Generalized hyperbolic fractional equation for transient-wave propagation in layered rigid-frame porous materials

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This paper provides a temporal model for the propagation of transient ultrasonic waves in a layered isotropic porous material having a rigid frame. A temporal equivalent fluid model is considered, in which the acoustic wave propagates only in the fluid saturating the material. In this model, the inertial effects are described by the layered tortuosity and the viscous and thermal losses of the medium are described by two layered susceptibility kernels which depend on the viscous and thermal characteristic lengths. The medium is one dimensional and its physical parameters (porosity, tortuosity, and characteristics lengths) are depth dependent. A generalized hyperbolic fractional equation for transient sound wave propagation in layered material is established.

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I. INTRODUCTION

The ultrasonic characterization of porous materials saturated by air [1,2] such as plastic foams, fibrous, or granular materials is of great interest for a wide range of industrial applications. These materials are frequently used in the automotive and aeronautics industries and in the building trade. When a sound wave travels in an air-saturated porous medium, the dispersive effects are due to the frequency dependence of the complex functions [1-4] of the physical parameters of the medium, hereafter referred to as the generalized susceptibilities [5,6], which describe the fluid-structure interactions. The analysis of the propagation of transient waves in such media encounters two different problems arising from direct and inverse scattering. The direct scattering problem [7–12] is that of determining the scattered fields when the incident wave is known. In most cases, the determination of medium parameters is done in the frequency domain [13,14].

This time-domain model is an alternative to the classical frequency-domain approach. It is an advantage of the timedomain method [7,8,15-17] that the results are immediate and direct. The attraction of a time-domain based approach is that analysis is naturally bounded by the finite duration of ultrasonic pressures and it is consequently the most appropriate approach for transient signals. However, for wave propagation generated by time harmonic incident waves and sources (monochromatic waves), the frequency analysis is more appropriate [1,13,14]. A time-domain approach differs from frequency analysis in that the susceptibility functions describing viscous and thermal effects are convolution operators acting on velocity and pressure, and therefore a different algebraic formalism must be applied to solve the wave equation. The time-domain response of the material is described by an instantaneous response and a susceptibility kernel responsible for memory effects [7–12].

The acoustic propagation in homogeneous porous materials has been well studied, different methods and techniques were developed in frequency [1–3,13,14] and time domains [7–11,20–22] for the acoustic characterization. All these techniques are valid only for homogeneous porous materials, in which their physical parameters are constants inside the porous medium. However, in the general case, the porous media are layered [23,24] and their physical properties are locally constants, i.e., they are constant in the elementary volume of homogenization [23], but they may vary point to point in the porous medium. For this general case, a good understanding of the acoustic propagation is necessary for developing new methods of characterization.

This work follows the investigation previously done in Ref. [7], in which a time-domain approach was developed. Here, a general expression for the equation of wave propagation in a layered porous medium is derived.

The outline of this paper is as follows. Section II shows the equivalent fluid model; the relaxation functions describing the inertial, viscous, and thermal interactions between fluid and structure are recalled. In this section, the connection between the fractional derivatives and wave propagation in rigid homogeneous porous media in the high frequency range is established. Finally, in Sec. III the analytical derivation of the general propagation equation is given in time domain. The different terms of this equation are discussed.

II. THE EQUIVALENT FLUID MODEL

In air saturated porous media, the structure is assumed to be motionless: the acoustic waves travel only in the fluid filling the pores. The wave propagation is described by the equivalent fluid model which is a particular case of Biot's theory [18]. In this model, the interactions between the fluid and the structure are taken into account in two frequency

dependent response factors which are the generalized susceptibilities: the dynamic tortuosity of the medium $\alpha(\omega)$ [3] and the dynamic compressibility of the air included in the medium $\beta(\omega)$ [1,4]. These two response factors are complex functions which heavily depend on the frequency $f=\omega/2\pi$. These functions represent the deviation from the behavior of the fluid in the free space as the frequency increases. Their theoretical expressions are given by Johnson *et al.* [4], and Allard [1] and Lafarge *et al.* [4]:

$$\alpha(\omega) = \alpha_{\infty} \left(1 + \frac{\phi \sigma}{i \omega \alpha_{\infty} \rho} \sqrt{1 + i \frac{4 \alpha_{\infty}^2 \eta \rho \omega}{\sigma^2 \Lambda^2 \phi^2}} \right), \tag{1}$$

$$\beta(\omega) = \gamma - (\gamma - 1) \left(1 + \frac{\eta \phi}{i \omega \rho k_0' P_r} \sqrt{1 + i \frac{4k_0'^2 \rho \omega P_r}{\eta \phi^2 \Lambda'^2}} \right)^{-1}, \tag{2}$$

where $i^2 = -1$, γ represents the adiabatic constant, P_r is the Prandtl number, α_{∞} is the tortuosity, σ is the flow resistivity, k'_0 is the thermal permeability [4], Λ and Λ' are the viscous and thermal characteristic lengths [1,3,4], η is the fluid viscosity, ϕ is the porosity, and ρ is the fluid density. This model was initially developed by Johnson [3], and completed by Allard [1] by adding the description of thermal effects. Later on, Lafarge [4] introduced the parameter k'_0 which describes the additional damping of sound waves due to the thermal exchanges between fluid and structure at the surface of the pores. Generally the ration between Λ' and Λ is between 2 and 3. For the porous materials having cylindrical pores, the characteristic lengths are equal to the radius of the pores. For the most resistive porous materials $\Lambda = 10 \ \mu m$ (sandstone, cancellous bone), and for the less resistive porous materials $\Lambda = 400 \ \mu m$ (plastic foam, glass wool).

The functions $\alpha(\omega)$ and $\beta(\omega)$ express the viscous and thermal exchanges between the air and the structure which are responsible for the sound damping in acoustic materials. These exchanges are due on the one hand to the fluidstructure relative motion and on the other hand to the air compressions-dilatations produced by the wave motion. The part of the fluid affected by these exchanges can be estimated by the ratio of a microscopic characteristic length of the media, as, for example, the sizes of the pores, to the viscous and thermal skin depth thickness $\delta = (2 \eta / \omega \rho)^{1/2}$ and δ' = $(2 \eta / \omega \rho P_r)^{1/2}$. For the viscous effects this domain corresponds to the region of the fluid in which the velocity distribution is perturbed by the frictional forces at the interface between the viscous fluid and the motionless structure. For the thermal effects, it is the fluid volume affected by the heat exchanges between the two phases of the porous medium, the solid skeleton being seen as a heat sink. At high frequencies, the viscous and thermal skin thicknesses are very small compared to the radius of the pore r. The viscous and thermal effects are concentrated in a small volume near the surface of the frame $\delta/r \ll 1$ and $\delta'/r \ll 1$. In this case, the expressions of the dynamic tortuosity and compressibility are given by the relations

$$\alpha(\omega) = \alpha_{\infty} \left[1 + \frac{2}{\Lambda} \left(\frac{\eta}{i\omega\rho} \right)^{1/2} \right], \tag{3}$$

$$\beta(\omega) = 1 + \frac{2(\gamma - 1)}{\Lambda'} \left(\frac{\eta}{i\omega P_r \rho} \right)^{1/2}.$$
 (4)

In the time domain, these factors are operators and their asymptotic expressions are given by Ref. [7] as

$$\alpha(t) = \alpha_{\infty} \left[\delta(t) + \frac{2}{\Lambda} \left(\frac{\eta}{\pi \rho} \right)^{1/2} t^{-1/2} \right], \tag{5}$$

$$\beta(t) = \left[\delta(t) + \frac{2(\gamma - 1)}{\Lambda'} \left(\frac{\eta}{\pi P r \rho} \right)^{1/2} t^{-1/2} \right]. \tag{6}$$

In each of these equations the first term in the right-hand side is the instantaneous response of the medium $[\delta(t)]$ is the Dirac function] while the second term is the memory function. In electromagnetism, the instantaneous response is called optical response. It describes all the processes which cannot be resolved by the signal. The time convolution of $t^{-1/2}$ with a function is interpreted as a fractional derivative operator according to the definition (for order ν) given by Samko and colleagues [19],

$$D^{\nu}[x(t)] = \frac{1}{\Gamma(-\nu)} \int_0^t (t-u)^{-\nu-1} x(u) du, \tag{7}$$

where $\Gamma(x)$ is the Gamma function.

In this framework, the basic equations of the acoustic waves propagation along the ox axis are

$$\rho \widetilde{\alpha}(t) * \frac{\partial w}{\partial t} = -\phi \frac{\partial p}{\partial x}, \tag{8}$$

$$\frac{\phi \tilde{\beta}(t)}{K_a} * \frac{\partial p}{\partial t} = -\frac{\partial w}{\partial x}.$$
 (9)

The first equation is the Euler equation, the second one is the constitutive equation. K_a is the bulk modulus of air, p is the acoustic pressure, and $\mathbf{w} = \phi \mathbf{v}$ where \mathbf{v} is the particle velocity, * denotes the shorthand notation for the time convolution

$$(f * g)(t) = \int_{0}^{t} f(t - t')g(t')dt'.$$
 (10)

The wave equation is deduced from these equations:

$$\frac{\partial^2 p}{\partial x^2} - A \frac{\partial^2 p}{\partial t^2} - B \int_0^t \frac{\partial^2 p / \partial t'^2}{\sqrt{t - t'}} dt' - C \frac{\partial p}{\partial t} = 0, \tag{11}$$

where coefficients A, B, and C are constants given by

$$A = \frac{\rho \alpha_{\infty}}{K_a}, \quad B = \frac{2\alpha_{\infty}}{K_a} \sqrt{\frac{\rho \eta}{\pi}} \left(\frac{1}{\Lambda} + \frac{\gamma - 1}{\sqrt{Pr}\Lambda'} \right),$$

$$C = \frac{4\alpha_{\infty}(\gamma - 1)\eta}{K \Lambda \Lambda' \sqrt{Pr}}.$$
(12)

The first coefficient is related to the velocity $c=1/\sqrt{\rho\alpha_{\infty}/K_a}$ of the wave in the air included in the porous material. The

other coefficients are essentially dependent of the characteristic lengths Λ and Λ' and express the viscous and thermal interactions between the fluid and the structure, respectively. The coefficient B governs the spreading of the signal, while C is responsible of the attenuation of the wave. This propagation equation has been solved analytically in Ref. [9]. The direct [10–12] and inverse [20–22] scattering problem for a slab of porous material has been studied given a good estimation of the physical parameters (tortuosity α_{∞} , porosity ϕ , and characteristic length Λ and Λ').

III. GENERALIZED PROPAGATION EQUATION IN LAYERED POROUS MATERIALS

Consider the propagation of transient acoustic waves in a layered porous material having rigid frame. In this material, the acoustical parameters (porosity, tortuosity, viscous, and thermal characteristic lengths) depend on the thickness. For a wave propagating along the x axis, the fluid-structure interactions are described by the layered relaxation operators $\alpha(x,t)$ and $\beta(x,t)$ given by

$$\alpha(x,t) = \alpha_{\infty}(x) \left[\delta(t) + \frac{2}{\Lambda(x)} \left(\frac{\eta}{\pi \rho} \right)^{1/2} t^{-1/2} \right], \tag{13}$$

$$\beta(x,t) = \left[\delta(t) + \frac{2(\gamma - 1)}{\Lambda'(x)} \left(\frac{\eta}{\pi P r \rho}\right)^{1/2} t^{-1/2}\right]. \tag{14}$$

In these equations, the tortuosity $\alpha_{\infty}(x)$ and viscous and thermal characteristic lengths $\Lambda(x)$ and $\Lambda'(x)$ depend on the thickness of the porous material for describing the layered losses in the material.

In this framework, the basic equations [23,25] for our model can be written as

$$\rho\alpha(x,t) * \frac{\partial w(x,t)}{\partial t} = -\phi(x) \frac{\partial p(x,t)}{\partial x}, \qquad (15)$$

$$\frac{\phi(x)}{K_a}\beta(x,t) * \frac{\partial p(x,t)}{\partial t} = -\frac{\partial w(x,t)}{\partial x},$$
 (16)

where $\phi(x)$ represents the variation of porosity with depth. In the next section, the generalized propagation equation in layered porous material having an acoustical parameter varying with depth is derived. The derivation of the generalized wave equation in layered porous material is important for computing the propagation of an acoustic pulse inside the medium, and for solving the direct and inverse scattering problems.

Let us consider the Euler equation (15) and the constitutive one (16) in an infinite layered porous material. By putting $a(x) = \frac{2}{\Lambda(x)} \sqrt{\frac{\eta}{\rho \pi}}$ and $b(x) = \frac{2(\gamma - 1)}{\Lambda'(x)} \sqrt{\frac{\eta}{P_r \rho \pi}}$, we obtain

$$\rho \alpha_{\infty}(x) \left(\delta(t) + \frac{a(x)}{\sqrt{t}} \right) * \frac{\partial w(x,t)}{\partial t} = -\phi(x) \frac{\partial p(x,t)}{\partial x}, \quad (17)$$

$$\frac{\phi(x)}{K_a} \left(\delta(t) + \frac{b(x)}{\sqrt{t}} \right) * \frac{\partial p(x,t)}{\partial t} = -\frac{\partial w(x,t)}{\partial x}. \tag{18}$$

We note P(x,z), the Laplace transform of p(x,t), defined by

$$P(x,z) = \mathcal{L}[p(x,t)] = \int_0^\infty \exp(-zt)p(x,t)dt.$$
 (19)

The Laplace transform of Eqs. (17) and (18) yields

$$\rho \alpha_{\infty}(x) \left(1 + a(x) \sqrt{\frac{\pi}{z}} \right) z W(x, z) = -\phi(x) \frac{\partial P(x, z)}{\partial x}, \quad (20)$$

$$\frac{\phi(x)}{K_a} \left(1 + b(x) \sqrt{\frac{\pi}{z}} \right) z P(x, z) = -\frac{\partial W}{\partial x}(x, z), \tag{21}$$

where W(x,z) is the Laplace transform of w(x,t).

Using Eqs. (20) and (21) (see the Appendix), we obtain the following equation:

$$\frac{1}{c^{2}(x)}z^{2}P(x,z) + B'(x)\sqrt{\frac{\pi}{z}}z^{2}P(x,z) + D'(x)zP(x,z)
+ \frac{\partial a(x)}{\partial x}\frac{1}{c^{2}(x)\phi(x)}
\times \int_{0}^{x}\phi(y)\left(\sqrt{\frac{\pi}{z}}z^{2}P(y,z) + \pi b(y)zP(y,z)\right)dy
= \frac{\partial^{2}P(x,z)}{\partial x^{2}} + \frac{\partial P(x,z)}{\partial x}\left(\frac{\partial \ln \phi(x)}{\partial x} - \frac{\partial \ln \alpha_{\infty}(x)}{\partial x}\right), (22)$$

where

$$\frac{\rho \alpha_{\infty}(x)}{K_a} = \frac{1}{c^2(x)}; \quad \frac{\rho \alpha_{\infty}(x)}{K_a} [a(x) + b(x)] = B'(x)$$
and
$$\frac{\rho \alpha_{\infty}(x)}{K_a} \pi a(x) b(x) = D'(x).$$

Using the inverse Laplace transform of Eq. (22) and the initial conditions $[9,12] \frac{\partial p}{\partial t}(x,0) = p(x,0) = 0$, we find the generalized propagation equation in time domain,

$$\frac{\partial^{2} p}{\partial x^{2}}(x,t) - \frac{1}{c^{2}(x)} \frac{\partial^{2} p}{\partial t^{2}}(x,t) - B'(x) \int_{0}^{t} \frac{\partial^{2} p}{\partial t^{2}}(x,t-\tau) \frac{d\tau}{\sqrt{\tau}}$$

$$- D'(x) \frac{\partial p}{\partial t}(x,t) - \frac{\partial a(x)}{\partial x} \frac{1}{c^{2}(x)\phi(x)}$$

$$\times \int_{0}^{x} \phi(y) \left(\int_{0}^{t} \frac{\partial^{2} p}{\partial t^{2}}(y,t-\tau) \frac{d\tau}{\sqrt{\tau}} + \pi b(y) \frac{\partial p}{\partial t}(y,t) \right)$$

$$\times dy - \frac{\partial p}{\partial x}(x,t)\varphi(x) = 0, \tag{23}$$

with

$$\varphi(x) = \frac{\partial}{\partial x} \ln \frac{\alpha_{\infty}(x)}{\phi(x)}.$$

Equation (23) is the generalized propagation equation for lossy layered porous material. This equation is very important for treating the direct and inverse scattering problems in layered porous materials in time domain. It is easy to find the special case of homogeneous porous medium, i.e., when $\alpha_{\infty}(x)$, $\phi(x)$, $\Lambda(x)$, and $\Lambda'(x)$ become constants (independent

of x), we find B'(x)=B, D'(x)=C, $\varphi(x)=\partial a(x)/\partial x=0$. In this case, the generalized wave propagation [Eq. (23)] is reduced to the propagation equation in homogeneous material [Eq. (11)].

The first and second term in the propagation equation (23), $\frac{\partial^2 p}{\partial x^2}(x,t) - \frac{1}{c^2(x)} \frac{\partial^2 p}{\partial t^2}(x,t)$, describe the propagation (time translation) via the front wave velocity c(x). The layered tortuosity $\alpha_{\infty}(x)$ appears as the refractive index of the medium which changes the wave velocity from $c_0 = \sqrt{K_a/\rho}$ in free space to $c(x) = c_0/\sqrt{\alpha_{\infty}(x)}$ in the porous medium. From this equation, it can be seen that only the inertial effects [represented by the spatial profile of the tortuosity $\alpha_{\infty}(x)$] modify the front wave velocity.

The third term in the propagation equation (23), $B'(x)\int_0^t \frac{\partial^2 p}{\partial t^2}(x,t-\tau)\frac{d\tau}{\sqrt{\tau}}$, contains a time fractional derivative of order 3/2 [see the definition of fractional derivatives in Eq. (7)]. This term is the most important one for describing the dispersion, memory effects (historical phenomena due to relaxations times), and the acoustic attenuation in porous materials. These effects are due to losses in the medium modeled by the viscous and thermal exchanges between fluid and structure, and described by the characteristic lengths $\Lambda(x)$ and $\Lambda'(x)$. This term results from the time convolution of the fractional derivatives operators of tortuosity $\alpha(x,t)$ and compressibility $\beta(x,t)$. It is sensitive to the spatial variation of the tortuosity $\alpha_{\infty}(x)$. The high frequency components of the transient signal are the most sensitive to this term (due to the fractional derivative).

The fourth term in the propagation equation (23), $D'(x)\frac{\partial p}{\partial t}(x,t)$, is an attenuating term; it results in the attenuation of the wave without dispersion. This term describes the acoustic attenuation due to the viscous and thermal interactions between fluid and structure, and to acoustic attenuation caused by the spatial variation of the tortuosity. The low frequency components of the transient signal are the most sensitive to this term.

The final term, $\varphi(x)\frac{\partial p}{\partial x}(x,t)$, describes the attenuation caused by the spatial variation of the tortuosity and the porosity. In contrast to the other terms, theses two terms are independent of the relaxations times of the medium and thus to the frequency component of the acoustic signal (i.e., there is no temporal derivative).

The spatial variation of the porosity $\phi(x)$ appears in the propagation equation only via the two end terms. We recall that in the homogeneous case, the propagation equation [Eq. (11)] is independent of the porosity; this parameter appears in the response of the homogeneous medium when the boundary conditions of the problem are introduced [10].

Finally, the term $-\frac{\partial a(x)}{\partial x}\frac{1}{c^2(x)\phi(x)}\int_0^x \int_0^x \int_0^{t} \frac{\partial^2 p}{\partial t^2}(y,t-\tau)\frac{d\tau}{\sqrt{\tau}} + \pi b(y)\frac{\partial p}{\partial t}(y,t)\right]dy$ describes the spatial variation of the inhomogeneity of the porous medium due to the temporal dispersion (viscous and thermal) of the medium.

IV. CONCLUSION

In this paper the generalized wave equation in layered porous material is established using fractional calculus. The different terms of the propagation equation show how the spatial variation of the tortuosity, porosity, and characteristic length affect the propagation. Future studies will concentrate on the direct and inverse scattering problems, and methods and inversion algorithms will be developed to optimize the acoustic properties of layered porous media.

APPENDIX

By differentiating both sides of Eq. (20) with respect to x, one finds that

$$\rho \frac{\partial \alpha_{\infty}(x)}{\partial x} \left(1 + a(x) \sqrt{\frac{\pi}{z}} \right) z W(x, z) + \rho \alpha_{\infty}(x) \frac{\partial a(x)}{\partial x}$$

$$\times \sqrt{\frac{\pi}{z}} z W(x, z) + \rho \alpha_{\infty}(x) \left(1 + a(x) \sqrt{\frac{\pi}{z}} \right) z \frac{\partial W(x, z)}{\partial x}$$

$$= -\phi(x) \frac{\partial^2 P(x, z)}{\partial x^2} - \frac{\partial P(x, z)}{\partial x} \frac{\partial \phi(x)}{\partial x}.$$
(A1)

The first term of Eq. (A1) gives

$$\rho \frac{\partial \alpha_{\infty}(x)}{\partial x} \left(1 + a(x) \sqrt{\frac{\pi}{z}} \right) z W(x, z)$$

$$= \frac{\partial \alpha_{\infty}(x)}{\alpha_{\infty}(x) \partial x} \rho \alpha_{\infty}(x) \left(1 + a(x) \sqrt{\frac{\pi}{z}} \right) z W(x, z),$$

and by taking into account Eq. (20), we obtain

$$\rho \frac{\partial \alpha_{\infty}(x)}{\partial x} \left(1 + a(x) \sqrt{\frac{\pi}{z}} \right) z W(x, z)$$

$$= -\frac{\partial P(x, z)}{\partial x} \phi(x) \frac{\partial \ln[\alpha_{\infty}(x)]}{\partial x}.$$
(A2)

The spatial integration of Eq. (21) from 0 to x yields

$$W(x,z) = W(0,z) - \frac{1}{K_a} \int_0^x \phi(y) \left(1 + b(y) \sqrt{\frac{\pi}{z}} \right) z P(y,z) dy.$$

Assuming the same initial conditions than those given in Refs. [9,12], which means that the medium is at rest for $t \le 0$, $v(0,t) = \frac{\partial v(0,t)}{\partial t} = 0 \Rightarrow W(0,z) = 0$, and by multiplying the two members by z,

$$zW(x,z) = -\frac{1}{K_a} \int_0^x \phi(y) \left(1 + b(y) \sqrt{\frac{\pi}{z}} \right) z^2 P(y,z) dy.$$
(A3)

Using Eqs. (A1) and (A3), we obtain

$$\rho \alpha_{\infty}(x) \frac{\partial a(x)}{\partial x} \sqrt{\frac{\pi}{z}} z W(x, z)$$

$$= -\frac{\rho \alpha_{\infty}(x)}{K_a} \frac{\partial a(x)}{\partial x} \int_0^x \phi(y) \left(\sqrt{\frac{\pi}{z}} z^2 P(y, z) + \pi b(y) z P(y, z) \right) dy. \tag{A4}$$

By replacing $\frac{\partial W(x,z)}{\partial x}$ by its expression given in Eq. (21) into Eq. (A1), we obtain

$$\rho \alpha_{\infty}(x) \left(1 + a(x) \sqrt{\frac{\pi}{z}} \right) z \frac{\partial W(x, z)}{\partial x}$$

$$= -\frac{\rho \alpha_{\infty}(x) \phi(x)}{K_a} \left(1 + a(x) \sqrt{\frac{\pi}{z}} \right) \left(1 + b(x) \sqrt{\frac{\pi}{z}} \right) z^2 P(x, z)$$

$$= -\frac{\rho \alpha_{\infty}(x) \phi(x)}{K_a} \left(1 + \left[a(x) + b(x) \right] \sqrt{\frac{\pi}{z}} \right)$$

$$+ \frac{\pi a(x) b(x)}{z} z^2 P(x, z). \tag{A5}$$

Equation (A1) takes the following form

$$-\frac{\partial P(x,z)}{\partial x}\phi(x)\frac{\partial \ln \alpha_{\infty}(x)}{\partial x} - \frac{\rho \alpha_{\infty}(x)}{K_{a}}\frac{\partial a(x)}{\partial x}$$

$$\times \int_{0}^{x}\phi(y)\left(\sqrt{\frac{\pi}{z}}z^{2}P(y,z) + \pi b(y)zP(y,z)\right)dy$$

$$-\frac{\rho \alpha_{\infty}(x)\phi(x)}{K_{a}}\left(1 + (a(x) + b(x))\sqrt{\frac{\pi}{z}}\right)$$

$$+\frac{\pi a(x)b(x)}{z}z^{2}P(x,z)$$

$$= -\phi(x)\frac{\partial^{2}P(x,z)}{\partial x^{2}} - \frac{\partial P(x,z)}{\partial x}\frac{\partial \phi(x)}{\partial x}.$$
(A6)

After some changes Eq. (A6) can be written as Eq. (22).

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