# Weakly nonlinear propagation of pressure waves in bubbly liquids with a polydispersity based on two-fluid model equations 

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#### Abstract

Herein, the weak, nonlinear propagation of pressure waves in an initially quiescent liquid containing many small spherical gas bubbles is theoretically studied. We focus on the initial, polydispersity features of the bubble radius and bubble number density. Our analysis was not based on any assumptions about explicit polydispersity forms. Using equations based on a two-fluid model and the method of multiple scales with perturbation expansions, the Korteweg-de Vries-Burgers equation for a low-frequency long wave and nonlinear Schrödinger equation for a high-frequency short wave were derived. In both cases, the polydispersity contributes to the advection and dissipation effects of waves, and every coefficient in both equations includes the initial void fraction as one of the most important parameters, owing to the use of the two-fluid model.


## 1. Introduction

The propagation properties of pressure (acoustic) waves in liquids containing microbubbles are considerably different from those in pure liquids (see, e.g., van Wijngaarden, 1972). In the case of pure liquids, the pressure wave generally evolves into a shock wave owing to the balance between the nonlinear effect of waves and the dissipation effect of the medium. However, in the case of bubbly liquids, volumetric oscillations of bubbles induce a dispersion effect of waves (van Wijngaarden, 1968), which then evolves into (acoustic) soliton as a result of the balance between the nonlinear and dispersion effects for a nondissipative medium (van Wijngaarden, 1968). Because the physical properties of shock waves and solitons are considerably different, the correct prediction of the evolved waveform is considerably important. Hence, the estimation of the magnitude of the three effects (i.e., nonlinear, dissipation, and dispersion effects) is effective for predicting the evolved waveform. Propagation properties of pressure waves in bubbly liquids have long been theoretically (e.g., van Wijngaarden, 1968, 1972; Noordzij and van Wijngaarden, 1974; Kuznetsov et al., 1978; Biesheuvel and van Wijngaarden, 1984; Caflisch et al., 1985; Commander and Prosperetti, 1989; Nigmatulin, 1991; Prosperetti, 1991; Gumerov, 1992a,b; Nakoryakov et al., 1993; Akhatov et al., 1994, 1996; Jamshidi and Brenner, 2013; Gubaidullin and Fedorov, 2013; Gubaidullin et al., 2013, 2022; Kudryashov and Sinelshchikov, 2014; Gubaidullin and Fedorov, 2015, 2016; Gumerov and Akhatov, 2017), numerically (e.g., Nigmatulin, 1991; Nakoryakov et al., 1993; Kameda
and Matsumoto, 1996; Kameda et al., 1998; Vanhille and CamposPozuelo, 2009; Ando et al., 2011; Frolov et al., 2017), and experimentally (e.g., Kuznetsov et al., 1978; Nigmatulin, 1991; Nakoryakov et al., 1993; Kameda and Matsumoto, 1996; Kameda et al., 1998; Frolov et al., 2017) studied. However, we cannot obtain the magnitude of the three effects through experiments and direct numerical simulations of the governing equations. The key role in obtaining the three effects is a theoretical derivation of the weakly nonlinear wave equation as a linear combination of the three effects. By deriving nonlinear wave equations via theoretical analysis and estimating the magnitude of the three effects, experiments and numerical calculations are supported.

Various studies on nonlinear wave equations in bubbly liquids have been published (van Wijngaarden, 1968, 1972; Noordzij and van Wijngaarden, 1974; Kuznetsov et al., 1978; Nigmatulin, 1991; Prosperetti, 1991; Gumerov, 1992a,b; Nakoryakov et al., 1993; Akhatov et al., 1994, 1996; Gubaidullin and Fedorov, 2013; Gubaidullin et al., 2013, 2022; Kudryashov and Sinelshchikov, 2014; Gubaidullin and Fedorov, 2015, 2016). In particular, the derivation of Korteweg-de Vries (KdV) and KdV-Burgers (KdVB) equations by van Wijngaarden (van Wijngaarden, 1968, 1972) is the most famous work. Recently, our group (Kanagawa et al., 2010, 2021a; Yano et al., 2013; Kanagawa, 2015) derived two equations, that is, the KdVB equation for low frequency long waves (Kanagawa et al., 2010, 2021a; Yano et al., 2013; Kanagawa, 2015) and the nonlinear Schrödinger (NLS) equation for high frequency short waves (Kanagawa et al., 2010, 2021a; Yano et al.,

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Fig. 1. The linear dispersion relation for pressure waves in bubbly liquids, and a decomposition into two frequency bands, that is, the low frequency band and the high frequency band described by the KdVB equation and the NLS equation, respectively (Kanagawa et al., 2010, 2021a; Yano et al., 2013).

2013; Kanagawa, 2015), based on a systematic decomposition of two frequency bands, as illustrated in Fig. 1 (Kanagawa et al., 2010). In this study, only one set of basic equations is used throughout this paper, but the equations for scaling (see (13) below) are switched to derive two types of equations (KdVB and NLS) corresponding to the two bands in Fig. 1.

Nonlinear wave equations are derived from model (or basic) equations for bubbly flows (or bubbly liquids). Although numerous model equations have been proposed, volumetric averaged models (Biesheuvel and van Wijngaarden, 1984; Caflisch et al., 1985; Jones and Prosperetti, 1985; Nigmatulin, 1991; Egashira et al., 2004) have been generally used. All the researchers except for our group (Kanagawa et al., 2010, 2021a; Yano et al., 2013; Kanagawa, 2015) have long been utilizing mixtures or homogeneous models (see, e.g., van Wijngaarden, 1968, 1972; Noordzij and van Wijngaarden, 1974; Kuznetsov et al., 1978; Biesheuvel and van Wijngaarden, 1984; Nigmatulin, 1991; Prosperetti, 1991; Nakoryakov et al., 1993; Akhatov et al., 1994, 1996; Kudryashov and Sinelshchikov, 2014). Our recent study (Yano et al., 2013) revealed that the use of a mixture model cannot express the dependence of coefficients in nonlinear, dissipation, and dispersion terms of nonlinear wave equations on the initial void fraction, which is a fatal disadvantage from the perspective of engineering or industrial applications; in contrast, the use of two-fluid model equations (Biesheuvel and van Wijngaarden, 1984; Egashira et al., 2004; Kanagawa et al., 2010; Yano et al., 2013; Kanagawa et al., 2021a) can reflect the dependence of each coefficient on the initial void fraction (Kanagawa et al., 2010; Yano et al., 2013). However, our previous studies (e.g., Kanagawa et al., 2010; Yano et al., 2013) except for Kanagawa et al. (2021a) and Kanagawa et al. (2022) assumed initial monodispersity of a bubbly liquid. In the real situation, the bubbly liquid has polydispersity (i.e., nonuniformity of the bubble radius and spatial distribution). Although some studies (Gumerov, 1992a,b; Gubaidullin and Fedorov, 2013; Gubaidullin et al., 2013, 2022; Gubaidullin and Fedorov, 2015, 2016) incorporated the initial polydispersity, no study using the two-fluid model incorporated the initial polydispersity despite the independence of using the two-fluid model and incorporating polydispersity. Although our previous study (Kanagawa et al., 2021a) succeeded in incorporating the initial small polydispersity into our original monodisperse case (Kanagawa et al., 2010), Kanagawa et al. (2021a) contains a critical restriction of polydispersity. In Kanagawa et al. (2021a), when the field was divided into several regions, an assumption was made that polydispersity appeared only in specific regions. Thus, the purpose of this study is to incorporate general initial small polydispersity in all regions using nonlinear wave equations from two-fluid model equations that model the initial polydispersity (see Fig. 2).

The contents of the present paper are as follows: Section 2 introduces the basic equations and perturbation expansions based on the multiple-scale method, with a special focus on the formulation of the initial polydispersity. Section 3 derives the resultant KdVB equation


Fig. 2. Difference between (a) our previous study (Kanagawa et al., 2021a) and (b) this study for KdVB equation. Although the polydispersity of bubble radius (not bubble number density) does not appear in near field, the polydispersity of bubble radius appears in this figure.
and Section 4 derives the resultant NLS equation including new terms. Section 5 discusses the physical meanings of the terms of the resultant equations and compares the new equations with those in previous studies. Section 6 is devoted to the summary of this study.

## 2. Formulation of the problem

### 2.1. Problem statement

The propagation of nonlinear pressure waves in initially quiescent compressible liquids containing a number of small spherical gas bubbles is theoretically studied. We focus on the one-dimensional propagation because bubble oscillations are assumed as spherically symmetric, and then the phenomenon can be regarded as a one-directional dependency. Initially, the bubbly liquid has a small (see (28) and (29) below) polydispersity (i.e., nonuniformity of bubble radius, bubble number density, and void fraction). The amplitude of the pressure wave is finite but is sufficiently small (i.e., a weakly nonlinear wave (e.g., Jeffrey and Kawahara, 1982)) The bubbles do not coalesce, break up, appear, and disappear. For simplicity, the gas viscosity, thermal conduction (e.g., Prosperetti, 1991; Kameda and Matsumoto, 1996; Preston et al., 2007; Stricker et al., 2011; Kanagawa and Kamei, 2021; Kamei et al., 2021), and mass transport across the bubble-liquid interface are ignored. We consider the liquid viscosity at the bubble-liquid interface. The drag (Yatabe et al., 2021; Kanagawa et al., 2021b; Arai et al., 2022) and lift forces acting on the bubbles, Reynolds stress, and gravitation are not considered.

### 2.2. Basic equations

The basic equations are composed of nine equations: four conservation equations, bubble dynamics equations, and four constitutive equations. As volumetric-averaged equations based on a twofluid model (Egashira et al., 2004; Yano et al., 2006) to describe the dependence of the initial void fraction (Kanagawa et al., 2010), conservation laws of mass and momentum for gas and liquid phases are first introduced as follows:
$\frac{\partial}{\partial t^{*}}\left(\alpha \rho_{\mathrm{G}}^{*}\right)+\frac{\partial}{\partial x^{*}}\left(\alpha \rho_{\mathrm{G}}^{*} u_{\mathrm{G}}^{*}\right)=0$,
$\frac{\partial}{\partial t^{*}}\left[(1-\alpha) \rho_{\mathrm{L}}^{*}\right]+\frac{\partial}{\partial x^{*}}\left[(1-\alpha) \rho_{\mathrm{L}}^{*} u_{\mathrm{L}}^{*}\right]=0$,
$\frac{\partial}{\partial t^{*}}\left(\alpha \rho_{\mathrm{G}}^{*} u_{\mathrm{G}}^{*}\right)+\frac{\partial}{\partial x^{*}}\left(\alpha \rho_{\mathrm{G}}^{*} u_{\mathrm{G}}^{* 2}\right)+\alpha \frac{\partial p_{\mathrm{G}}^{*}}{\partial x^{*}}=F^{*}$,
$\frac{\partial}{\partial t^{*}}\left[(1-\alpha) \rho_{\mathrm{L}}^{*} u_{\mathrm{L}}^{*}\right]+\frac{\partial}{\partial x^{*}}\left[(1-\alpha) \rho_{\mathrm{L}}^{*} u_{\mathrm{L}}^{* 2}\right]+(1-\alpha) \frac{\partial p_{\mathrm{L}}^{*}}{\partial x^{*}}+P^{*} \frac{\partial \alpha}{\partial x^{*}}=-F^{*}$,
where $t^{*}$ is time, $x^{*}$ is the space coordinate, $\alpha$ is the void fraction, $\rho^{*}$ is the density, $u^{*}$ is the fluid velocity, $p^{*}$ is the pressure, and $P^{*}$ is the surface-averaged pressure (Jones and Prosperetti, 1985); the subscripts $G$ and $L$ denote the gas and liquid phases, respectively.

As the interfacial transport term of momentum $F^{*}$, we use the virtual mass force model (Yano et al., 2006; Eames and Hunt, 2004):
$F^{*}=-\beta_{1} \alpha \rho_{\mathrm{L}}^{*}\left(\frac{\mathrm{D}_{\mathrm{G}} u_{\mathrm{G}}^{*}}{\mathrm{D} t^{*}}-\frac{\mathrm{D}_{\mathrm{L}} u_{\mathrm{L}}^{*}}{\mathrm{D} t^{*}}\right)-\beta_{2} \rho_{\mathrm{L}}^{*}\left(u_{\mathrm{G}}^{*}-u_{\mathrm{L}}^{*}\right) \frac{\mathrm{D}_{\mathrm{G}} \alpha}{\mathrm{D} t^{*}}-\beta_{3} \alpha\left(u_{\mathrm{G}}^{*}-u_{\mathrm{L}}^{*}\right) \frac{\mathrm{D}_{\mathrm{G}} \rho_{\mathrm{L}}^{*}}{\mathrm{D} t^{*}}$,
where $\beta_{1}, \beta_{2}$, and $\beta_{3}$ are the virtual mass coefficients and are $1 / 2$ for the case of spherical bubbles. Furthermore, $\mathrm{D}_{\mathrm{G}} / \mathrm{D} t^{*}$ and $\mathrm{D}_{\mathrm{L}} / \mathrm{D} t^{*}$ are material derivatives for the gas and liquid phases, respectively, as follows:
$\frac{\mathrm{D}_{\mathrm{G}}}{\mathrm{D} t^{*}}=\frac{\partial}{\partial t^{*}}+u_{\mathrm{G}}^{*} \frac{\partial}{\partial x^{*}}$,
$\frac{\mathrm{D}_{\mathrm{L}}}{\mathrm{D} t^{*}}=\frac{\partial}{\partial t^{*}}+u_{\mathrm{L}}^{*} \frac{\partial}{\partial x^{*}}$.
For bubble dynamics, the Keller equation (Keller and Kolodner, 1956; Keller and Miksis, 1980) for spherically symmetric volume oscillations of bubbles in a compressible liquid is utilized, as follows:

$$
\begin{align*}
(1- & \left.\frac{1}{c_{\mathrm{L} 0}^{*}} \frac{\mathrm{D}_{\mathrm{G}} R^{*}}{\mathrm{D} t^{*}}\right) R^{*} \frac{\mathrm{D}_{\mathrm{G}}^{2} R^{*}}{\mathrm{D} t^{* 2}}+\frac{3}{2}\left(1-\frac{1}{3 c_{\mathrm{L} 0}^{*}} \frac{\mathrm{D}_{\mathrm{G}} R^{*}}{\mathrm{D} t^{*}}\right)\left(\frac{\mathrm{D}_{\mathrm{G}} R^{*}}{\mathrm{D} t^{*}}\right)^{2} \\
& =\left(1+\frac{1}{c_{\mathrm{L} 0}^{*}} \frac{\mathrm{D}_{\mathrm{G}} R^{*}}{\mathrm{D} t^{*}}\right) \frac{P^{*}}{\rho_{\mathrm{L} 0}^{*}}+\frac{R^{*}}{\rho_{\mathrm{L} 0}^{*} c_{\mathrm{L} 0}^{*}} \frac{\mathrm{D}_{\mathrm{G}}}{\mathrm{D} t^{*}}\left(p_{\mathrm{L}}^{*}+P^{*}\right) \tag{8}
\end{align*}
$$

where $R^{*}$ is the bubble radius, $c_{\mathrm{L} 0}^{*}$ is the speed of sound in the pure liquid, and the subscript 0 denotes the initial quantity.

The following constitutive equations close (1)-(8):
(i) Tait equation of state for liquid
$p_{\mathrm{L}}^{*}=p_{\mathrm{L} 0}^{*}+\frac{\rho_{\mathrm{L} 0}^{*} c_{\mathrm{L} 0}^{*}{ }^{2}}{n}\left[\left(\frac{\rho_{\mathrm{L}}^{*}}{\rho_{\mathrm{L} 0}^{*}}\right)^{n}-1\right]$,
where $n$ is material constant; e.g., $n=7.15$ for water, (ii) polytropic equation of state for gas,
$\frac{p_{\mathrm{G}}^{*}}{p_{\mathrm{G} 0}^{*}}=\left(\frac{\rho_{\mathrm{G}}^{*}}{\rho_{\mathrm{G} 0}^{*}}\right)^{\gamma}$,
where $\gamma$ is polytropic exponent, (iii) the conservation law of mass inside bubble,
$\frac{\rho_{\mathrm{G}}^{*}}{\rho_{\mathrm{G} 0}^{*}}=\left(\frac{R_{0}^{*}}{R^{*}}\right)^{3}$,
(iv) the balance of normal stress across the bubble-liquid interface,
$p_{\mathrm{G}}^{*}-\left(p_{\mathrm{L}}^{*}+P^{*}\right)=\frac{2 \sigma^{*}}{R^{*}}+\frac{4 \mu^{*}}{R^{*}} \frac{\mathrm{D}_{\mathrm{G}} R^{*}}{\mathrm{D} t^{*}}$,
where $\sigma^{*}$ is the surface tension and $\mu^{*}$ is the liquid viscosity.

### 2.3. Parameter scaling

The scaling relations of nondimensional ratios among the physical parameters appropriate to the low-frequency and long-wavelength band (i.e., weak dispersion band) and the high-frequency and shortwavelength band (i.e., strong dispersion band) shown in Fig. 1 are defined as follows (Kanagawa et al., 2010).
$\left(\frac{R_{00}^{*}}{L^{*}}, \frac{\omega^{*}}{\omega_{\mathrm{B}}^{*}}, \frac{U^{*}}{c_{\mathrm{L} 0}}\right) \equiv\left\{\begin{array}{c}(O(\sqrt{\epsilon}), O(\sqrt{\epsilon}), O(\sqrt{\epsilon})) \\ \equiv(\Delta \sqrt{\epsilon}, \Omega \sqrt{\epsilon}, V \sqrt{\epsilon}), \quad(\mathrm{KdVB}), \\ \left(O(1), O(1), O\left(\epsilon^{2}\right)\right) \equiv\left(\Delta, \Omega, V \epsilon^{2}\right), \quad(\mathrm{NLS}),\end{array}\right.$
where $\epsilon$ is a nondimensional wave amplitude $(0<\epsilon \ll 1)$; $\Delta, \Omega$, and $V$ are parameters of $O(1), R_{00}^{*}$ is the initial radius of a single bubble, $L^{*}$ is a typical wavelength, $\omega^{*}$ is an angular frequency of incident waves, and $\omega_{\mathrm{B}}^{*}$ is the natural angular frequency of the linear spherical symmetric oscillation of a typical bubble given by
$\omega_{\mathrm{B}}^{*}=\sqrt{\frac{3 \gamma\left(p_{\mathrm{L} 0}^{*}+2 \sigma^{*} / R_{00}^{*}\right)-2 \sigma^{*} / R_{00}^{*}}{\rho_{\mathrm{L} 0}^{*} R_{00}^{*}{ }^{2}}}$,
and $U^{*}$ is a phase velocity of the incident wave. This study derives the KdVB and NLS equations. Here and hereafter, the labels (KdVB) and (NLS) at the end of equations signify the KdVB and NLS equations, respectively.

The ratio for length scale $R_{00}^{*} / L^{*}$ (or $\Delta$ ) in (13) signifies the magnitude of the dispersion effect of waves in bubbly liquid, caused by bubble oscillations (van Wijngaarden, 1968). The long wavelength case (the KdVB equation) is relatively weakly dispersive compared with the short wavelength case (the NLS equation), because the dispersion effect is owing to bubble oscillations. Bubble oscillations can be clearly observed in the short wavelength case compared with the low frequency case. This explains the right-hand side of (13).

Furthermore, the scaling relation of the liquid viscosity $\mu^{*}$ is
$\frac{\mu^{*}}{\rho_{\mathrm{L} 0}^{*} U^{*} L^{*}} \equiv\left\{\begin{array}{l}O(\epsilon) \equiv \mu \epsilon, \quad(\mathrm{KdVB}), \\ O\left(\epsilon^{2}\right) \equiv \mu \epsilon^{2}, \quad(\mathrm{NLS}),\end{array}\right.$
where $\mu$ is a constant of $O(1)$. By changing the right-hand side of (13) and (15), two types of nonlinear wave equations are derived without changing the basic equations (i.e., (1)-(12)).

### 2.4. Perturbation expansions and multiple scales analysis

First, independent variables $t^{*}$ and $x^{*}$ are nondimensionalized:
$t=\frac{t^{*}}{T^{*}}, \quad x=\frac{x^{*}}{L^{*}}$,
where $T^{*}$ is a typical period of the waves and satisfies the following:
$\frac{1}{T^{*}}= \begin{cases}\omega^{*}, & (\text { KdVB }), \\ \omega_{\mathrm{B}}^{*}, & (\mathrm{NLS}) .\end{cases}$
Using $t$ and $x$, the near-field, far-field I, and far-field II are described by extended independent variables in the method of multiple scales (e.g., Jeffrey and Kawahara, 1982):
$t_{0}=\epsilon^{0} t, \quad x_{0}=\epsilon^{0} x, \quad$ (near field),
$t_{1}=\epsilon^{1} t, \quad x_{1}=\epsilon^{1} x, \quad($ far field I),
$t_{2}=\epsilon^{2} t, \quad x_{2}=\epsilon^{2} x, \quad($ far field II).
In the case of the KdVB equation, far field II is not used (see Section 3), and far field I is called far field for simplicity. The differential operators are then expanded as follows:
$\frac{\partial}{\partial t}=\sum_{i=0}^{j} \epsilon^{i} \frac{\partial}{\partial t_{i}}, \quad \frac{\partial}{\partial x}=\sum_{i=0}^{j} \epsilon^{i} \frac{\partial}{\partial x_{i}}$,
where $j=1$ for the KdVB equation and $j=2$ for the NLS equation.
Dependent variables are now regarded as functions of the extended independent variables in (18)-(20). The expansions of dependent variables except for bubble radius $R^{*}$ and void fraction $\alpha$ are as follows (Kanagawa et al., 2010, 2021a)
$u_{\mathrm{G}}^{*} / U^{*}=\left\{\begin{array}{l}\epsilon^{1} u_{\mathrm{G} 1}+\epsilon^{2} u_{\mathrm{G} 2}+O\left(\epsilon^{3}\right), \quad(\mathrm{KdVB}), \\ \epsilon^{1} u_{\mathrm{G} 1}+\epsilon^{2} u_{\mathrm{G} 2}+\epsilon^{3} u_{\mathrm{G} 3}+O\left(\epsilon^{4}\right), \quad(\mathrm{NLS}),\end{array}\right.$
$u_{\mathrm{L}}^{*} / U^{*}=\left\{\begin{array}{l}\epsilon^{1} u_{\mathrm{L} 1}+\epsilon^{2} u_{\mathrm{L} 2}+O\left(\epsilon^{3}\right), \quad(\mathrm{KdVB}), \\ \epsilon^{1} u_{\mathrm{L} 1}+\epsilon^{2} u_{\mathrm{L} 2}+\epsilon^{3} u_{\mathrm{L} 3}+O\left(\epsilon^{4}\right), \quad(\mathrm{NLS}),\end{array}\right.$
$p_{\mathrm{L}}^{*} /\left(\rho_{\mathrm{L} 0}^{*} U^{* 2}\right)=\left\{\begin{array}{l}p_{\mathrm{L} 0}+\epsilon^{1} p_{\mathrm{L} 1}+\epsilon^{2} p_{\mathrm{L} 2}+O\left(\epsilon^{3}\right), \quad(\mathrm{KdVB}), \\ p_{\mathrm{L} 0}+\epsilon^{1} p_{\mathrm{L} 1}+\epsilon^{2} p_{\mathrm{L} 2}+\epsilon^{3} p_{\mathrm{L} 3}+O\left(\epsilon^{4}\right), \quad(\mathrm{NLS}),\end{array}\right.$
$\rho_{\mathrm{L}}^{*} / \rho_{\mathrm{L} 0}^{*}= \begin{cases}1+\epsilon^{2} \rho_{\mathrm{L} 1}+\epsilon^{3} \rho_{\mathrm{L} 2}+O\left(\epsilon^{4}\right), & (\mathrm{KdVB}), \\ 1+\epsilon^{5} \rho_{\mathrm{L} 1}+\epsilon^{6} \rho_{\mathrm{L} 2}+O\left(\epsilon^{7}\right), & (\mathrm{NLS}),\end{cases}$
where (25) is determined by (9) and (13) (Kanagawa et al., 2010). Equation (25) expresses the difference of compressibility between gas and liquid (i.e., gas is more compressible than liquid). We assume an initially quiescent bubbly liquid, and the effect of the initial flow of velocity is neglected, and the terms of $O(1)$ do not appear in the right hand sides of (22) and (23). As constants, the nondimensional pressures for the gas and liquid phases in the unperturbed state $p_{\mathrm{G} 0}$ and $p_{\mathrm{L} 0}$ are given as
$p_{\mathrm{G} 0}=\frac{p_{\mathrm{G} 00}^{*}}{\rho_{\mathrm{L} 0}^{*} U^{* 2}} \equiv O(1), \quad p_{\mathrm{L} 0}=\frac{p_{\mathrm{L} 0}^{*}}{\rho_{\mathrm{L} 0}^{*} U^{* 2}} \equiv O(1)$,
where $p_{\mathrm{G} 00}^{*}$ is the initial gas pressure in a single bubble. The ratio of the initial densities of the gas and liquid phases is
$\frac{\rho_{\mathrm{G} 00}^{*}}{\rho_{\mathrm{L} 0}^{*}}=O\left(\epsilon^{3}\right)$,
where $\rho_{\mathrm{G} 00}^{*}$ is the initial gas density of a single bubble.

### 2.5. Formulation of initial polydispersity

We incorporate a small initial polydispersity of the bubble radius $R^{*}$ and bubble number density $n^{*}$ (Kanagawa, 2015) by expanding $R^{*}$ and void fraction $\alpha$ as

$$
\begin{align*}
R^{*} / R_{00}^{*} & =\left\{\begin{array}{l}
1+\epsilon\left[R_{1}+\delta_{R 1}\left(x_{0}, x_{1}\right)\right]+\epsilon^{2} R_{2}+O\left(\epsilon^{3}\right), \quad(\mathrm{KdVB}), \\
1+\epsilon R_{1}+\epsilon^{2}\left[R_{2}+\delta_{R 2}\left(x_{0}, x_{1}, x_{2}\right)\right]+\epsilon^{3} R_{3}+O\left(\epsilon^{4}\right),
\end{array} \quad(\mathrm{NLS})\right. \\
\alpha / \alpha_{00} & =\left\{\begin{array}{l}
1+\epsilon\left[\alpha_{1}+\delta_{\alpha 1}\left(x_{0}, x_{1}\right)\right]+\epsilon^{2} \alpha_{2}+O\left(\epsilon^{3}\right), \quad(\mathrm{KdVB}), \\
1+\epsilon \alpha_{1}+\epsilon^{2}\left[\alpha_{2}+\delta_{\alpha 2}\left(x_{0}, x_{1}, x_{2}\right)\right]+\epsilon^{3} \alpha_{3}+O\left(\epsilon^{4}\right), \quad(\mathrm{NLS}),
\end{array}\right. \tag{28}
\end{align*}
$$

where $\delta_{R 1}, \delta_{R 2}, \delta_{\alpha 1}$, and $\delta_{\alpha 2}$ are initially given variables representing small initial nonuniformities of $R^{*}$ and $\alpha$, respectively, and $\alpha_{00}$ is the initial typical void fraction. For the monodisperse case (Kanagawa et al., 2010), $\delta_{R 1}, \delta_{R 2}, \delta_{\alpha 1}$, and $\delta_{\alpha 2}$ are equal to zero. The important difference between our previous polydisperse (Kanagawa et al., 2021a) and the present cases is that $\delta_{R 1}$ and $\delta_{\alpha 1}$ do not depend on $x_{0}$ in the KdVB equation, and $\delta_{R 2}$ and $\delta_{\alpha 2}$ do not depend on $x_{0}$ and $x_{1}$ in the NLS equation in our previous polydisperse case (Kanagawa et al., 2021a). From a practical viewpoint, a process of bubble production may cause manufacturing errors. Then, the polydispersity defined in (28) and (29) can be applied to express manufacturing errors, and various applications using microbubbles for medical use will also be expected.

The following relation among $R^{*}, \alpha$, and $n^{*}$ is imposed:
$\alpha=\frac{4}{3} \pi R^{* 3} n^{*}$.
We also express the polydispersity of the bubble number density, $\delta_{n 1}$ and $\delta_{n 2}$, by substituting (28) and (29) into (30) as follows:
$\left\{\begin{array}{l}n_{00}^{*}\left(1+\epsilon \delta_{n 1}+\epsilon^{2} \delta_{n 2}\right)=\frac{3 \alpha_{00}\left(1+\epsilon \delta_{\alpha 1}\right)}{4 \pi R_{00}^{* 3}\left(1+\epsilon \delta_{R 1}\right)^{3}}, \\ n_{00}^{*}\left(1+\epsilon^{2} \delta_{n 2}\right)=\frac{3 \alpha_{00}\left(1+\epsilon^{2} \delta_{\alpha 2}\right)}{4 \pi R_{00}^{* 3}\left(1+\epsilon^{2} \delta_{R 2}\right)^{3}}, \quad(\mathrm{KLS}) .\end{array}\right.$
From now on, $\delta_{R i}$ and $\delta_{\alpha i}(i=1,2)$ are explicitly used, and $\delta_{n 1}$ and $\delta_{n 2}$ do not appear. Furthermore, substituting (28) into (10) and (11) gives the expansions of gas pressure and gas density with initial nonuniformity,
respectively,

$$
\begin{align*}
p_{\mathrm{G}}^{*} / & \left(\rho_{L 0} U^{* 2}\right) \\
& =\left\{\begin{array}{l}
p_{\mathrm{G} 0}+\epsilon\left[p_{\mathrm{G} 1}+\delta_{p 1}(x)\right]+\epsilon^{2}\left[p_{\mathrm{G} 2}+\delta_{p 2}(x)+p_{\mathrm{G} 1} \frac{\delta_{p 1}(x)}{p_{\mathrm{G} 0}}\right] \\
\quad+O\left(\epsilon^{3}\right), \\
p_{\mathrm{G} 0}+\epsilon p_{\mathrm{G} 1}+\epsilon^{2}\left[\mathrm{KdVB}_{\mathrm{G} 2}+\delta_{p 2}(x)\right]+\epsilon^{3} p_{\mathrm{G} 3}+O\left(\epsilon^{4}\right),
\end{array}\right. \text { (NLS), } \tag{32}
\end{align*}
$$

where
$\delta_{p 1}=\gamma \delta_{\rho 1}, \quad(\mathrm{KdVB})$,
$\delta_{p 2}=-\left(\frac{\Delta^{2}}{\Omega^{2}}-3 \gamma p_{\mathrm{G} 0}\right) \delta_{R 1}^{2}, \quad(\mathrm{KdVB})$,
$\delta_{p 2}=\gamma \delta_{\rho 2}, \quad(\mathrm{NLS})$,
$\rho_{\mathrm{G}}^{*} / \rho_{\mathrm{G} 00}^{*}=\left\{\begin{array}{lr}1+\epsilon\left[\rho_{\mathrm{G} 1}+\delta_{\rho 1}(x)\right] \\ \quad+\epsilon^{2}\left[\rho_{\mathrm{G} 2}+\delta_{\rho 2}(x)+\rho_{\mathrm{G} 1} \delta_{\rho 1}(x)\right]+O\left(\epsilon^{3}\right), & (\mathrm{KdVB}), \\ 1+\epsilon \rho_{\mathrm{G} 1}+\epsilon^{2}\left[\rho_{\mathrm{G} 2}+\delta_{\rho 2}(x)\right]+\epsilon^{3} \rho_{\mathrm{G} 3}+O\left(\epsilon^{4}\right), & (\mathrm{NLS}),\end{array}\right.$
where
$\delta_{\rho 1}=\frac{1}{\gamma}\left(\frac{\Delta^{2}}{\Omega^{2}}-3 \gamma p_{\mathrm{G} 0}\right) \delta_{R 1}, \quad(\mathrm{KdVB})$,
$\delta_{\rho 2}=\frac{1}{\gamma}\left(\frac{\Delta^{2}}{\Omega^{2}}-3 \gamma p_{\mathrm{G} 0}\right)\left[\frac{\gamma-1}{2 \gamma}\left(\frac{\Delta^{2}}{\Omega^{2}}-3 \gamma p_{\mathrm{G} 0}\right)-1\right] \delta_{R 1}^{2}$,
(KdVB),
$\delta_{\rho 2}=\frac{\Delta^{2}-3 \gamma p_{\mathrm{G} 0}}{\gamma} \delta_{R 2}, \quad(\mathrm{NLS})$.

## 3. Derivation of KdVB equation for long waves

### 3.1. Linear propagation at near field

Substituting (13), (15) and (21)-(29), (32) and (36) into basic eqations (1)-(12) and equating each coefficient of like powers of $\epsilon$ in the set of resultant equations, we have a set of linear equations as the first-order equations:
$\frac{\partial \alpha_{1}}{\partial t_{0}}-3 \frac{\partial R_{1}}{\partial t_{0}}+\frac{\partial u_{\mathrm{G} 1}}{\partial x_{0}}=0$,
$\alpha_{00} \frac{\partial \alpha_{1}}{\partial t_{0}}-\left(1-\alpha_{00}\right) \frac{\partial u_{\mathrm{L} 1}}{\partial x_{0}}=0$,
$\beta_{1} \frac{\partial u_{\mathrm{G} 1}}{\partial t_{0}}-\beta_{1} \frac{\partial u_{\mathrm{L} 1}}{\partial t_{0}}-3 \gamma p_{\mathrm{G} 0} \frac{\partial R_{1}}{\partial x_{0}}+\frac{\partial \delta_{p 1}}{\partial x_{0}}=0$,
$\left(1-\alpha_{00}+\beta_{1} \alpha_{00}\right) \frac{\partial u_{\mathrm{L} 1}}{\partial t_{0}}-\beta_{1} \alpha_{00} \frac{\partial u_{\mathrm{G} 1}}{\partial t_{0}}+\left(1-\alpha_{00}\right) \frac{\partial p_{\mathrm{L} 1}}{\partial x_{0}}=0$,
$R_{1}+\frac{\Omega^{2}}{\Delta^{2}} p_{\mathrm{L} 1}=0$.
Removing $\alpha_{1}, u_{\mathrm{G} 1}, u_{\mathrm{L} 1}$, and $p_{\mathrm{L} 1}$ from (40)-(44), we have the linear wave equation for unknown $R_{1}$ :
$\frac{\partial^{2} R_{1}}{\partial t_{0}^{2}}-v_{\mathrm{p}}^{2} \frac{\partial^{2} R_{1}}{\partial x_{0}^{2}}=-\frac{1-\alpha_{00}+\beta_{1}}{3 \beta_{1}\left(1-\alpha_{00}\right)} \frac{\partial^{2} \delta_{p 1}}{\partial x_{0}^{2}}$,
where the phase velocity $v_{\mathrm{p}}$ is given by
$v_{\mathrm{p}}=\sqrt{\frac{3 \alpha_{00}\left(1-\alpha_{00}+\beta_{1}\right) \gamma p_{\mathrm{G} 0}+\beta_{1}\left(1-\alpha_{00}\right) \Delta^{2} / \Omega^{2}}{3 \beta_{1} \alpha_{00}\left(1-\alpha_{00}\right)}}$,
and the explicit form of $U^{*}$ is then determined as follows:
$U^{*}=\sqrt{\frac{3 \alpha_{00}\left(1-\alpha_{00}+\beta_{1}\right) \gamma p_{\mathrm{G} 00}^{*} / \rho_{\mathrm{L} 0}^{*}+\beta_{1}\left(1-\alpha_{00}\right) R_{0}^{* 2} \omega_{\mathrm{B}}^{* 2}}{3 \beta_{1} \alpha_{00}\left(1-\alpha_{00}\right) v_{\mathrm{p}}^{2}}}$,
note that $L^{*} \equiv U^{*} T^{*}=U^{*} / \omega^{*}$ is simultaneously determined. Now, we assume $v_{\mathrm{p}}=1$ for simplicity.

Hereafter, we focus on only the right-running wave $R_{1}=f\left(\varphi_{0}\right.$; $t_{1}, x_{1}$ ), and the phase function $\varphi_{0}$ is then defined as
$\varphi_{0}\left(t_{0}, x_{0}\right) \equiv x_{0}-t_{0}$.
The independent variables in (40)-(44) are rewritten using $\varphi_{0}$; all the first-order variables, i.e., $\alpha_{1}, u_{\mathrm{G} 1}, u_{\mathrm{L} 1}$, and $p_{\mathrm{L} 1}$ are expressed via $f\left(=R_{1}\right)$ :
$\alpha_{1}=s_{1} f, \quad u_{\mathrm{G} 1}=s_{2} f, \quad u_{\mathrm{L} 1}=s_{3} f, \quad p_{\mathrm{L} 1}=s_{4} f$,
with
$s_{4}=-\frac{\Delta^{2}}{\Omega^{2}}, \quad s_{1}=\frac{\left(1-\alpha_{00}\right)\left[3 \alpha_{00} \beta_{1}-\left(1-\alpha_{00}\right) s_{4}\right]}{\alpha_{00}\left(1-\alpha_{00}+\beta_{1}\right)}$,
$s_{2}=s_{1}-3, \quad s_{3}=-\frac{\alpha_{00} s_{1}}{1-\alpha_{00}}$.
The constants of integration are omitted because the boundary conditions at $x_{0} \rightarrow \infty$ (where the bubbly liquid is at rest) and $\partial \delta_{p 1} / \partial x_{0}$ are equal to zero because the conditions at $t_{0} \rightarrow \infty$ (when $\alpha_{1}, u_{\mathrm{G} 1}, u_{\mathrm{L} 1}$, and $p_{\mathrm{L} 1}$ ) should not be $\infty$.

The results of the leading order of approximation coincide with those of our monodisperse case (Kanagawa et al., 2010), and the initial polydispersity appears in the following order.

### 3.2. Nonlinear propagation at far field

We proceed with the approximation of $O\left(\epsilon^{2}\right)$, and the second-order set of equations is derived as
$\frac{\partial \alpha_{2}}{\partial t_{0}}-3 \frac{\partial R_{2}}{\partial t_{0}}+\frac{\partial u_{\mathrm{G} 2}}{\partial x_{0}}=\hat{K}_{1}$,
$\alpha_{00} \frac{\partial \alpha_{2}}{\partial t_{0}}-\left(1-\alpha_{00}\right) \frac{\partial u_{\mathrm{L} 2}}{\partial x_{0}}=\hat{K}_{2}$,
$\beta_{1} \frac{\partial u_{\mathrm{G} 2}}{\partial t_{0}}-\beta_{1} \frac{\partial u_{\mathrm{L} 2}}{\partial t_{0}}-3 \gamma p_{\mathrm{G} 0} \frac{\partial R_{2}}{\partial x_{0}}=\hat{K}_{3}$,
$\left(1-\alpha_{00}+\beta_{1} \alpha_{00}\right) \frac{\partial u_{\mathrm{L} 2}}{\partial t_{0}}-\beta_{1} \alpha_{00} \frac{\partial u_{\mathrm{G} 2}}{\partial t_{0}}+\left(1-\alpha_{00}\right) \frac{\partial p_{\mathrm{L} 2}}{\partial x_{0}}=\hat{K}_{4}$,
$R_{2}+\frac{\Omega^{2}}{\Delta^{2}} p_{\mathrm{L} 2}=\hat{K}_{5}$,
where the forms of inhomogeneous terms $\hat{K}_{i}(i=1,2,3,4,5)$ are explicitly presented in Appendix B, and new terms including polydispersity, $\delta_{R 1}$ and $\delta_{\alpha 1}$, appear in $\hat{K}_{i}$. Excluding the second-order variables except for $R_{2}$ from (51)-(55), we have the inhomogeneous wave equation as counterpart of (45):

$$
\begin{align*}
\frac{\partial^{2} R_{2}}{\partial t_{0}^{2}}-\frac{\partial^{2} R_{2}}{\partial x_{0}^{2}}= & \hat{K}\left(R_{1} ; \varphi_{0}, t_{1}, x_{1}\right) \\
= & -\frac{1}{3} \frac{\partial \hat{K}_{1}}{\partial t_{0}}+\frac{1}{3 \alpha_{00}} \frac{\partial \hat{K}_{2}}{\partial t_{0}}+\frac{1-\alpha_{00}+\beta_{1}}{3 \beta_{1}\left(1-\alpha_{00}\right)} \frac{\partial \hat{K}_{3}}{\partial x_{0}} \\
& +\frac{1}{3 \alpha_{00}\left(1-\alpha_{00}\right)} \frac{\partial \hat{K}_{4}}{\partial x_{0}}-\frac{\Delta^{2}}{3 \alpha_{00} \Omega^{2}} \frac{\partial^{2} \hat{K}_{5}}{\partial x_{0}^{2}} \tag{56}
\end{align*}
$$

Rewriting $\hat{K}_{i}$ using $\varphi_{0}$ and substituting the first-order variables, which are proportional to $R_{1}$ (see (49)), into $\hat{K}$ yields

$$
\begin{align*}
\hat{K}=2 \frac{\partial}{\partial \varphi_{0}}\left\{\frac{\partial R_{1}}{\partial t_{1}}\right. & +\frac{\partial R_{1}}{\partial x_{1}}+\left[\Pi_{0}+\hat{\Pi}_{4}\left(x_{0}, x_{1}\right)\right] \frac{\partial R_{1}}{\partial \varphi_{0}} \\
& +\Pi_{1} R_{1} \frac{\partial R_{1}}{\partial \varphi_{0}}+\Pi_{2} \frac{\partial^{2} R_{1}}{\partial \varphi_{0}^{2}}+\Pi_{3} \frac{\partial^{3} R_{1}}{\partial \varphi_{0}^{3}} \\
& \left.+\hat{\Pi}_{5}\left(x_{0}, x_{1}\right) R_{1}-\frac{1-\alpha_{00}+\beta_{1}}{6 \beta_{1}\left(1-\alpha_{00}\right)} \frac{\partial \delta_{p 1}}{\partial x_{1}}\right\} . \tag{57}
\end{align*}
$$

Now, we impose the solvability (i.e., non-secular) condition for (56) (Kanagawa et al., 2010; Jeffrey and Kawahara, 1982) to discuss the uniformly valid approximate solution $R_{1}$ (Kanagawa et al., 2010). From (21) and (48), the independent variables $\varphi_{0}, t_{1}$, and $x_{1}$ in (57) are
expressed by the transformed independent variables $\tau$ and $\xi$ (see (59) below), and we have the KdVB equation with a correction term:
$\frac{\partial R_{1}}{\partial \tau}+\Pi_{1} R_{1} \frac{\partial R_{1}}{\partial \xi}+\Pi_{2} \frac{\partial^{2} R_{1}}{\partial \xi^{2}}+\Pi_{3} \frac{\partial^{3} R_{1}}{\partial \xi^{3}}+\hat{\Pi}_{5} R_{1}-\frac{1}{\epsilon} \frac{1-\alpha_{00}+\beta_{1}}{6 \beta_{1}\left(1-\alpha_{00}\right)} \frac{\partial \delta_{p 1}}{\partial \xi}=0$,
$\tau \equiv \epsilon t, \quad \xi \equiv x-\left\{1+\epsilon\left[\Pi_{0}+\hat{\Pi}_{4}(x)\right]\right\} t$,
where $\Pi_{i}(i=0,1,2,3)$ is the constant coefficient, and $\hat{\Pi}_{j}(j=4,5)$ is the variable coefficient containing $\delta_{R 1}$ and $\delta_{\alpha 1}$. Here, $\Pi_{0}, \Pi_{1}, \Pi_{2}$, and $\Pi_{3}$ in (58) and (59) are the same as those in our previous monodisperse results (Kanagawa et al., 2010), and explicit forms are omitted from this paper.

The coefficients $\hat{\Pi}_{4}$ and $\hat{\Pi}_{5}$ in (58) and (59) do not appear in our previous monodisperse result (Kanagawa et al., 2010) and are given as
$\hat{\Pi}_{4}=\hat{\Pi}_{41} \delta_{R 1}+\hat{\Pi}_{42} \delta_{\alpha 1}$,
where
$\hat{\Pi}_{41}=1+\frac{s_{1}}{3} \frac{\delta_{\rho 1}}{\delta_{R 1}}-\frac{\delta_{\rho 1}}{\delta_{R 1}}-\frac{s_{2}}{3} \frac{\delta_{\rho 1}}{\delta_{R 1}}+\frac{1-\alpha_{00}+\beta_{1}}{\beta_{1}\left(1-\alpha_{00}\right)} \gamma\left(\frac{\delta_{p 1}}{\delta_{R 1}}-p_{\mathrm{G} 0}\right)$

$$
\begin{equation*}
-\frac{1}{3 \alpha_{00}}\left[3 \gamma p_{\mathrm{G} 0}+(3 \gamma-2)\left(3 \gamma p_{\mathrm{G} 0}-\frac{\Delta^{2}}{\Omega^{2}}\right)\right] \tag{61}
\end{equation*}
$$

$\hat{\Pi}_{42}=-1+\frac{1-\alpha_{00}+\beta_{1}}{\beta_{1}\left(1-\alpha_{00}\right)} \gamma p_{\mathrm{G} 0}+\frac{s_{4}-s_{3}}{3\left(1-\alpha_{0}\right)}$,
$\hat{\Pi}_{5}=\hat{\Pi}_{50} \frac{\partial \delta_{\alpha 1}}{\partial x_{0}}$,
where
$\hat{\Pi}_{50}=\frac{1}{2}\left[\frac{1-\alpha_{00}+\beta_{1}}{\beta_{1}\left(1-\alpha_{00}\right)} \gamma p_{\mathrm{G} 0}+\frac{s_{4}-s_{3}}{3\left(1-\alpha_{00}\right)}\right]$,
where $\hat{\Pi}_{41}, \hat{\Pi}_{42}$, and $\hat{\Pi}_{50}$ are constants, and $s_{i}(i=1,2,3,4)$ are presented in Appendix A. The coefficients $\hat{\Pi}_{4}$ and $\hat{\Pi}_{5}$ include $\delta_{R 1}$ and $\delta_{\alpha 1}$, and they are due to the polydispersity.

## 4. Derivation of NLS equation for short waves

### 4.1. Linear propagation of carrier waves at a near field

As in the previous case of (40)-(44), we have the following set of linear equations:
$\frac{\partial \alpha_{1}}{\partial t_{0}}-3 \frac{\partial R_{1}}{\partial t_{0}}+\frac{\partial u_{\mathrm{G} 1}}{\partial x_{0}}=0$,
$\alpha_{00} \frac{\partial \alpha_{1}}{\partial t_{0}}-\left(1-\alpha_{00}\right) \frac{\partial u_{\mathrm{L} 1}}{\partial x_{0}}=0$,
$\beta_{1} \frac{\partial u_{\mathrm{G} 1}}{\partial t_{0}}-\beta_{1} \frac{\partial u_{\mathrm{L} 1}}{\partial t_{0}}-3 \gamma p_{\mathrm{G} 0} \frac{\partial R_{1}}{\partial x_{0}}=0$,
$\left(1-\alpha_{00}+\beta_{1} \alpha_{00}\right) \frac{\partial u_{\mathrm{L} 1}}{\partial t_{0}}-\beta_{1} \alpha_{00} \frac{\partial u_{\mathrm{G} 1}}{\partial t_{0}}+\left(1-\alpha_{00}\right) \frac{\partial p_{\mathrm{L} 1}}{\partial x_{0}}=0$,
$\frac{\partial^{2} R_{1}}{\partial t_{0}^{2}}+R_{1}+\frac{p_{\mathrm{L} 1}}{\Delta^{2}}=0$.
The difference between the KdVB and NLS cases is the form of the linearized Keller equation (i.e., (44) and (69)). In the NLS case, the dispersion effect is stronger than that in the KdVB case, and the dispersion effect appears in the leading order of approximation (i.e., the second-order derivative of $R_{1}$ ).

By combining five equations (65)-(69) into a single equation, the five unknown variables in (65)-(69) are reduced to only $R_{1}$ :
$\mathcal{L}_{1}\left[R_{1}\right]=0, \quad \mathcal{L}_{1} \equiv \frac{\partial^{2}}{\partial t_{0}^{2}}-\left[\frac{\Delta^{2}}{3 \alpha_{00}}+\frac{\left(1-\alpha_{0}+\beta_{1}\right) \gamma p_{\mathrm{G} 0}}{\beta_{1}\left(1-\alpha_{00}\right)}\right] \frac{\partial^{2}}{\partial x_{0}^{2}}-\frac{\Delta^{2}}{3 \alpha_{00}} \frac{\partial^{4}}{\partial t_{0}^{2} \partial x_{0}^{2}}$.

The fourth order derivative of (70) represents the dispersion term owing to the second-order derivative (i.e., acceleration of the bubble wall) in (69), which did not appear in the near field in the KdVB case (see (44)). We now assume the form of the solution of (70) as a quasi-monochromatic wave train evolving into a slowly modulated wave packet:
$R_{1}=A\left(t_{1}, t_{2}, x_{1}, x_{2}\right) \mathrm{e}^{\mathrm{i} \theta}+$ c.c.,
where
$\theta=k x_{0}-\Omega(k) t_{0}$,
where $A$ is the slowly varying complex amplitude depending only on slow-scale variables (i.e., $t_{1}, t_{2}, x_{1}$, and $x_{2}$ ) and i represents the imaginary unit, c.c. represents the complex conjugate. $A$ is clearly constant in the near field denoted by fast scale variables (i.e., $t_{0}$ and $x_{0}$ ). Substituting (71) into (65)-(69) gives
$\alpha_{1}=b_{1} R_{1}, \quad u_{\mathrm{G} 1}=b_{2} R_{1}, \quad u_{\mathrm{L} 1}=b_{3} R_{1}, \quad p_{\mathrm{L} 1}=b_{4} R_{1}$,
with
$b_{4}=\Delta^{2}\left(\Omega^{2}-1\right), \quad b_{1}=\frac{\left(1-\alpha_{00}\right)\left[3 \beta_{1} \alpha_{00}-\left(1-\alpha_{00}\right) b_{4} k^{2} / \Omega^{2}\right]}{\alpha_{1}\left(1-\alpha_{00}+\beta_{1}\right)}$,
$b_{2}=\left(b_{1}-3\right) \frac{\Omega}{k}, \quad b_{3}=-\frac{\alpha_{00} b_{1} \Omega}{\left(1-\alpha_{00}\right) k}$,
where $\Omega$ depends on $k$ through a linear dispersion relation
$D \equiv \frac{\Delta^{2} k^{2}\left(1-\Omega^{2}\right)}{3 \alpha_{00}}+\frac{\left(1-\alpha_{00}+\beta_{1}\right) \gamma p_{\mathrm{G} 0}}{\beta_{1}\left(1-\alpha_{00}\right)} k^{2}-\Omega^{2}=0$,
or
$\Omega= \pm k \sqrt{\frac{\Delta^{2}}{3 \alpha_{00}+\Delta^{2} k^{2}}+\frac{3 \alpha_{00}\left(1-\alpha_{00}+\beta_{1}\right) \gamma p_{\mathrm{G} 0}}{\beta_{1}\left(1-\alpha_{00}\right)\left(3 \alpha_{00}+\Delta^{2} k^{2}\right)}}$.
The positive $\Omega$ in (76) corresponds to the right-running carrier wave. The nondimensional phase velocity $v_{\mathrm{p}}$ and group velocity $v_{\mathrm{g}}$ are immediately obtained as follows:
$v_{\mathrm{p}}=\frac{\Omega}{k}, \quad v_{\mathrm{g}}=\frac{\mathrm{d} \Omega}{\mathrm{d} k}=\frac{3 \alpha_{00} \Omega}{k\left(3 \alpha_{00}+\Delta^{2} k^{2}\right)}$.
Imposing $v_{\mathrm{p}}=1$ under $\Omega=1$, the explicit form of $U^{*}$ is determined as (Kanagawa et al., 2010)
$U^{*}=\sqrt{\frac{\left(1-\alpha_{00}+\beta_{1}\right) \gamma p_{\mathrm{G} 0}^{*}}{\beta_{1}\left(1-\alpha_{00}\right) \rho_{\mathrm{L} 0}^{*}}}$.
Note that $L \equiv U^{*} T^{*}=U^{*} / \omega_{\mathrm{B}}^{*}$ is simultaneously determined (see (17)). The initial polydispersity does not affect the results of the approximation in near field.

### 4.2. Linear propagation of envelopes at a far field I

The set of equations as the second-order equations is derived as follows:
$\frac{\partial \alpha_{2}}{\partial t_{0}}-3 \frac{\partial R_{2}}{\partial t_{0}}+\frac{\partial u_{\mathrm{G} 2}}{\partial x_{0}}=M_{1}$,
$\alpha_{00} \frac{\partial \alpha_{2}}{\partial t_{0}}-\left(1-\alpha_{00}\right) \frac{\partial u_{\mathrm{L} 2}}{\partial x_{0}}=M_{2}$,
$\beta_{1} \frac{\partial u_{\mathrm{G} 2}}{\partial t_{0}}-\beta_{1} \frac{\partial u_{\mathrm{L} 2}}{\partial t_{0}}-3 \gamma p_{\mathrm{G} 0} \frac{\partial R_{2}}{\partial x_{0}}=M_{3}-\frac{\partial \delta_{p 2}}{\partial x_{0}}$,
$\left(1-\alpha_{00}+\beta_{1} \alpha_{00}\right) \frac{\partial u_{\mathrm{L} 2}}{\partial t_{0}}-\beta_{1} \alpha_{00} \frac{\partial u_{\mathrm{G} 2}}{\partial t_{0}}+\left(1-\alpha_{00}\right) \frac{\partial p_{\mathrm{L} 2}}{\partial x_{0}}=M_{4}$,
$\frac{\partial^{2} R_{2}}{\partial t_{0}^{2}}+R_{2}+\frac{p_{\mathrm{L} 2}}{\Delta^{2}}=M_{5}$,
where $M_{i}(i=1,2,3,4,5)$ are in the same form in our original monodisperse case (Kanagawa et al., 2010). The reduction of (79)-(83) to a single equation for $R_{2}$ is performed as follows:
$\mathcal{L}_{1}\left[R_{2}\right]=-\frac{1}{3} \frac{\partial M_{1}}{\partial t_{0}}+\frac{1}{3 \alpha_{00}} \frac{\partial M_{2}}{\partial t_{0}}+\frac{1-\alpha_{00}+\beta_{1}}{3 \beta_{1}\left(1-\alpha_{00}\right)} \frac{\partial M_{3}}{\partial x_{0}}$

$$
\begin{align*}
& +\frac{1}{3 \alpha_{00}\left(1-\alpha_{00}\right)} \frac{\partial M_{4}}{\partial x_{0}}-\frac{\Delta^{2}}{3 \alpha_{00}} \frac{\partial^{2} M_{5}}{\partial x_{0}^{2}}-\frac{1-\alpha_{00}+\beta_{1}}{3 \beta_{1}\left(1-\alpha_{00}\right)} \frac{\partial \delta_{p 2}}{\partial x_{0}} \\
= & \Gamma A^{2} \mathrm{e}^{2 \mathrm{i} \theta}+\mathrm{i}\left(-\frac{\partial D}{\partial \Omega}\right)\left(\frac{\partial A}{\partial t_{1}}+v_{\mathrm{g}} \frac{\partial A}{\partial x_{1}}\right) \mathrm{e}^{\mathrm{i} \theta} \\
& + \text { c.c. }-\frac{1-\alpha_{00}+\beta_{1}}{3 \beta_{1}\left(1-\alpha_{00}\right)} \frac{\partial \delta_{p 2}}{\partial x_{0}}, \tag{84}
\end{align*}
$$

where $\Gamma$ is the same form in our original monodisperse case (Kanagawa et al., 2010). From the solvability condition for (84), the coefficient of $\mathrm{e}^{\mathrm{i} \theta}$ on the right-hand side of (84) must be zero. Thus, we have
$\frac{\partial A}{\partial t_{1}}+v_{\mathrm{g}} \frac{\partial A}{\partial x_{1}}=0$.
Using (85) to (84), $R_{2}$ is considered such that the coefficients other than the term of $\mathrm{e}^{2 \mathrm{i} \theta}$ on the right-hand side are zero, a uniformly valid solution up to the far field of (86) is obtained:
$R_{2}=c_{0} A \mathrm{e}^{2 \mathrm{i} \theta}+$ c.c..
Substituting (86) into (79)-(83) yields the following:
$\left(\begin{array}{c}\alpha_{2} \\ u_{\mathrm{G} 2} \\ u_{\mathrm{L} 2} \\ p_{\mathrm{L} 2}\end{array}\right)=\left(\begin{array}{lll}c_{1} & d_{1} & 0 \\ c_{2} & d_{2} & 0 \\ c_{3} & d_{3} & 0 \\ c_{4} & d_{4} & c_{\mathrm{s}}\end{array}\right)\left(\begin{array}{c}A^{2} \mathrm{e}^{2 \mathrm{i} \theta}+\text { c.c. } \\ \mathrm{i}\left(\partial A / \partial t_{1}\right) \mathrm{e}^{\mathrm{i} \theta}+\mathrm{c} . \mathrm{c} . \\ \left|A^{2}\right|\end{array}\right)$,
where the explicit forms of $c_{0}, c_{i}, d_{i}(i=1,2,3,4,5)$, and $c_{\mathrm{s}}$ are the same as those in our monodisperse case (Kanagawa et al., 2010).

As in the leading order of approximation in 4.1, the forms of independent valuables of the second-order approximation in this section coincides with our original monodisperse case (Kanagawa et al., 2010). The effect of initial polydispersity appears next order of approximation.

### 4.3. Nonlinear propagation of envelope waves at a far field II

The third-order of approximation gives
$\frac{\partial \alpha_{3}}{\partial t_{0}}-3 \frac{\partial R_{3}}{\partial t_{0}}+\frac{\partial u_{\mathrm{G} 3}}{\partial x_{0}}=\hat{N}_{1}$,
$\alpha_{00} \frac{\partial \alpha_{3}}{\partial t_{0}}-\left(1-\alpha_{00}\right) \frac{\partial u_{\mathrm{L} 3}}{\partial x_{0}}=\hat{N}_{2}$,
$\beta_{1} \frac{\partial u_{\mathrm{G} 3}}{\partial t_{0}}-\beta_{1} \frac{\partial u_{\mathrm{L} 3}}{\partial t_{0}}-3 \gamma p_{\mathrm{G} 0} \frac{\partial R_{3}}{\partial x_{0}}=\hat{N}_{3}$,
$\left(1-\alpha_{00}+\beta_{1} \alpha_{00}\right) \frac{\partial u_{\mathrm{L} 3}}{\partial t_{0}}-\beta_{1} \alpha_{00} \frac{\partial u_{\mathrm{G} 3}}{\partial t_{0}}+\left(1-\alpha_{00}\right) \frac{\partial p_{\mathrm{L} 3}}{\partial x_{0}}=\hat{N}_{4}$,
$\frac{\partial^{2} R_{3}}{\partial t_{0}^{2}}+R_{3}+\frac{p_{\mathrm{L} 3}}{\Delta^{2}}=\hat{N}_{5}$,
where the explicit forms of the inhomogeneous terms $\hat{N}_{i}(i=1,2,3,4,5)$ are quite complex, as presented in Appendix B. The initial polydispersity (i.e., $\delta_{R 2}$ and $\delta_{\alpha 2}$ ) appears in $\hat{N}_{i}$.

The single inhomogeneous equation for $R_{3}$ is
$\mathcal{L}_{1}\left[R_{3}\right]=\Lambda_{1} \mathrm{e}^{\mathrm{i} \mathrm{i} \theta}+\Lambda_{2} \mathrm{e}^{2 \mathrm{i} \theta}+\hat{\Lambda}_{3} \mathrm{e}^{\mathrm{i} \theta}+$ c.c. $-\frac{1-\alpha_{0}+\beta_{1}}{3 \beta_{1}\left(1-\alpha_{0}\right)} \frac{\partial \delta_{p 2}}{\partial x_{1}}$,
where $\Lambda_{i}(i=1,2,3)$ are the complex variables composed of $A$. Here, $\Lambda_{1}$ and $\Lambda_{2}$ are not shown because they are not essential to derive the NLS equation; $\hat{\Lambda}_{3}$ is given as

$$
\begin{align*}
\hat{\Lambda}_{3}=\left(-\frac{\partial D}{\partial \Omega}\right)\{ & \mathrm{i}\left(\frac{\partial A}{\partial t_{2}}+v_{\mathrm{g}} \frac{\partial A}{\partial x_{2}}\right)+\frac{1}{2} \frac{\mathrm{~d} v_{\mathrm{g}}}{\mathrm{~d} k} \frac{\partial^{2} A}{\partial x_{1}^{2}}+v_{1}|A|^{2} A \\
& \left.+\mathrm{i}\left[v_{2}+\hat{v}_{4}\left(x_{0}, x_{1}, x_{2}\right)\right] A+\hat{v}_{3}\left(x_{0}, x_{1}, x_{2}\right) A\right\} \tag{94}
\end{align*}
$$

where $D$ is shown in (75), and should be zero under the non-secular condition for (93). Using (21) and (70), we rewrite (94) as the following NLS equation with a correction term:
$\mathrm{i} \frac{\partial A}{\partial \tau}+\frac{1}{2} \frac{\mathrm{~d} v_{\mathrm{g}}}{\mathrm{d} k} \frac{\partial^{2} A}{\partial \xi^{2}}+v_{1}|A|^{2} A+\mathrm{i}\left(v_{2}+\hat{v}_{4}\right) A=0$,
via a variable transformation
$\tau \equiv \epsilon^{2} t, \quad \xi \equiv \epsilon\left(x-v_{\mathrm{g}} t\right)+\epsilon^{2} \frac{\hat{\imath}_{3}}{K}$,
where $K$ is the non-dimensional wave number of the envelope wave, $v_{1}$ and $v_{2}$ are the constant coefficients, and $\hat{v}_{3}$ and $\hat{v}_{4}$ are the variable coefficients, including $\delta_{R 2}$ and $\delta_{\alpha 2}$. Here, $v_{1}$ and $v_{2}$ in (95) are the same as those in our previous monodisperse results (Kanagawa et al., 2010), and explicit forms are omitted in this paper.

The coefficients $\hat{\nu}_{3}$ and $\hat{v}_{4}$ in (95) and (96) do not appear in our previous monodisperse result (Kanagawa et al., 2010) and are given as
$\hat{v}_{3}=\hat{v}_{31} \delta_{R 2}+\hat{v}_{32} \delta_{\alpha 2}+\hat{v}_{33} \frac{\partial^{2} \delta_{\alpha 2}}{\partial x_{0}^{2}}$,
where

$$
\begin{align*}
\left(-\frac{\partial D}{\partial \Omega}\right) \hat{v}_{31}= & \left(\frac{\delta_{\rho 2}}{\delta_{R 2}}-1-\frac{b_{1}}{3} \frac{\delta_{\rho 2}}{\delta_{R 2}}\right) \Omega^{2}+\frac{1}{3} b_{2} k \Omega \frac{\delta_{\rho 2}}{\delta_{R 2}} \\
& -\frac{\left(1-\alpha_{00}+\beta_{1}\right) \gamma k^{2}}{\beta_{1}\left(1-\alpha_{00}\right)}\left(\frac{\delta_{p 2}}{\delta_{R 2}}-p_{\mathrm{G} 0}\right) \\
& +\frac{k^{2}}{3 \alpha_{00}}\left[3 \gamma p_{\mathrm{G} 0}(3 \gamma-1)-(3 \gamma-2) \Delta^{2}+\Delta^{2} \Omega^{2}\right],  \tag{98}\\
\left(-\frac{\partial D}{\partial \Omega}\right) \hat{\nu}_{32}= & \Omega^{2}-\frac{1-\alpha_{00}+\beta_{1}}{\beta_{1}\left(1-\alpha_{00}\right)} \gamma p_{\mathrm{G} 0} k^{2}+\frac{b_{3} \Omega-b_{4} k}{3\left(1-\alpha_{00}\right)} k,  \tag{99}\\
\left(-\frac{\partial D}{\partial \Omega}\right) \hat{v}_{33}= & \frac{\Delta^{2} \Omega^{2}}{3\left(1-\alpha_{00}\right)},  \tag{100}\\
\hat{v}_{4}= & \hat{v}_{40} \frac{\partial \delta_{\alpha 2}}{\partial x_{0}}, \tag{101}
\end{align*}
$$

where
$\left(-\frac{\partial D}{\partial \Omega}\right) \hat{\nu}_{40}=\left[\frac{1-\alpha_{00}+\beta_{1}}{\beta_{1}\left(1-\alpha_{00}\right)} \gamma p_{\mathrm{G} 0} k+\frac{\Delta^{2} \Omega^{2} k+b_{4} k-b_{3} \Omega}{3\left(1-\alpha_{00}\right)}\right]$,
where $\hat{\nu}_{3 i}(i=1,2,3)$ and $\hat{v}_{40}$ are constants. The coefficients $\hat{v}_{3}$ and $\hat{v}_{4}$ include $\delta_{R 2}$ and $\delta_{\alpha 2}$, and they are due to the polydispersity as in $\hat{\Pi}_{4}$ and $\hat{\Pi}_{5}$ in (58).

## 5. Effect of polydispersity

Again, let us show the resultant equations derived in the previous sections: KdVB equation,
$\frac{\partial R_{1}}{\partial \tau}+\Pi_{1} R_{1} \frac{\partial R_{1}}{\partial \xi}+\Pi_{2} \frac{\partial^{2} R_{1}}{\partial \xi^{2}}+\Pi_{3} \frac{\partial^{3} R_{1}}{\partial \xi^{3}}+\hat{\Pi}_{5} R_{1}-\frac{1}{\epsilon} \frac{1-\alpha_{00}+\beta_{1}}{6 \beta_{1}\left(1-\alpha_{00}\right)} \frac{\partial \delta_{p 1}}{\partial \xi}=0$,
$\tau \equiv \epsilon t, \quad \xi \equiv x-\left\{1+\epsilon\left[\Pi_{0}+\hat{\Pi}_{4}(x)\right]\right\} t$,
and NLS equation,
$\mathrm{i} \frac{\partial A}{\partial \tau}+\frac{1}{2} \frac{\mathrm{~d} v_{\mathrm{g}}}{\mathrm{d} k} \frac{\partial^{2} A}{\partial \xi^{2}}+v_{1}|A|^{2} A+\mathrm{i}\left(v_{2}+\hat{v}_{4}\right) A=0$,
$\tau \equiv \epsilon^{2} t, \quad \xi \equiv \epsilon\left(x-v_{\mathrm{g}} t\right)+\epsilon^{2} \frac{\hat{v}_{3}}{K}$.
Both the KdVB equation (58) and NLS equation (95) describe the weakly nonlinear propagation of waves. They are composed of a linear combination of terms representing the three effects (i.e., nonlinear, dissipation, and dispersion effects), and the sixth term of (58) is the inhomogeneous term.

In the following, we discuss the physical meaning of each term in detail. The second term of (58) and the third term of (95) are nonlinear terms and represent the size of the nonlinear effect. Deriving the linear dispersion relation of (58) and (95) reveals the physical meanings of other terms. The third and fifth terms of (58) and the fourth term of (95) represent the dissipation effect. Here, $\Pi_{2}$ and $v_{2}$ are related to the liquid viscosity, and $\hat{\Pi}_{5}$ and $\hat{\nu}_{4}$ the polydispersity; $\hat{\Pi}_{5}$ and $\hat{\nu}_{4}$ did not appear in our previous polydisperse result (Kanagawa et al., 2021a).


Fig. 3. Dependence of the constant dissipation coefficient $\hat{\Pi}_{50}$ in (64) on the initial void fraction $\alpha_{00}$ for the case of $\Omega=1, \sqrt{\epsilon}=0.15, \gamma=1.4, \beta_{1}=0.5, R_{00}^{*}=10 \mu \mathrm{~m}$, and the normal condition of the air-water system.

The fourth term of (58) and the second term of (95) represent the dispersion effect. The coefficients $\hat{I}_{4}$ and $\hat{v}_{3}$ in the moving coordinate $\xi$ in (59) and (96) represent the advection effect of waves due to the polydispersity. Hence, polydispersity contributes $\hat{\Pi}_{4}$ and $\hat{\Pi}_{5}$ in (58) and $\hat{v}_{3}$ and $\hat{v}_{4}$ in (95) and affects the dissipation and advection effects of waves.

The advection effect only moves the wave and does not essentially affect the waveform (see (59) and (96)). The dissipation coefficients $\hat{\Pi}_{5}$ and $\hat{v}_{4}$ are expressed by using $\partial \delta_{\alpha} / \partial x_{0}$, where $\delta_{\alpha}$ represents the polydispersity of the void fraction.

$$
\begin{align*}
\hat{\Pi}_{5} & =\hat{\Pi}_{50} \frac{\partial \delta_{\alpha 1}}{\partial x_{0}},  \tag{63}\\
\hat{\Pi}_{50} & =\frac{1}{2}\left[\frac{1-\alpha_{00}+\beta_{1}}{\beta_{1}\left(1-\alpha_{00}\right)} \gamma p_{\mathrm{G} 0}+\frac{s_{4}-s_{3}}{3\left(1-\alpha_{00}\right)}\right],  \tag{64}\\
\hat{v}_{4} & =\hat{v}_{40} \frac{\partial \delta_{\alpha 2}}{\partial x_{0}},  \tag{101}\\
\left(-\frac{\partial D}{\partial \Omega}\right) \hat{v}_{40} & =\left[\frac{1-\alpha_{00}+\beta_{1}}{\beta_{1}\left(1-\alpha_{00}\right)} \gamma p_{\mathrm{G} 0} k+\frac{\Delta^{2} \Omega^{2} k+b_{4} k-b_{3} \Omega}{3\left(1-\alpha_{00}\right)}\right] . \tag{102}
\end{align*}
$$

The dependences of each coefficient in (64) and (102) on the initial void fraction $\alpha_{00}$ and the wavenumber $k$ are shown in Figs. 3 and 4, respectively. Here, $\hat{\Pi}_{50}$ and $\hat{\nu}_{40}$ are always positive. Importantly, the positive and/or negative of $\hat{\Pi}_{5}$ and $\hat{\nu}_{4}$ depends on the value of the slope $\partial \delta_{\alpha} / \partial x_{0}$. As can be seen in the dissipation terms in (58) and (95), the larger the coefficients $\hat{\Pi}_{5}$ and $\hat{\nu}_{4}$, the stronger the dissipation effect. The dissipation effect is strong where the polydispersity of the void fraction is larger and is weak where it is smaller. Hence, the magnitude of the dissipation effect can be controlled by the value of the initial void fraction.

## 6. Conclusions

We theoretically investigated the propagation of nonlinear pressure waves in a liquid containing numerous gas bubbles and particularly focused on the initial small polydispersity of bubble size and bubble number density. Polydispersity exists in all of the regions; it is introduced into the expansion of the void fraction and the bubble radius without determining the explicit form and appears in the expansions of the gas density and pressure. Thus, we derived two nonlinear wave equations (i.e., KdVB (58) and NLS (95)), including terms to represent the polydispersity using the basic equations based on the two-fluid model.


Fig. 4. Dependence of the constant dissipation coefficient $\hat{v}_{40}$ on (a) the wavenumber $k$, and dependence of $\hat{v}_{40}$ on (b) the initial void fraction $\alpha_{00}$ for the case of $\epsilon=0.07$, $\gamma=1.4, \beta_{1}=0.5, R_{00}^{*}=10 \mu \mathrm{~m}$, and the normal condition of the air-water system.

First, we clarified that the polydispersity contributes to the dissipation and advection effects of waves and accordingly induces variable coefficients in the KdVB and NLS equations; the dissipation terms owing to the polydispersity do not appear in our previous polydisperse study (Kanagawa et al., 2021a). Since the dissipation effect becomes smaller in a field such that the void fraction decreases, the dissipation effect can be artificially changed by controlling the value of the void fraction. Second, we successfully incorporated the dependence of the initial void fraction on every coefficient in the KdVB and NLS equations owing to the use of the two-fluid model. These results highlight the effect of the initial polydispersity on weakly nonlinear waves in bubbly liquid.

In a forthcoming paper, we will apply the present theory to physicomathematical modelling of potential applications such as a medical ultrasound enhanced by microbubbles. Especially, ultrasound-contrastagent as microbubbles coated by a visco-elastic shell (e.g., Kanagawa et al., 2023; Kikuchi et al., 2023) and thermal ablation of tumor by using microbubble enhanced focused ultrasound (e.g., Kagami and Kanagawa, 2022) will be developed based on the present model for polydisperse bubbly liquids.

## CRediT authorship contribution statement

Takuma Kawame: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Software, Visualization, Validation,

Writing - original draft, Writing - review \& editing. Tetsuya Kanagawa: Conceptualization, Methodology, Formal analysis, Investigation, Supervision, Funding acquisition, Project administration, Validation, Writing - original draft, Writing - review \& editing.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Tetsuya Kanagawa reports financial support was provided by University of Tsukuba.

## Data availability

Data will be made available on request.

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## Appendix A. Explicit forms of $\hat{K}_{i}$

Explicit forms of the inhomogeneous terms $\hat{K}_{i}(i=1,2,3,4,5)$ in (51)-(55) are given by
$\hat{K}_{1}=K_{1}+\left(3 \delta_{\alpha 1}-3 \delta_{R 1}-s_{1} \delta_{\rho 1}+3 \delta_{\rho 1}\right) \frac{\partial R_{1}}{\partial t_{0}}$

$$
\begin{equation*}
-\left(\delta_{\alpha 1}+\delta_{\rho 1}\right) s_{2} \frac{\partial R_{1}}{\partial x_{0}}-s_{2} \frac{\partial \delta_{\alpha 1}}{\partial x_{0}} R_{1}, \tag{A.1}
\end{equation*}
$$

$\hat{K}_{2}=K_{2}-\alpha_{00} s_{3} \delta_{\alpha 1} \frac{\partial R_{1}}{\partial x_{0}}-\alpha_{00} s_{3} \frac{\partial \delta_{\alpha 1}}{\partial x_{0}} R_{1}$,
$\hat{K}_{3}=K_{3}-\beta_{1}\left(s_{2}-s_{3}\right) \delta_{\alpha 1} \frac{\partial R_{1}}{\partial t_{0}}+3 \gamma\left(\delta_{p 1}-p_{\mathrm{G} 0} \delta_{R 1}+p_{\mathrm{G} 0} \delta_{\alpha 1}\right) \frac{\partial R_{1}}{\partial x_{0}}-\frac{\partial \delta_{p 1}}{\partial x_{1}}$,
$\hat{K}_{4}=K_{4}+\alpha_{00}\left[\beta_{1}\left(s_{2}-s_{3}\right)+s_{3}\right] \delta_{\alpha 1} \frac{\partial R_{1}}{\partial t_{0}}+\alpha_{00} s_{4} \delta_{\alpha 1} \frac{\partial R_{1}}{\partial x_{0}}$,
$\hat{K}_{5}=K_{5}+\frac{\Omega^{2}}{\Delta^{2}}\left[3 \gamma p_{\mathrm{G} 0}+(3 \gamma-2)\left(3 \gamma p_{\mathrm{G} 0}-\frac{\Delta^{2}}{\Omega^{2}}\right)\right] R_{1} \delta_{R 1}$,
where $K_{i}(i=1,2,3,4,5)$ is the original inhomogeneous term for the monodisperse case (Kanagawa et al., 2010).

## Appendix B. Explicit forms of $\hat{N}_{i}$

Explicit forms of the inhomogeneous terms $\hat{N}_{i}(i=1,2,3,4,5)$ in (88)-(92) are given by
$\hat{N}_{1}=N_{1}+\left(3 \delta_{\alpha 2}+3 \delta_{\rho 2}-3 \delta_{R 2}-b_{1} \delta_{\rho 2}\right) \frac{\partial R_{1}}{\partial t_{0}}$

$$
\begin{equation*}
-b_{2}\left(\delta_{\alpha 2}+\delta_{\rho 2}\right) \frac{\partial R_{1}}{\partial x_{0}}-b_{2} \frac{\partial \delta_{\alpha 2}}{\partial x_{0}} R_{1} \tag{B.1}
\end{equation*}
$$

$\hat{N}_{2}=N_{2}-\alpha_{00} b_{3} \delta_{\alpha 2} \frac{\partial R_{1}}{\partial x_{0}}-\alpha_{00} b_{3} \frac{\partial \delta_{\alpha 2}}{\partial x_{0}} R_{1}$,
$\hat{N}_{3}=N_{3}-\beta_{1}\left(b_{2}-b_{3}\right) \delta_{\alpha 2} \frac{\partial R_{1}}{\partial t_{0}}$

$$
\begin{equation*}
+3 \gamma\left(\delta_{p 2}+p_{\mathrm{G} 0} \delta_{\alpha 2}-p_{\mathrm{G} 0} \delta_{R 2}\right) \frac{\partial R_{1}}{\partial x_{0}}-\frac{\partial \delta_{p 2}}{\partial x_{1}}, \tag{B.3}
\end{equation*}
$$

$\hat{N}_{4}=N_{4}+\alpha_{00}\left[\beta_{1}\left(b_{2}-b_{3}\right)+b_{3}\right] \delta_{\alpha 2} \frac{\partial R_{1}}{\partial t_{0}}$

$$
\begin{gather*}
+\alpha_{00} b_{4} \delta_{\alpha 2} \frac{\partial R_{1}}{\partial x_{0}}+\alpha_{00} \Delta^{2} \Omega^{2} \frac{\partial \delta_{\alpha 2}}{\partial x_{0}} R_{1},  \tag{B.4}\\
\hat{N}_{5}=  \tag{B.5}\\
N_{5}+\left[\frac{3 \gamma p_{\mathrm{G} 0}(3 \gamma-1)}{\Delta^{2}}-(3 \gamma-2)+\Omega^{2}\right] \delta_{R 2} R_{1},
\end{gather*}
$$

where $N_{i}(i=1,2,3,4,5)$ is the original inhomogeneous term for the monodisperse case (Kanagawa et al., 2010).

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