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(54) **METHOD FOR PREDICTING DROP SIZE DISTRIBUTION**

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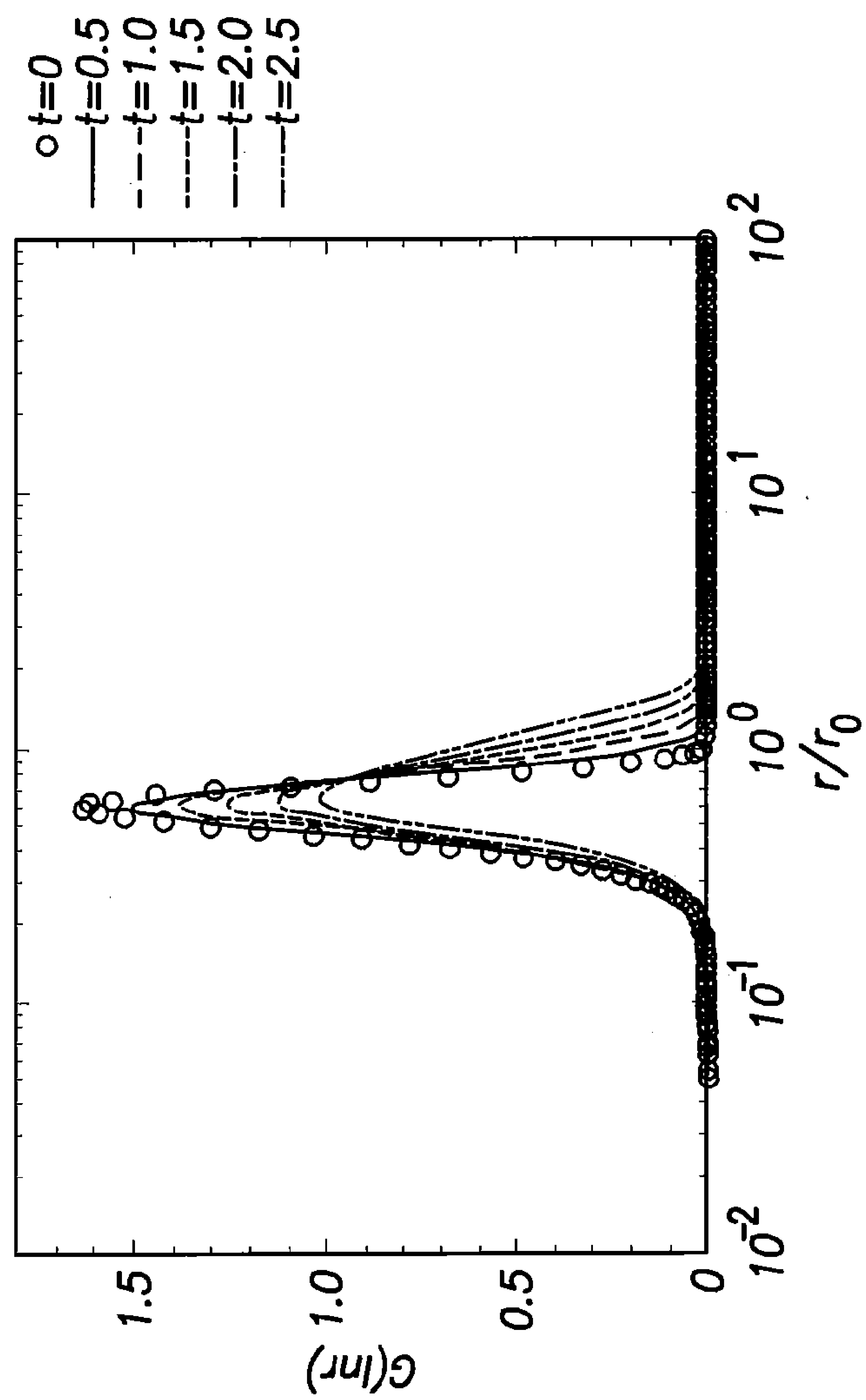
(57) **ABSTRACT**

The present invention relates to crude oil-water separation processes, specifically desalting in a petroleum refinery. More particularly, the present invention relates to a method and system for increase coalescence rates of water drops in a desalter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10^{-1}
10^{0}
10^{1}
10^{2}
r/r_0

FIG.2A

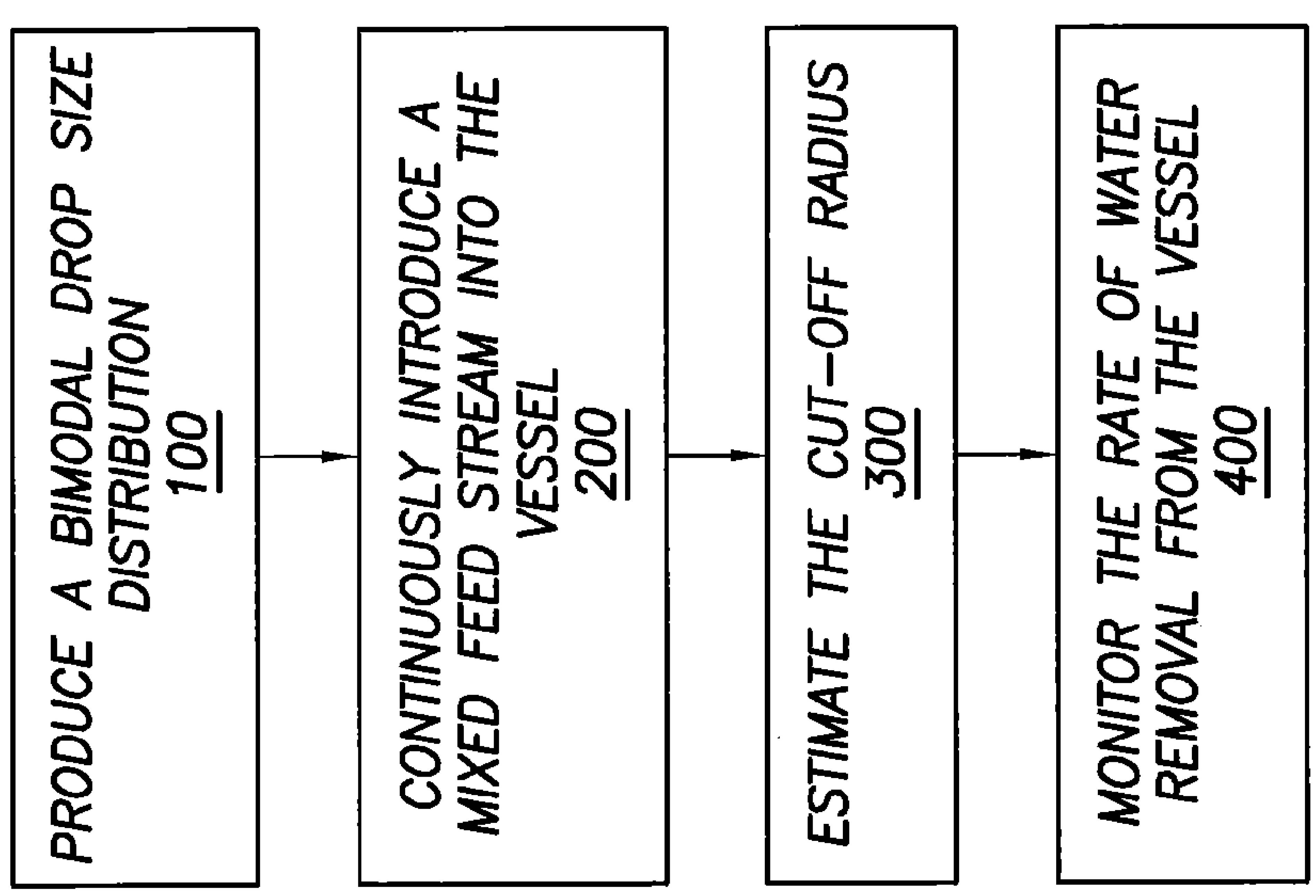
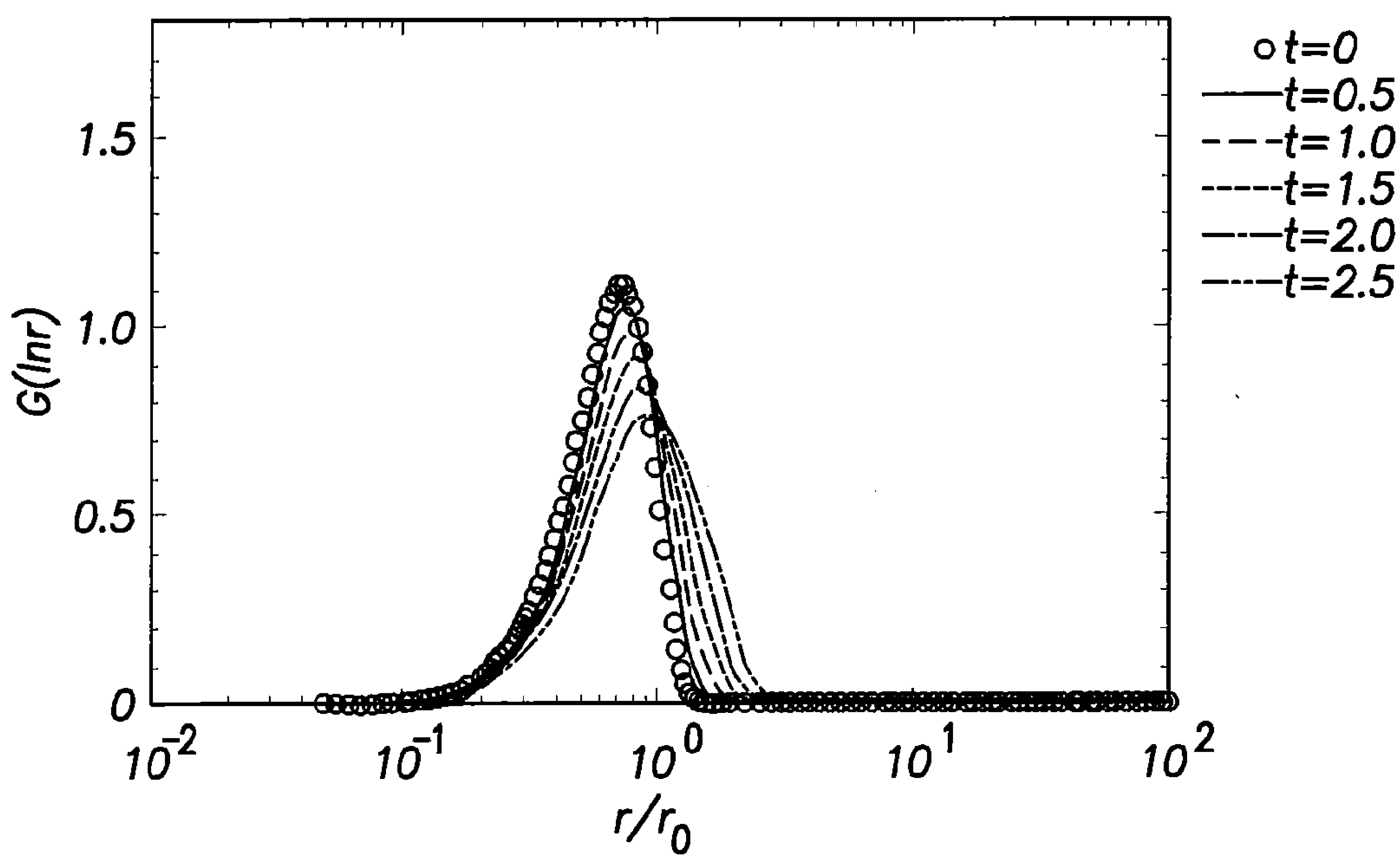

PRODUCE A BIMODAL DROP SIZE DISTRIBUTION
100
CONTINUOUSLY INTRODUCE A MIXED FEED STREAM INTO THE VESSEL
200
ESTIMATE THE CUT-OFF RADIUS
300
MONITOR THE RATE OF WATER REMOVAL FROM THE VESSEL
400

FIG.1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t=2.5
G(lnr)
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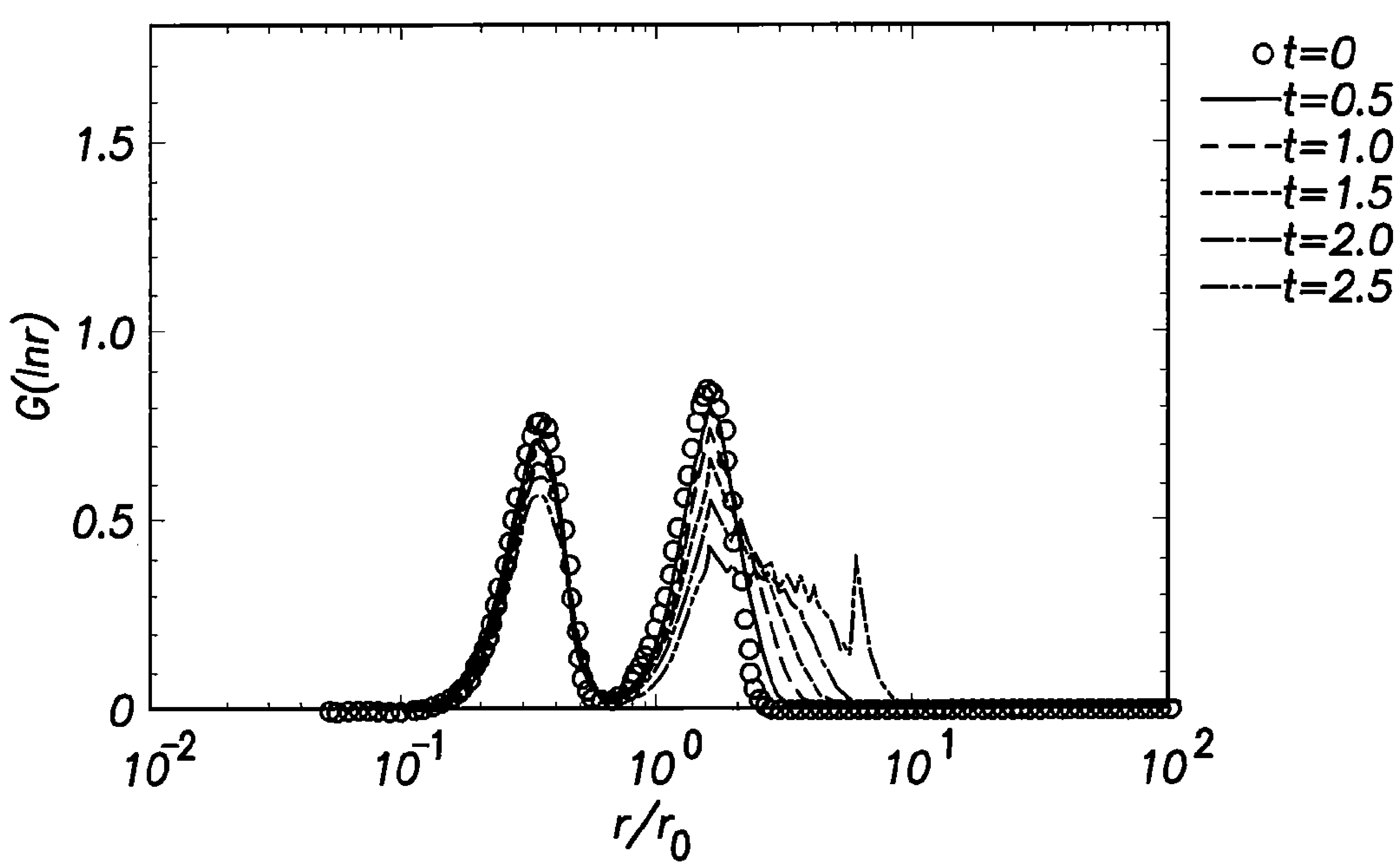
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}
r/r_0

FIG.2B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t=1.5
t=2.0
t=2.5
G(lnr)
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1.0
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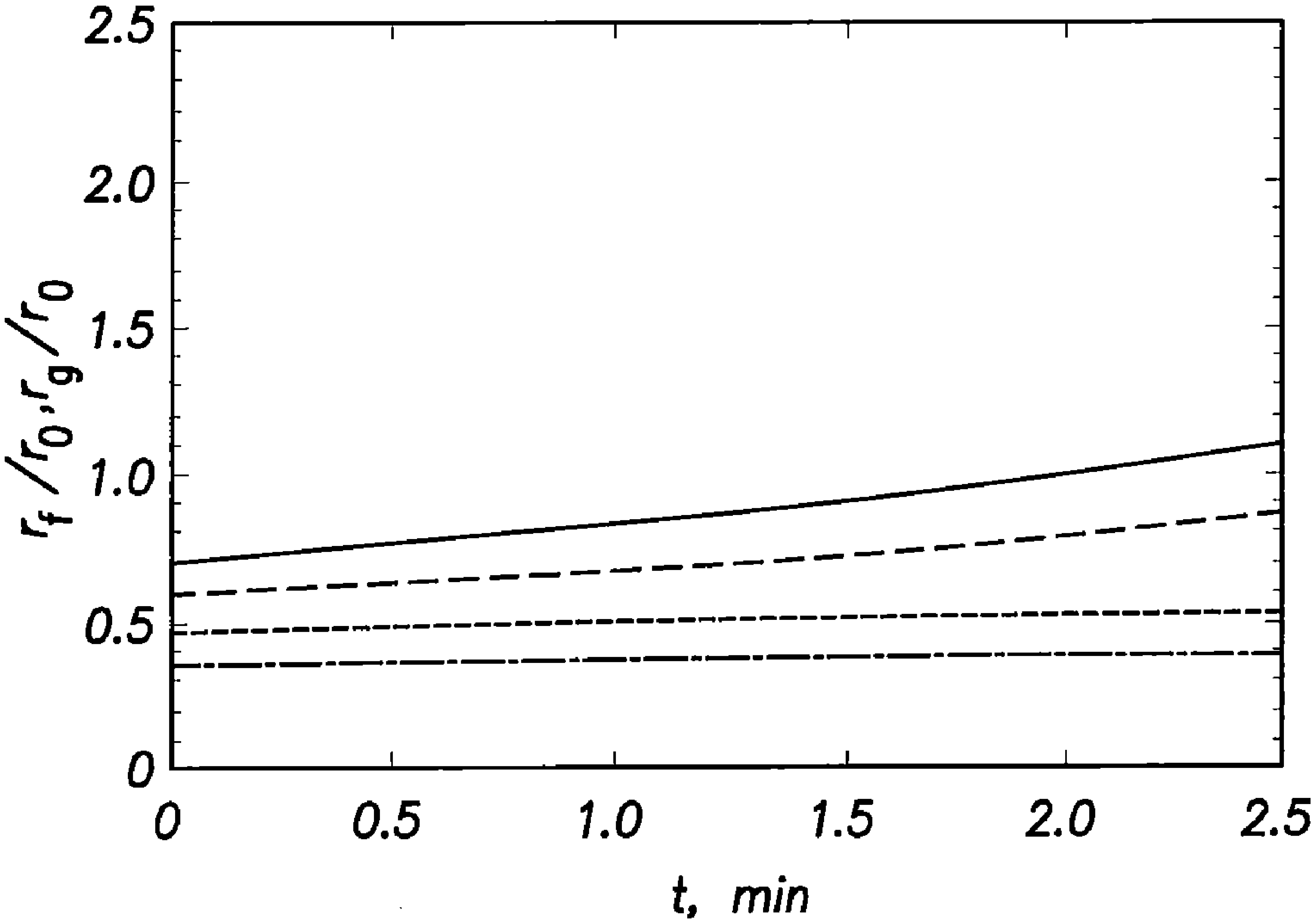
0
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}
r/r_0

FIG.2C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$r_f/r_0, r_g/r_0$
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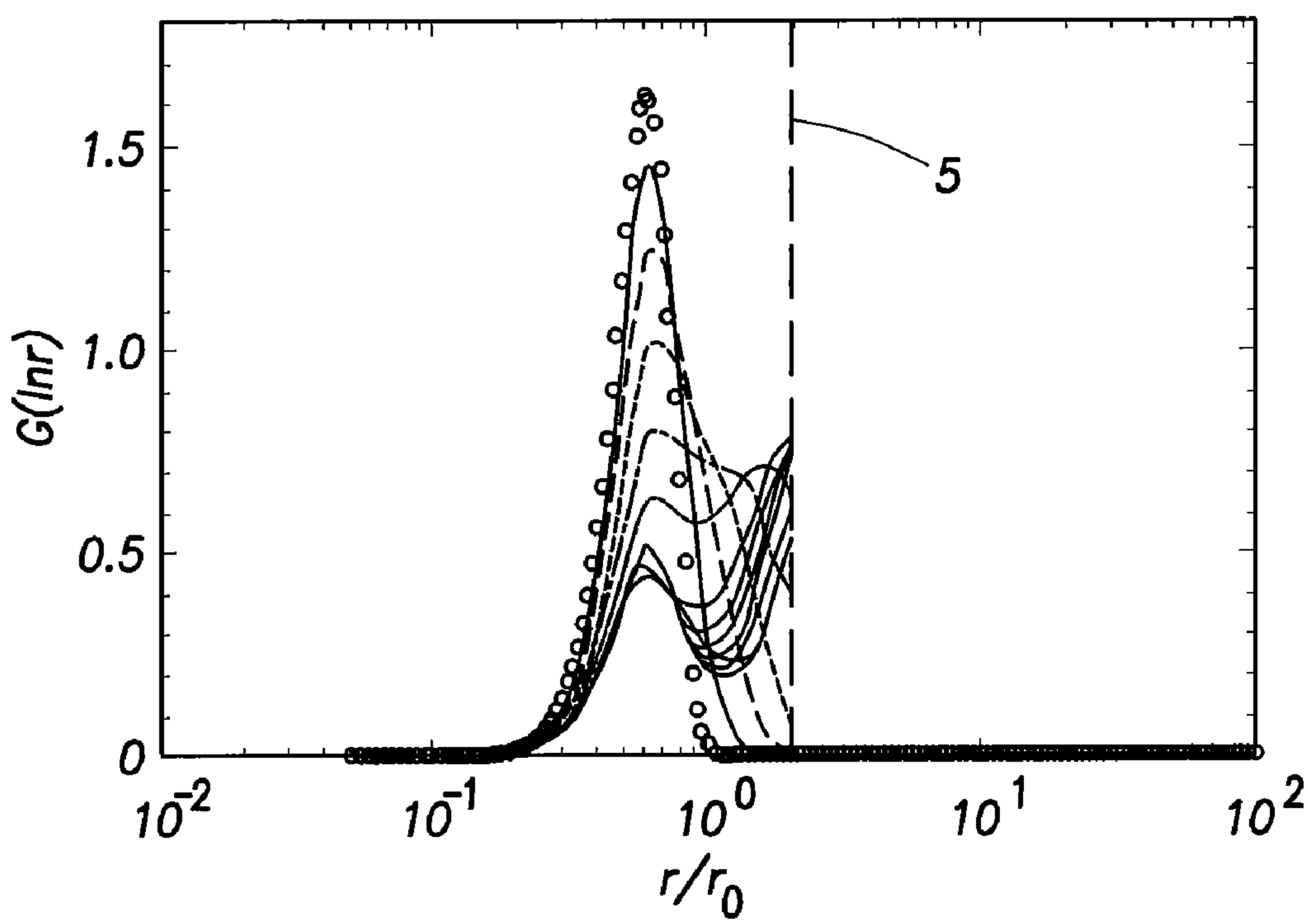
1.5
2.0
2.5
t, min

FIG.2D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0
G(lnr)
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10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}
r/r_0

FIG.3A

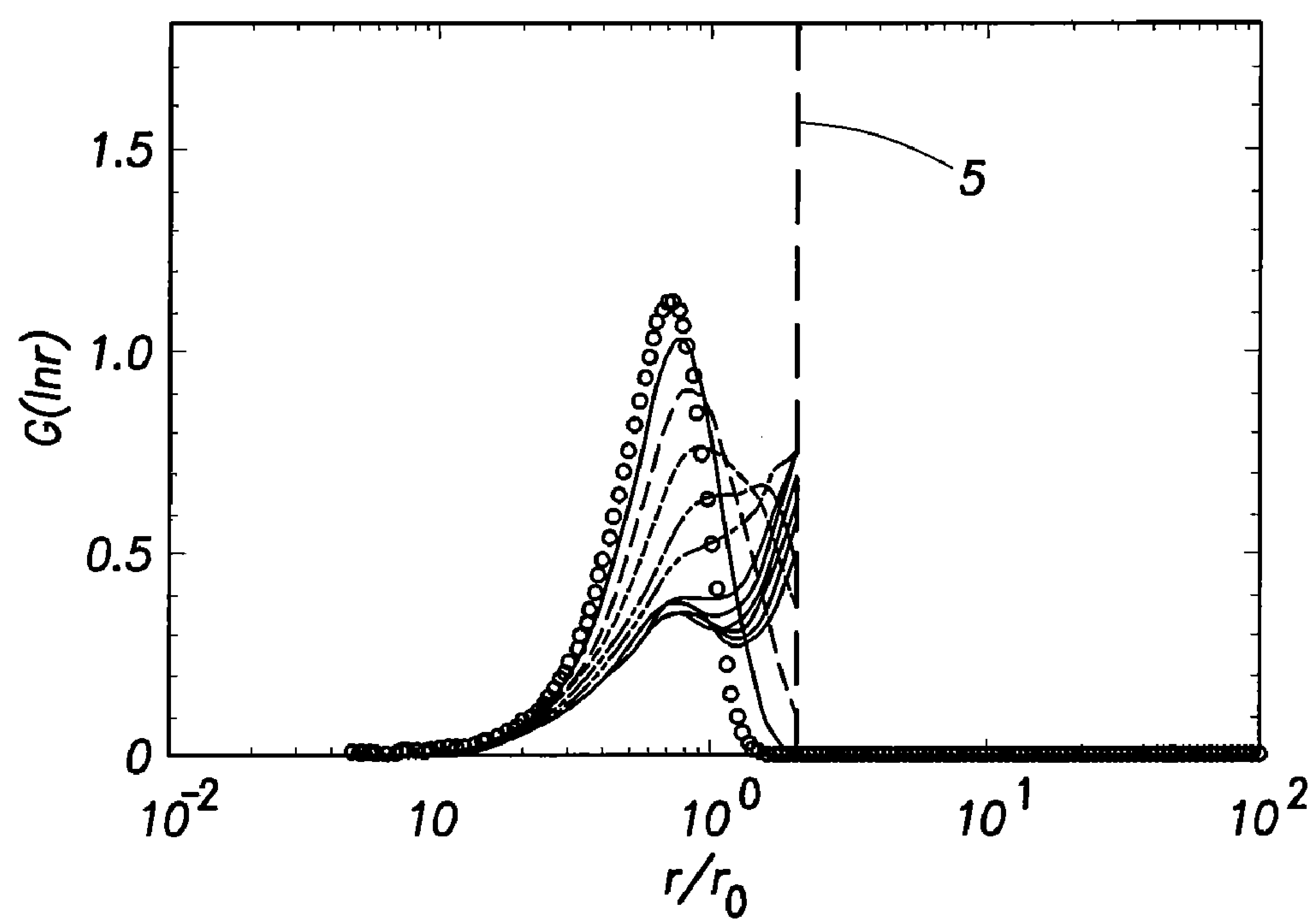

1.5
1.0
0.5
0
G(lnr)
5
10^{-2}
10
10^{0}
10^{1}
10^{2}
r/r_0

FIG.3B

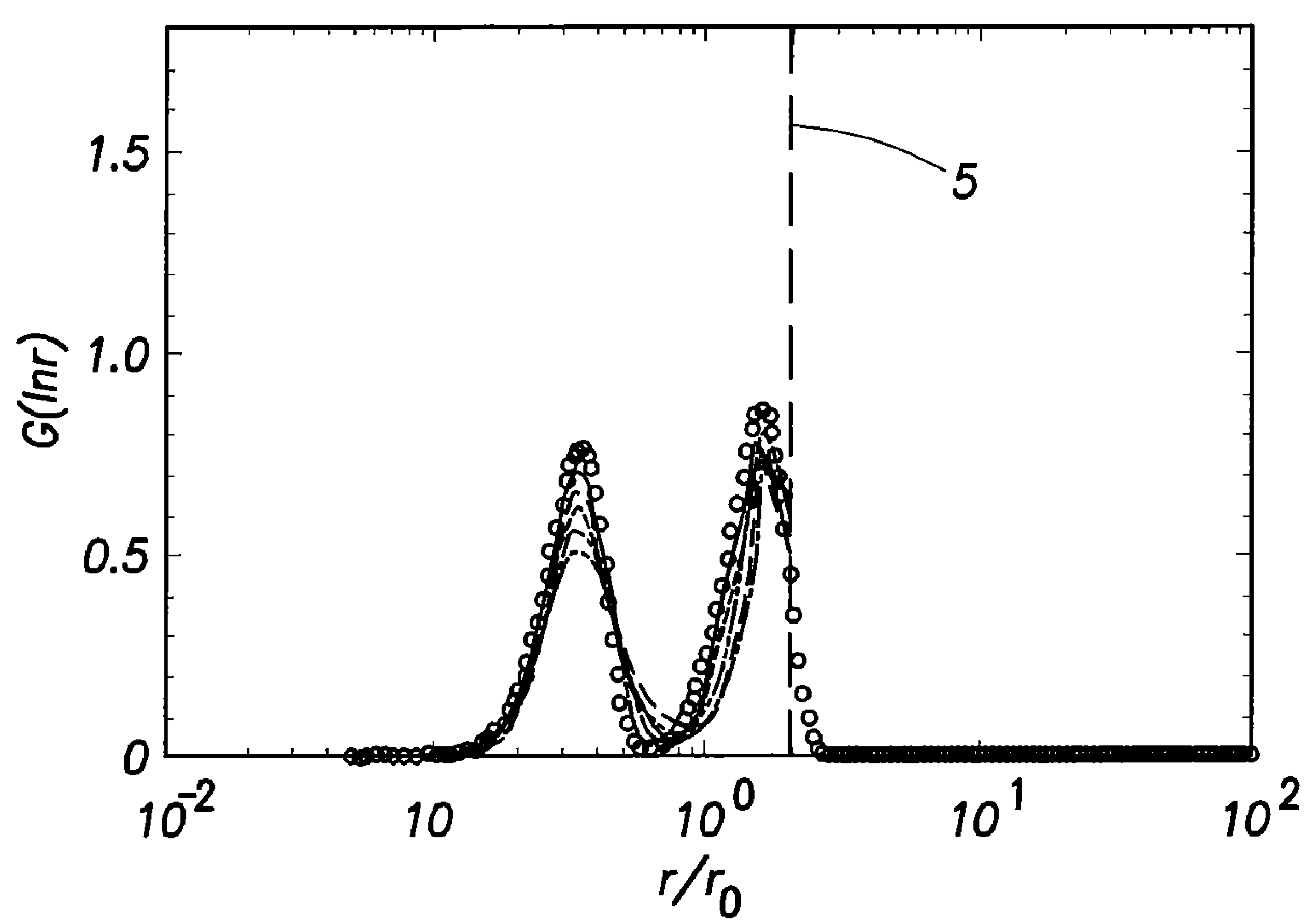
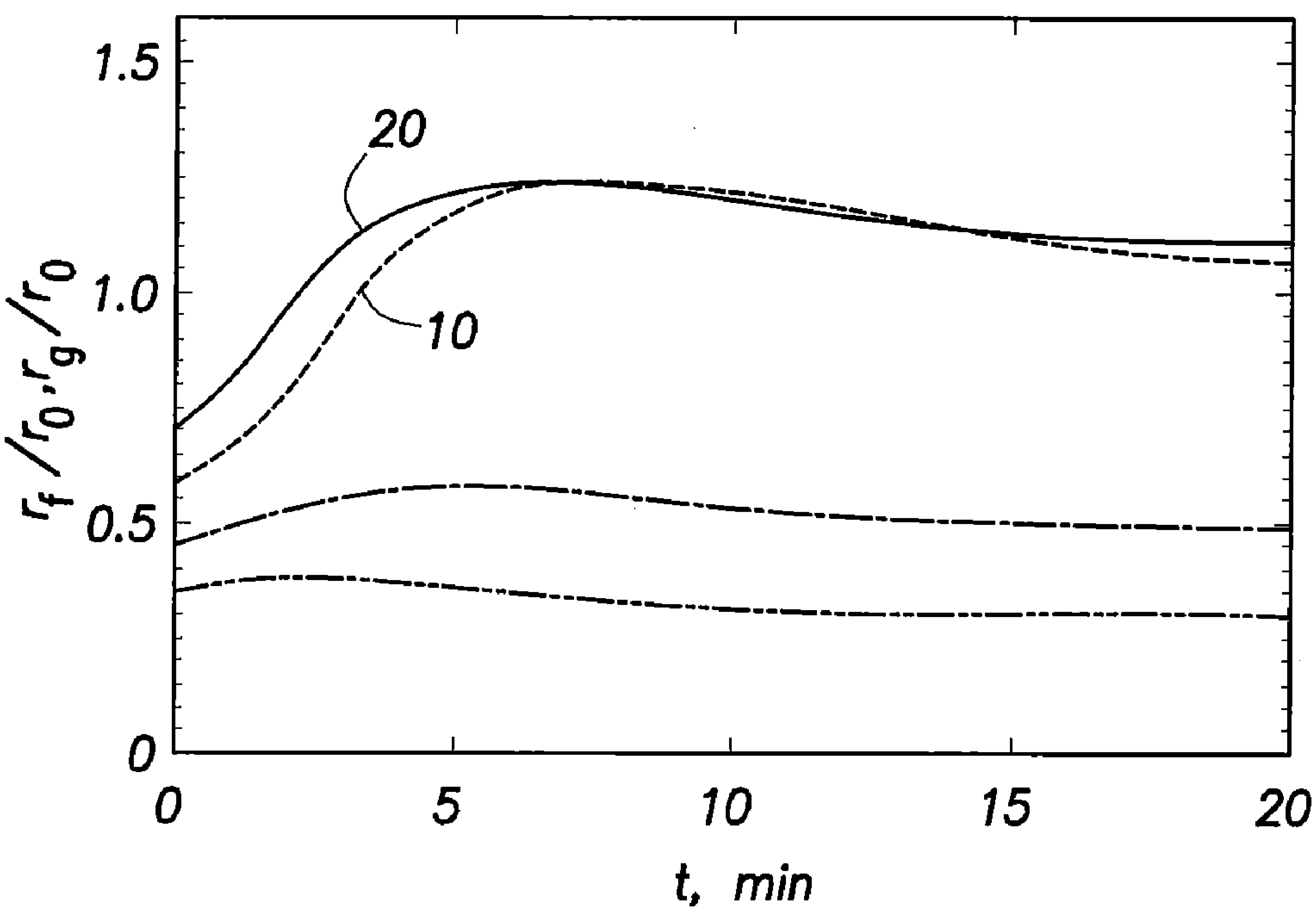

1.5
1.0
0.5
0
G(lnr)
5
10^{-2}
10
10^{0}
10^{1}
10^{2}
r/r_0

FIG.3C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1.0
0.5
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10
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5
10
15
20
t, min

FIG. 4A

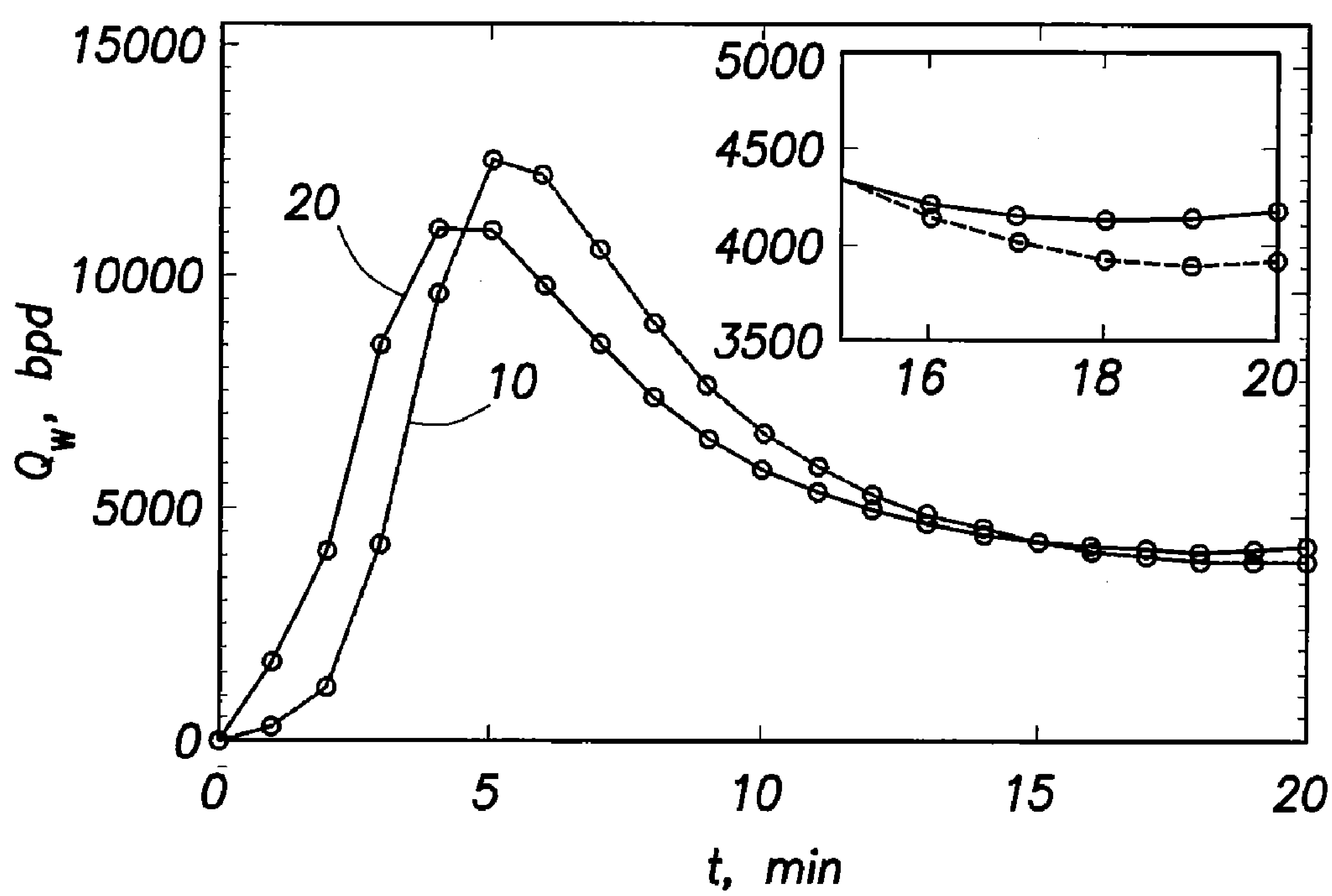
15000
10000
5000
0
Q_w, bpd
20
10
5000
4500
4000
3500
16
18
20
0
5
10
15
20
t, min

FIG. 4B

METHOD FOR PREDICTING DROP SIZE DISTRIBUTION

CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] This application claims priority benefit under 35 U.S.C. Section 119(e) to U.S. Provisional Patent Ser. No. 61/406,256 filed on Oct. 25, 2010 the entire disclosure of which is incorporated herein by reference.

FIELD OF THE INVENTION

[0002] The present invention relates to crude oil-water separation processes, specifically desalting in a petroleum refinery. More particularly, the present invention relates to a method and system for increase coalescence rates of water drops in a desalter.

BACKGROUND OF THE INVENTION

[0003] Desalting is the first process crude oil undergoes in a refinery. The primary purpose of desalting is to remove mineral salts present in crude oil, along with solids, metals, and water. Salts, mostly chlorides of sodium, potassium, and calcium naturally occur in soil and are associated with produced crude oil. Most of the salt is present as dissolved salt, in a small amount of water also associated with the crude oil. When this crude oil enters the refinery, it is necessary to remove these salts. Incomplete removal of salts can cause several problems, ranging from fouling and corrosion in heat exchangers and columns to catalyst poisoning.

[0004] Prior to entering the desalter vessel, crude oil is contacted with wash water by passing the two through a mixing valve. As a result, salt present in the crude oil is mixed with, and dissolved in the wash water. The mixing process also creates an emulsion of water drops in oil, which must be separated in order to remove the dissolved salt. This is accomplished in a desalter vessel, where the emulsion flows in at very slow velocities. An electric field in the desalter vessel promotes collision between drops, which leads to the formation of larger drops. When drops are sufficiently large, gravity forces the drops to settle to the bottom of the desalter vessel. Thus, the desalter vessel can be considered a gravity-based separation device, enhanced by the application of eclectic fields. In addition, chemicals may be added to promote drop coalescence.

[0005] With refineries increasingly processing more heavy crude oil, consistent desalting has become a challenge. The high density and viscosity of heavy crude oil, and its ability to form highly stable water-in-oil emulsions are mainly responsible for this inconsistent performance. Due to economic implications of poor desalting processes, it has become necessary to closely examine the fundamentals of the desalting process to investigate possible modifications. The fundamental process of water removal in a desalter vessel is the settling of water drops due to buoyant forces, which strongly depend on the size of the droplet.

[0006] Currently, optimization of desalter mixing is done primarily empirically, by tracing the performance of the desalter against mixing intensity usually controlled via the pressure drop across the mixing valve. Although there are some general principles on selecting the optimum pressure drop, there are no other controls used to alter mixing characteristics. This limits current desalting operation to the range of mixing produced by a single mixing valve.

[0007] Therefore, a need exists for a method for predicting the drop size of water in a desalter.

SUMMARY OF THE INVENTION

[0008] In an embodiment, a method of predicting drop size distribution including: (a) producing a bimodal drop size distribution, wherein the bimodal drop size distribution is produced by providing a constant fresh water stream and a constant crude oil stream through at least one mixing valve resulting in a mixed feed stream; (b) continuously introducing the mixed feed stream into a vessel, wherein the vessel includes an electric field; (c) estimating a cut-off radius, wherein any drops larger than the cut-off radius settle to the bottom of the vessel; and (d) monitoring the rate of water removal from the vessel.

[0009] In another embodiment, a method of predicting drop size distribution includes: (a) producing a bimodal drop size distribution, wherein the bimodal drop size distribution is produced by providing a constant fresh water stream and a constant crude oil stream through at least one mixing valve resulting in a mixed feed stream wherein the bimodal distribution is the linear combination of two unimodal distributions; (b) continuously introducing the mixed feed stream into a vessel, wherein the vessel includes an electric field, wherein the vessel is a desalter; (c) estimating a cut-off radius, wherein any drops larger than the cut-off radius settle to the bottom of the vessel; and (d) monitoring the rate of water removal from the vessel.

BRIEF DESCRIPTION OF THE DRAWINGS

[0010] The invention, together with further advantages thereof, may best be understood by reference to the following description taken in conjunction with the accompanying drawings in which:

[0011] FIG. **1** is a flow chart in accordance with an embodiment of the present invention.

[0012] FIGS. **2**(*a*)-(*d*) are plots from numerical simulations showing the evolution of drop size distribution in a desalter operating under batch conditions in accordance with the present invention.

[0013] FIGS. **3**(*a*)-(*c*) are plots from numerical simulations showing the evolution of drop size distribution in a desalter operating under continuous conditions in accordance with the present invention.

[0014] FIGS. **4**(*a*)-(*b*) are plots from numerical simulations showing rate of water drop growth and water removal in a desalter operating under continuous conditions in accordance with the present invention.

DETAILED DESCRIPTION OF THE INVENTION

[0015] Reference will now be made in detail to embodiments of the present invention, one or more examples of which are illustrated in the accompanying drawings. Each example is provided by way of explanation of the invention, not as a limitation of the invention. It will be apparent to those skilled in the art that various modifications and variations can be made without departing from the scope or spirit of the invention. For instance, features illustrated or described as part of one embodiment can be used on another embodiment to yield a still further embodiment. Thus, it is intended that the present invention cover such modifications and variations that come within the scope of the appended claims and their equivalents.

[0016] The primary driving force for drop collision is the relative settling velocity between drops of different sizes. The desalter is primarily a gravity-settling device, where separation occurs due to a density difference between the two fluids, water and oil. The density difference between a water drop and the surrounding oil results in a buoyant force, the magnitude of which depends on the drop size, following Stokes' law. For a rigid particle of radius r, the Stokes' settling equation reads,

$$v_{settle} = \frac{2r^2(\rho_w - \rho_o)g}{9\eta_o} \quad (1)$$

where g is the acceleration due to gravity; η_o is the dynamic oil viscosity; ρ_w is the density of water; and ρ_o is the density of oil. Eq. (1) provides that the magnitude of the settling velocity strongly depends on the size of the drop. Under the assumption of small water concentration, the average separation between water drops is too large for any drop-drop interaction. However, due to the difference in drop sizes, and hence settling velocities, the larger drops settle faster than the smaller drops, and collide with the smaller, slower moving drops as they settle.

[0017] The function of the electric field is to only enhance the probability of collision, after two drops are brought within a certain separation, by gravity. Thus, the driving force for collision (and hence drop coalescence) is the difference in the settling velocities. The electric field merely (albeit significantly) increases the probability of collision. In the extreme case of drops of equal size, there is no collision and hence no drop growth, even in the presence of an electrical field.

[0018] The drops size distribution (DSD) is quantified by a density function $f(x)$, defined such that $f(x)dx$ is the number of drops per unit volume in the size interval of (x, x+dx), where x is the mass of the drop, related to the radius by $x=4/3\pi r^3\rho_w$. It is preferred to work with mass (or equivalent volume) instead of drop radius, because in a collision process, the mass (and volume) of the resultant drop is simply the sum of the two colliding drops.

[0019] The evolution of the DSD can be written as[1],

$$\frac{\partial f(x)}{\partial t} = \int_0^{x/2} f(x_c)V(x_c \mid x')f(x')dx' - \int_0^{\infty} f(x)V(x \mid x')dx' \quad (2)$$

where $x_c=x-x'$. Eq. (2) can be read as follows: The rate of change of the number of drops of size x is equal to the rate of formation drops of size x minus the rate of consumption of drops of size x. The formation of drops of size x is by the coalescence of drops of size x' and x_c. The rate of consumption of drop x is by the coalescence of drops of size x with any other drop. The probability of collision is contained in the quantity, V(x|x'), termed the "collection kernel."[1] For discussion, it suffices to say that V(x|x') is a quantity which represents the probability of drops of size x>x' colliding with drops of size x'. The collection kernel is calculated using

$$V(x|x')=\pi(r+r')^2|v-v'|e(x,x') \quad (3)$$

where r and r' are the radii of drops of size x and x', v and v' are the settling velocities of drops x and x' under the influence of only gravity, and e(x, x') is a collision efficiency, which will be briefly described shortly. The velocity of a drop settling under gravity is given by

$$v = \frac{2(\hat{\eta}+1)r^2(\rho_w - \rho_o)g}{3(3\hat{\eta}+1)\eta_o} \quad (4)$$

where $\hat{\eta}=\eta_w/\eta_o$ is the viscosity ratio of water to oil. Eq. (4) is similar to the Stokes' settling velocity for a rigid particle (Eq. (1)). It is clearly seen that in the limited viscosity ratios, the additional factor is due to the fact that the fluid velocity inside the drop is non-zero, creating a "slip" at the oil/water interface. The physical implication of this is that drops will settle faster than a rigid particle of equal size and density.

[0020] The collision efficiency, e(x, x'), is a quantity which quantifies the ratio of the actual collision rate to the rate of two drops under gravity settling ignoring all drop-drop interactions, including hydrodynamic forces, forces due to external fields such as electric fields, and van der Waals forces. The quantity e can take values ranging from several orders below unity to several orders above unity. The range of values depends on the crude oil and water properties such as density and viscosity, magnitude of the electric field, material dielectric property (which dictate van der Waals forces), etc.

[0021] A log-normal distribution is chosen to represent shape of the feed DSD. As used herein, a "feed drop size distribution" refers to the water drop size distribution in the incoming crude oil to the desalter, i.e., the crude oil coming out of the mixing valve into the desalter. The log-normal distribution was chosen because it is representative of emulsion drops generated by a variety of mixing devices. However, a regular distribution can also be used. An actual DSD derived from experiments can also be incorporated into the model. As used herein, "actual drop size distribution" refers to the drop size distribution inside the desalter, which changes under the influence of the electric field. The general form of the drop size distribution can be written as

$$f(x) = \left(\frac{N^2}{L}\right)\left(\frac{(\nu+1)^2}{\Gamma(\nu+1)}\right)s^{\nu}\exp[-(\nu+1)s] \quad (5)$$

where N is the total drop number density (# of drops per unit volume); L is the total water content (mass per unit volume contained in drops);

$$x_m = L/N$$

is the mean mass of drops; and

$$s = x/x_m.$$

N and L can be related back to the drop size distribution function of Eq. (5) via

$$N=\int f(x)dx \quad L=\int xf(x)dx \quad (6)$$

[0022] The relative variance of the drop size distribution is given by,

$$\mathrm{var}(x)=1/(v+1).$$

The drop size distribution function, $f(x)$, is difficult to interpret since the number of drops does not remain constant. It is therefore useful to transform the function into a mass distribution function. The mass distribution function is represented in terms of drop radii as,

$$G(\ln r)=xf(\ln r)=(3L)\left(\frac{(v+1)^{v+1}}{\Gamma(v+1)}\right)s^{v+2}\exp[-(v+1)s] \tag{7}$$

G can be described in the same manner as f. G(ln r)d ln r is the mass of the drops in the size range (ln r, ln r+d ln r) per unit volume. The radius is represented as a logarithm to accommodate for the wide range of values, typically seen in emulsions.

[0023] FIG. **1** is a flow chart representing a particular embodiment of the present invention illustrated in FIG. **1**. In alternative implementations, the functions noted in the various blocks may occur out of the order depicted in FIG. **1**. For example, two blocks shown in succession in FIG. **1** may in fact be executed substantially concurrently, or the blocks may sometimes be executed in the reverse order depending upon the functionality involved.

[0024] In step **100**, a bimodal drop size distribution is produced. The bimodal drop size distribution is produced by providing a constant fresh water stream and a constant crude oil stream through at least one mixing valve resulting in a mixed feed stream.

[0025] In step **200**, the mixed feed stream is continuously introduced into a vessel. The vessel includes an electric field. In an embodiment, the vessel is a desalter.

[0026] In step **300**, the cut-off radius is estimated. The cut-off radius is estimated based on the space velocity of the feed in the vessel and the physical properties of the crude and water. By re-arranging Stokes' equation, the cut-off radius can be estimated as,

$$r_{cut}=\sqrt{\frac{9\eta_o v_{feed}}{2(\rho_w-\rho_o)g}} \tag{8}$$

[0027] The space velocity feed, v_{feed}, can be calculated based on the feed volumetric flow rate and the vessel geometry. Any drops which are larger than the cut-off radius are assumed to settle down to the water layer, and are removed as brine from the vessel. The rate of water removal is tracked as a function of time.

[0028] In step **400**, the rate of water removal from the vessel is monitored.

[0029] The following examples of certain embodiments of the invention are given. Each example is provided by way of explanation of the invention, one of many embodiments of the invention, and the following examples should not be read to limit, or define, the scope of the invention.

[0030] For each example, the following assumptions are prevalent: (1) the desalted volume is homogenous; (2) the electric field is constant between the plates; (3) there is a negligible change in drop size distribution outside the electric field; (4) drops remain sphere-like; (5) the concentration of water is small; (6) laminar flow conditions prevail in the desalter vessel; (7) drop breakup is neglected; and (8) crude oil composition chemistry and the effect of added chemical is not explicitly considered, however, it can be empirically incorporated.

[0031] The simulations were performed using three different feed drop size distributions. Two of the feed DSD's were unimodal distributions differing in their variance (distribution width). The third was a bimodal distribution represented by $G=cG_1+(1-c)G_2$, with unimodal distributions G_1 and G_2. The bimodal distribution is constructed of a linear combination of two unimodal distributions.

[0032] The following table summarizes the parameters selected for the feed distributions for this batch simulation.

TABLE 1

Parameter for feed drops size distributions used in simulations

	Type	x_1/x_0	$1/(1+v_1)$	x_2/x_0	$1/(1+v_2)$	c
I	Unimodal - Narrow	1/10	1	—	—	—
II	Unimodal - Wide	1/303	100	—	—	—
III	Bimodal	1/50	1	2	1	0.417

Example 1

Batch Simulation

[0033] The first example simulates batch conditions where a constant volume of water-in-oil emulsion is subject to an electric field. The following parameters were chosen:

Density of water, ρ_w=1.00 g/cc

Density of oil, ρ_o=0.90 g/cc~26° API

Viscosity of water=0.1 cP

Electric field intensity=10,000 V/in

Water content=5% by volume

[0034] FIGS. **2**(*a*)-(*c*) show the evolution of the drop size distribution with time for three different feed conditions. FIGS. **2**(*a*)-(*c*) show the mass distribution function, G(ln r), plotted as a function of the radius, normalized by an arbitrary radius of r_o=50 μm. The open circles correspond to the feed DSD, i.e., DSD at time t=0.

[0035] FIG. **2**(*c*) shows two distinct peaks corresponding to the bimodal distribution. The shifting of the drop size distribution function with time, to larger radii represents drop growth. FIG. **2**(*c*) shows that the peaks decrease in height as the distribution moves to larger sizes since the total mass of water, i.e., the area under the curve, remains constant.

[0036] FIG. **2**(*d*) shows the evolution of mean radius, r_f, and "predominant radius,"[2] r_g as a function of time. The dashed lines correspond to the mean radii and the solid lines correspond to the predominant radii. The mean and predominant radii are defined as the radii corresponding to mean mass, x_f, and the predominant mass, x_g, which are statistical moments of the distribution. They are calculated as

$$x_f=\langle x\rangle=\frac{\int xf(x)dx}{\int f(x)dx} \tag{9}$$

-continued

$$x_g = \frac{\langle x^2 \rangle}{\langle x \rangle} = \frac{\int x^2 f(x) dx}{\int x f(x) dx}$$

[0037] The mean radius is the total mass of water (which remains constant) divided by the number of drops. The number of drops reduces with time, due to drop coalescence. Therefore, the mean mass increases over time. More useful, however, is the predominant mass, which is the ratio of the second moment of mass to the total mass of water. The predominant mass can be interpreted as the "most likely" drop one might except to encounter in the total emulsion volume. The corresponding radii are calculated as

$$r_f = \left(\frac{3}{4\pi\rho_w}\right)^{\frac{1}{3}} x_f^{\frac{1}{3}} \quad (10)$$

$$r_g = \left(\frac{3}{4\pi\rho_w}\right)^{\frac{1}{3}} x_g^{\frac{1}{3}}$$

[0038] FIGS. **2**(*a*)-(*d*), reveals that the predominant size increases with time, signifying the shift in the distribution to larger drop sizes. That rate of increase, however, different for different feed DSD's, and is the highest for the feed with the bimodal distribution.

Example 2

Continuous Simulation

[0039] The second example provides a continuous process with constant feed and outlet stream. The following parameters were chosen:

Density of water, ρ_w=1.00 g/cc
Density of oil, ρ_o=0.90 g/cc~26° API
Viscosity of water=0.1 cP
Electric field intensity=10,000 V/in
Water content=5% by volume
Crude oil flow rate=100,000 bpd
Desalter vessel capacity=7900 ft^3

[0040] In these simulations, a constant feed and product crude oil stream are assumed. The amount of water removed is estimated based on the mass of water contained in drops which are larger enough to settle by gravity. A reasonable cut-off radius of r_{cut}=100 μm is assumed. FIG. **3** shows the cut-off radius line **5**.

[0041] FIG. **3** shows the evolution of the DSD as a function of time for three feed conditions. The symbols and lines correspond to the feed DSD and DSD plotted in intervals of 1 minute. The total simulation time was 20 minutes. FIG. **3** shows a sharp transition at

$$r / r_o = 2,$$

where the value of the mass distribution function drops to zero. This corresponds to the cut-off radius, r_{cut}=100 μm, shown by the black dashed line. Assuming any drops larger than the cut-off radius settles down to the water layer, there is a sharp drop in the mass distribution curve. Thus, the distribution curve provided in FIG. **3** only includes water drops which are still part of the emulsion and does not count drops which have settled into the water layer.

[0042] FIG. **4**(*a*) shows the evolution of mean (dashed lines) and predominant (solid lines) radii for different feed conditions. It can be seen that the representative radii remain unchanged at ling times, confirming that the DSD's indeed reach a steady-state.

[0043] FIG. **4**(*b*) shows the instantaneous rate of water removal (in bpd) plotted as a function of time for the three feed conditions. The rate of water removal is related to the DSD as follows: In the emulsion, there is a continuous rate at which smaller drops coalesce to form larger drops. Once a drop exceeds the cut-off size, it settles by gravity. Consider the two unimodal distributions, curves **10** and **20**. Initially the drops are all smaller than the cut-off radius, as shown by the open circles in FIG. **3** being to the left of the cut-off radius line **5**. Accordingly, the rate of water removal is zero. In time, drops grow, the DSD crosses the cut-off radius size, and water is removed at a certain rate. This rate continues to increase as there are more and more large drops being formed, and a maximum water removal rate is attained. However, this rate corresponds to a high concentration of large drops, all of which settle at approximately the same time. This results in a crease in the water removal rate. At some point, the rates of drop growth and drop settling reach a steady-state, and this corresponds to the steady-state value of water removal rate.

[0044] The inset in FIG. **4**(*b*) is a magnified section of the plot long times, to clearly show the steady-state value. It is seen that, between the unimodal distributions, the wide distribution has a larger water removal rate than the narrow distribution.

[0045] The bimodal distribution curve **30** shows a qualitative difference compared to the unimodal distributions. Initially, the bimodal distribution curve is such that it already contains a small number of drops which are larger than the cut-off radius, as seen in FIG. **3**, with part of the DSD to the right of the cut-off line. This results in a non-zero water removal rate at t=0.

[0046] In closing, it should be noted that the discussion of any reference is not an admission that it is prior art to the present invention, especially any reference that may have a publication date after the priority date of this application. At the same time, each and every claim below is hereby incorporated into this detailed description or specification as additional embodiments of the present invention.

[0047] Although the systems and processes described herein have been described in detail, it should be understood that various changes, substitutions, and alterations can be made without departing from the spirit and scope of the invention as defined by the following claims. Those skilled in the art may be able to study the preferred embodiments and identify other ways to practice the invention that are not exactly as described herein. It is the intent of the inventors that variations and equivalents of the invention are within the scope of the claims while the description, abstract and drawings are not to be used to limit the scope of the invention. The invention is specifically intended to be as broad as the claims below and their equivalents.

REFERENCES

[0048] All of the references cited herein are expressly incorporated by reference. The discussion of any reference is not an admission that it is prior art to the present invention, especially any reference that may have a publication data after the priority date of this application. Incorporated references are listed again here for convenience:

[0049] 1. Berry, E. X. and R. L. Reinhardt, *An Analysis of Cloud Drop Growth by Collection: Part I. Double Distributions*. Journal of the Atmospheric Sciences, 1974. 31(7): p. 1814-1824.

1. A method of predicting drop size distribution comprising:

a. producing a bimodal drop size distribution, wherein the bimodal drop size distribution is produced by providing a constant fresh water stream and a constant crude oil stream through at least one mixing valve resulting in a mixed feed stream;

b. continuously introducing the mixed feed stream into a vessel, wherein the vessel includes an electric field;

c. estimating a cut-off radius, wherein any drops larger than the cut-off radius settle to the bottom of the vessel; and

d. monitoring the rate of water removal from the vessel.

2. The method according to claim **1**, wherein the bimodal distribution is the linear combination of two unimodal distributions.

3. The method according to claim **1**, wherein the vessel is a desalter.

4. A method of predicting drop size distribution comprising:

a. producing a bimodal drop size distribution, wherein the bimodal drop size distribution is produced by providing a constant fresh water stream and a constant crude oil stream through at least one mixing valve resulting in a mixed feed stream wherein the bimodal distribution is the linear combination of two unimodal distributions;

b. continuously introducing the mixed feed stream into a vessel, wherein the vessel includes an electric field, wherein the vessel is a desalter;

c. estimating a cut-off radius, wherein any drops larger than the cut-off radius settle to the bottom of the vessel; and

d. monitoring the rate of water removal from the vessel.

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