$, the derivative behaves like that in Fig 4(a) with the width narrower than that. Namely, the PCFD is as narrow as the second derivative and as shifted as the first derivative when $\gamma = 1$ and $\nu = 2$.

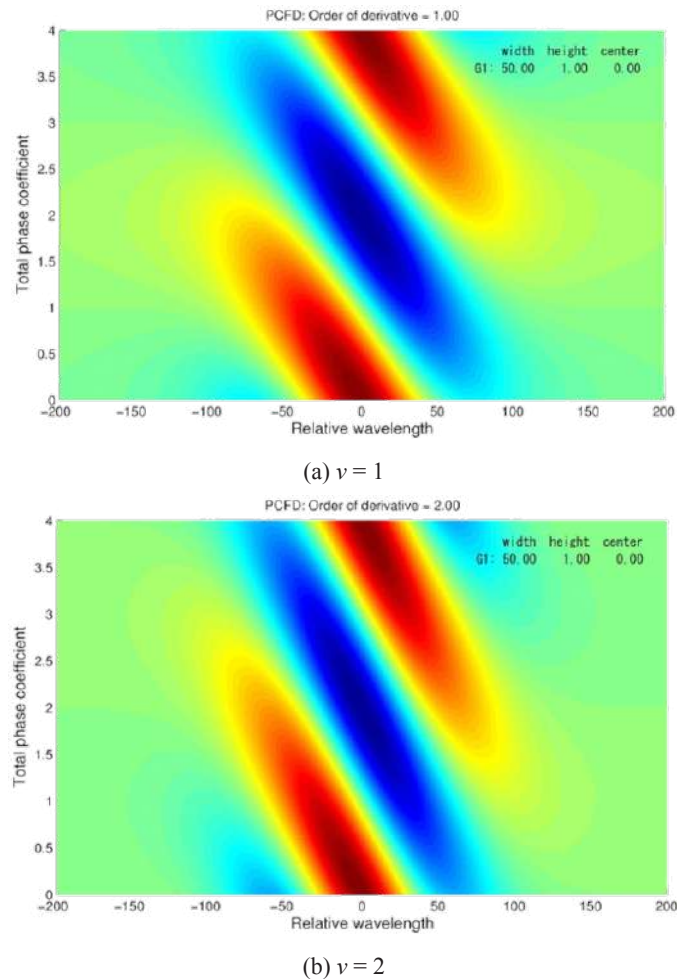


Fig 4. PCFD of a single Gaussian function for $0 \leq \gamma \leq 4$

A typical case where PCFD may be effective is shown in Fig 5. This figure illustrates the dependence of PCFD on ν of a linear combination of three Gaussian peaks having (width, height) = (80, 10), (20, 1) and (5, 0.1) with all the peaks centered on the origin, and Figs 5(a) and 5(b) are for $c = 1$ and $\gamma = 1$, respectively. Figure 5(a) corresponds to ordinary FDs, showing that the three peaks are barely separated by the ordinary fractional derivative in the region of $\nu \gtrsim 2.3$ on the right side of the original position. This is because the amount of the peak shift depends on the width of the peak as well as ν , and there are large differences in the

peak shifts of three Gaussian functions. In Fig 5(a), the narrowing of peaks is insufficient for $\nu \approx 1$, while there is no peak shift around $\nu \approx 2$ inspite of fairly good narrowing. In Fig 5(b), however, three peaks are separated appreciably for $1.5 \lesssim \nu$, because the PCFD for $\gamma = 1$ is forced to have large peak shift regardless of ν , and therefore the effect of increased narrowing with ν contributes directly to the peak separation.

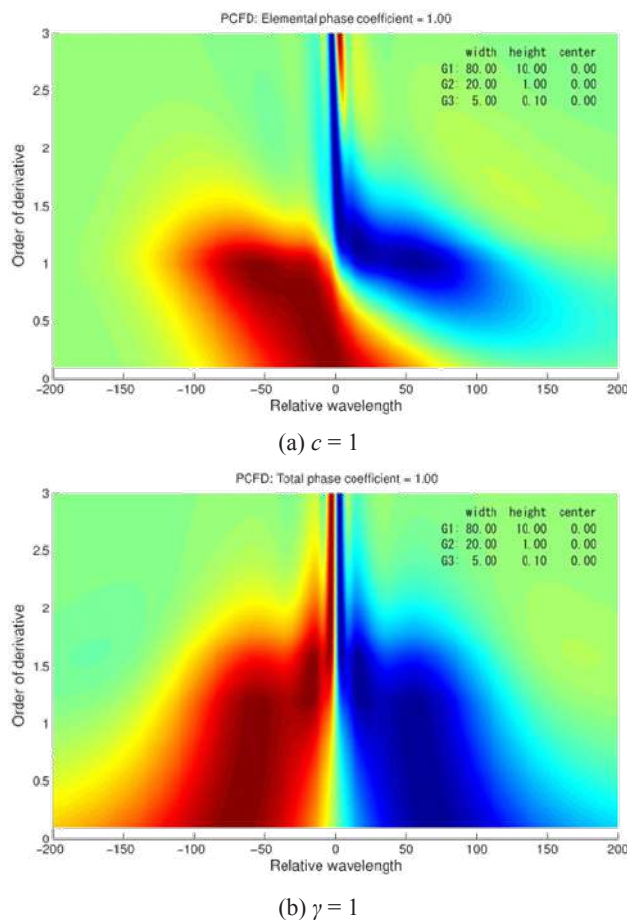


Fig 5. PCFD of three overlapped Gaussian peaks.

For the name of the derivative defined by the filter function of Eq (17), ‘phase-modified’ derivative may be more suitable than ‘phase-controlled’ derivative given above. However, the author would like to keep the current name, expecting the possibility of controlling the elemental or total phase coefficient with an external factor to adjust the peak shift automatically in the future.

The term ‘fractional derivative’ has already been recognized. However, it may need discussion in using the term ‘derivative’ in FAD and PCFD. Some of the original properties, such as a slope of the line tangent to the function, of derivatives are lost in FAD and PCFD. However, the same already holds for the FD of a truly fractional order. In this sense, PCFD is similar to FD as it includes conventional derivatives as its special cases. The important property common to the conventional, fractional, absolute and phase-controlled fractional derivatives is that the modulus of their filter functions in Fourier domain is the same scaling function, which has the property of enhancing higher frequency components in a scaling manner. This may justifies the use of the term ‘derivative’ for all of them.

3 Application to NIR spectra

3.1 FD and FAD spectra

To examine the properties and behaviors of FD, FAD and PCFD discussed above in a practical situation, some derivatives are calculated as a function of λ and ν for absorbance spectrum of rice flour in NIR region of $1100 \leq \lambda \leq 2500$ nm, where λ is the wavelength and is used instead of x in Eq (1). Figures 6(a) and (b) show the FD and FAD, respectively, for every 0.5 order in $0 \leq \nu \leq 2.0$. In the calculation of these derivatives, a low pass Gaussian function such as shown in Figs 1(b) and 2(b) was used to suppress high frequency noises. Characteristics of the FD and FAD are evident in these figures in accordance with the features seen for single Gaussian function in Figs 1(a) and 2(a). For the FD, as ν increases, absorption peaks are shifted to the left and small peaks are enhanced and separated gradually. In the case of FAD shown in Fig 6(b), however, absorption peaks remain almost unshifted, while other features of enhancement of small peaks and separation of complex peaks are present.

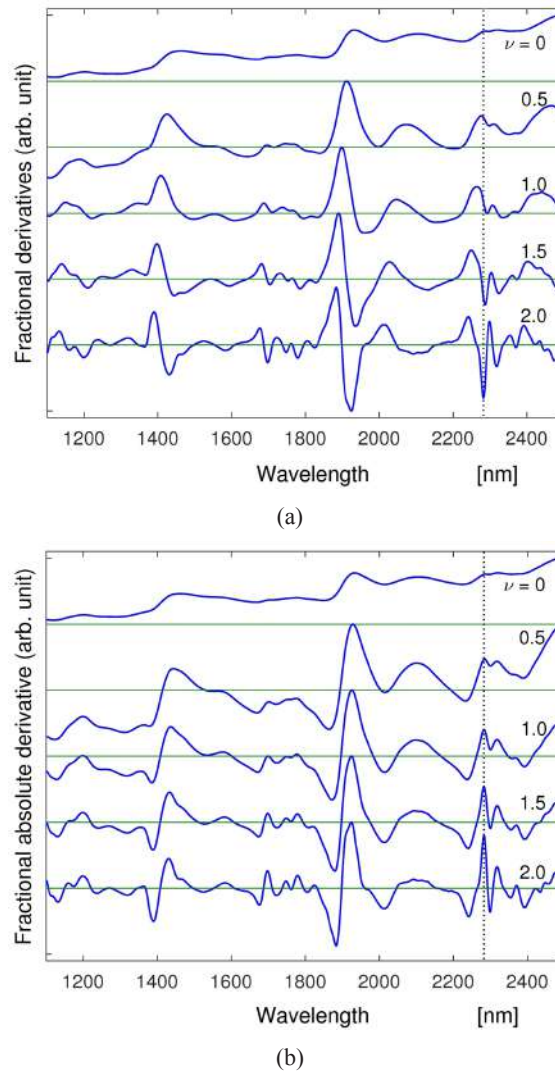


Fig 6. (a) FDs and (b) FADs of an NIR absorbance spectrum of rice flour for $0 \leq \nu \leq 2.0$.

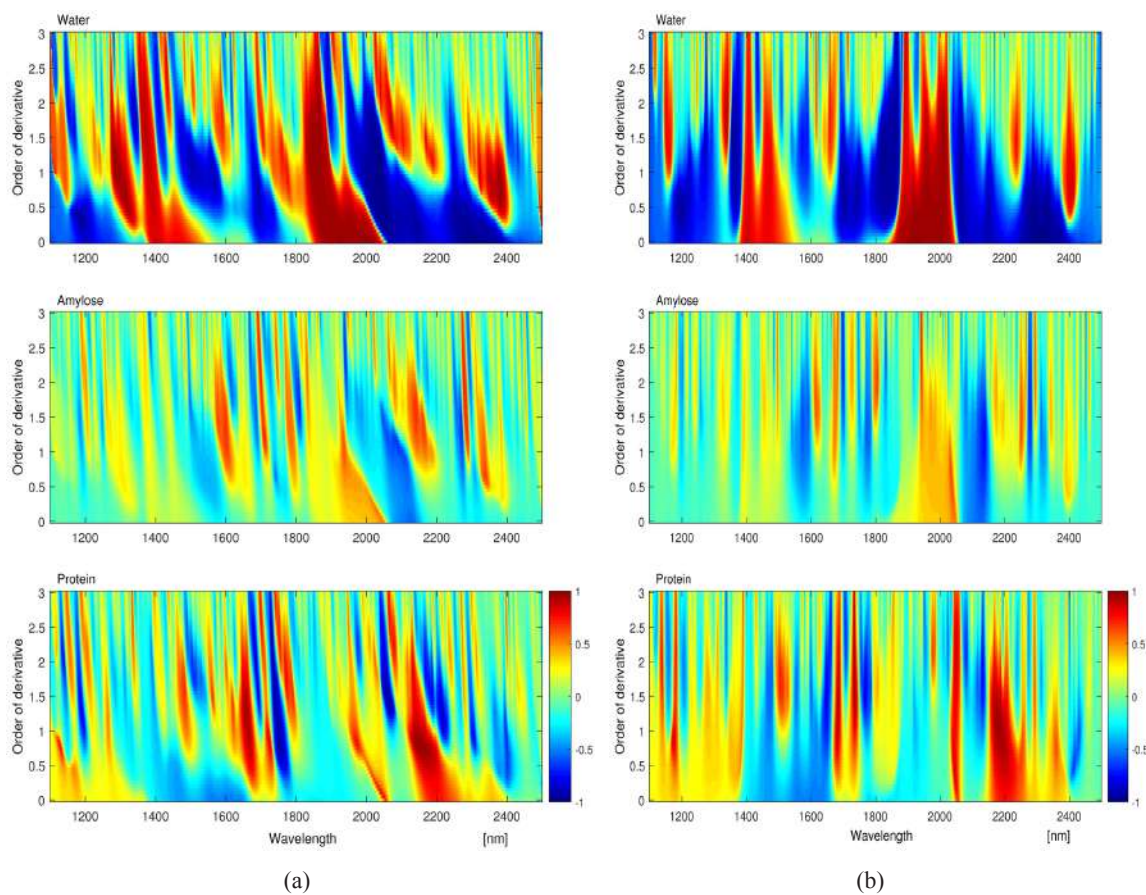


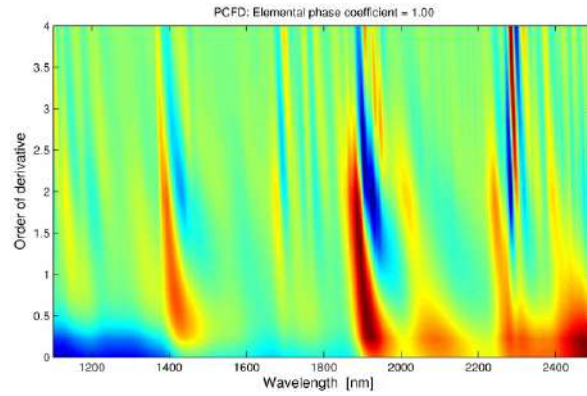
Fig 7. Correlation coefficients of water, amylose and protein concentrations with (a) FD and (b) FAD.

The difference in the behavior of peaks between the FD and FAD is most visible in the oil absorption band around 2300 nm, where broken lines are drawn for reference. It is seen from this figure that the FAD may be more suitable for analyzing peak structures of spectra than FD. It is again noted that, in the second derivatives, the FD is an inverted version of the FAD, but in the first derivatives, the FAD is quite different from the corresponding FD. While fine details of derivative spectra are clearly visible in Fig 6, the tendency of the continuous change of FD and FAD spectra with the change of the order is seen in Fig 8(a) and (b) as discussed later.

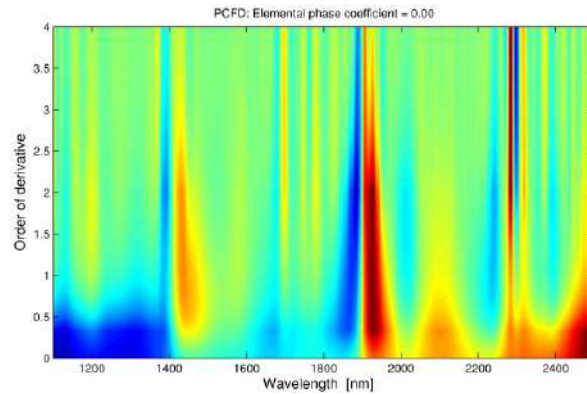
3.2 Correlations of FD and FAD with constituents of rice

NIR spectra measured at every two nanometers in $1100 \leq \lambda \leq 2500\text{nm}$ for 31 rice flour samples were first processed with the pretreatment of the multiplicative scatter correction [4] and their FDs and FADs were subsequently calculated for $0.0 \leq \nu \leq 3.0$. Then, for three major constituents, water, amylose and protein, obtained by conventional chemical procedures, correlation coefficients with each of the FD and FAD derivative spectra were calculated. The results are shown in Figs 7(a) and (b) for FD and FAD, respectively. It is seen from Fig 7(a) that the rate of the peak shift in the correlation coefficient calculated from the FD is fast for smaller ν and becomes slow for larger ν . This may be explained as follows. For small ν , wider peaks are dominant and exhibit larger shift, while for larger ν wide peaks are suppressed and/or decomposed into small ones, which give rise to smaller shift. In Fig 7(b), it is more clearly seen that, in general, each

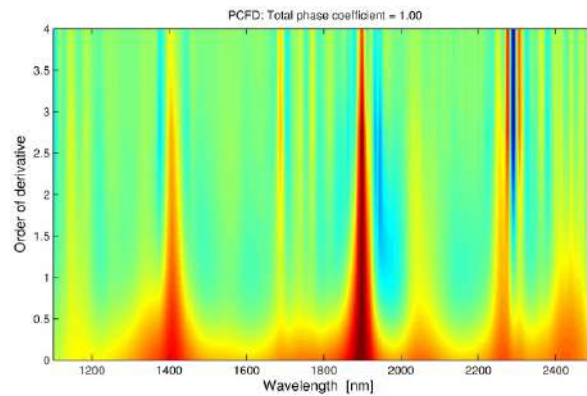
correlation peak is getting narrower and splits into many fine structures. It is also noted, that, there are some correlation peaks having maxima at some intermediate values of ν depending on their wavelengths, both for the FD and the FAD. This implies that the FD or FAD spectrum of a certain fractional order may give better calibration for the estimation of these constituents as compared with conventional derivatives of integral orders.



(a) $c = 1$ (FD)



(b) $c = 0$ (FAD)



(c) $\gamma = 1$

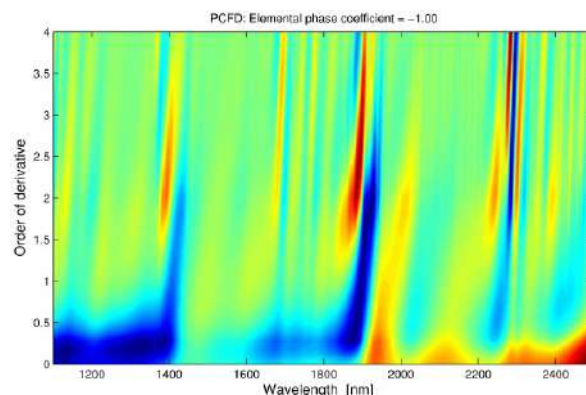
(d) $c = -1$

Fig 8. PCFD of an NIR spectrum of rice flour in four different cases.

Performance of NIR calibration constructed using FD and FAD for estimating water, amylose and protein concentrations in rice flour has been examined as a preliminary study and a slightly better result was actually reported as compared with ordinary integral derivatives spectra.[8,9].To investigate the interrelation between different wavelengths with respect to the correlation with the constituent concentrations, two-dimensional correlation analysis was also reported [15].

3.3 PCFD spectra

To examine the effect of PCFD in NIR spectra of rice flour, derivatives are calculated for four different cases of (a) $c = 1$, (b) $c = 0$, (c) $\gamma = 1$ and (d) $c = -1$, and the results are shown in Figs 8(a)–(d), respectively. It is noted that a Gaussian low-pass filter with an adequate width is again inserted in the integrand of inverse Fourier transform calculating PCFD to suppress noises. In Fig 8, the cases of (a) $c = 1$ and (b) $c = 0$ correspond to FD and FAD, respectively, and are continuous version of Figs 6(a) and (b), respectively. In Fig 8(c), the peak shift is forced to be of the degree of the first derivative, and therefore there may be some cases similar to Fig 5(b), giving rise to better separation contributing to improved performance of NIR spectroscopy. Figure 8(d) of $c = -1$ corresponds to the FD obtained by differentiation in negative direction of the wavelength and, hence, the peak shift is made directed to the right of the original positions. This extraordinary case may also give rise to better separation of overlapped peaks than the case of ordinary FD in some circumstances. However, suitable values of the parameters, ν , c and γ would depend on materials, constituents, vibrational modes and wavelengths.

4 Conclusion

The fractional derivative (FD) was revisited on the basis of the Riemann–Liouville definition, and the fractional absolute derivative (FAD) was defined as a modification to FD. The behaviors and properties of FD and FAD were discussed for a Gaussian function as a model of a single spectral peak. Phase-controlled fractional derivative (PCDF) was introduced by modifying the phase factor of the filter function and by defining new parameters of elemental and total phase coefficients, c and γ , respectively, in the definition of the fractional derivative. This process can control the degree of enhancing tiny peaks and the degree of the shift of peaks and troughs independently and continuously, and therefore can realize effective separation of overlapped peaks and good enhancement of small peaks at once. Behaviors of this new type of derivative were examined by employing a single and overlapped Gaussian functions. It was shown that the shift of peaks in PCFDs of a single Gaussian function can be controlled by c or γ effectively. Also shown was that

the derivative peaks of overlapped Gaussian peaks stay largely shifted and separated with $\gamma = 1$ for any derivative order larger than a certain minimum value needed to narrow component peaks. An NIR spectrum of rice flour was examined for the behavior of FD, FAD and PCFD. The derivatives of the spectrum show quite different appearances depending upon the parameters c and γ . The correlation properties of the PCFDs and constituent concentrations as well as calibration performance will be discussed in the future study.

The present study has been partly presented in conferences and published in their proceedings [8-10]. The author would like to express acknowledgement to Mr. Hideyuki Ogasawara and Mr. Masahiko Murata for their cooperation with respect to the present study.

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