



## Critical $\beta$ -values of all coil planet centrifuges

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### ABSTRACT

I-type, J-type and non-synchronous centrifuges are all coil planet centrifuges. Analysing the motion of I-type and J-type centrifuges has advanced the understanding of how to manufacture and use these centrifuges. This paper analyses the motion of non-synchronous centrifuges producing equations of motion that can be applied to all coil planet centrifuges. This has also produced simple equations to determine the critical  $\beta$ -values for any coil planet centrifuge. This paper also demonstrates that I-type centrifuges also have 2 critical  $\beta$ -values when it was thought that  $\beta$ -value did not influence the understanding of the processes within I-type centrifuges. For the I-type instrument both of these critical values are at bobbin radii approaching infinity. In practice this means all I-types function within one  $\beta$ -value range hence the unilateral distribution and type/effectiveness of the mixing is consistent. Finally the paper shows the influence that the tangential velocity has on the Archimedean screw effect and thus the unilateral distribution of the upper and lower phases in the columns of coil planet centrifuges. This explains why the maximum stationary phase retention in an I-type centrifuge is limited to 50%.

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### 1. Introduction

I-type, J-type and non-synchronous centrifuges are all coil planet centrifuges. The I-type and J-type centrifuges are classified as synchronous centrifuges because the bobbins/columns and rotor have the same rotational speed. The bobbin and rotor of an I-type instrument rotate in opposing directions, when viewed relative to the rotor, and thus have different rotational velocities. Where as the rotor and bobbins of a J-type rotate in the same direction and thus have the same rotational velocity.

Non-synchronous centrifuges are much more complex. The bobbin/column and rotor will rotate at different speeds and the directions of rotation may be the same or opposing one another. Thus it is important to derive general equations of motion and critical  $\beta$ -values for all non-synchronous centrifuges.

#### 1.1. Synchronous planetary centrifuges

##### 1.1.1. J-type and I-type centrifuges

For J-type centrifuges it is known that the unilateral (hydrodynamic) distribution [1,2] of the upper and lower phases within J-type centrifuges is greatly influenced by critical  $\beta$ -values. The critical  $\beta$ -values for a J-type instrument are 0.25 and 0.5 [2,3]. These critical values divide the column into three distinct ranges:

0–0.25, 0.25–0.5 and above 0.5. Within any of these ranges, both the unilateral distribution of a phase system and the type and effectiveness of the mixing and settling are consistent. However the unilateral distribution of the phase system and the effectiveness of mass transfer (mixing and settling) may be different in each range. Thus to understand the hydrodynamic processes inside a coil planet centrifuge the critical  $\beta$ -values must be known for that centrifuge.

The J-type centrifuge, with a column wound above a  $\beta$ -value of 0.5, is characterised by high stationary phase retention which can be greater than 95% and for this reason has become the most popular form of counter-current chromatography (CCC) device for separating and purifying small molecules. However, the wave mixing in such a J-type centrifuge [4] is not vigorous enough to separate biological samples using aqueous-polymer phase systems [5].

The I-type centrifuge is good at separating all types of molecules. Sutherland et al. showed that I-type centrifuges produce cascade mixing [4]. Cascade mixing is more vigorous than wave mixing creating a larger total surface area thus increasing mass transfer and chromatographic efficiency. Hence cascade mixing is particularly suitable for separating biological samples in aqueous-polymer phase systems [5]. However cascade mixing is compromised by the maximum stationary phase retention of 50% [6,7].

Both the I-type and J-type centrifuges allow simple paths for non-twisting flying leads due to the synchronous nature of the rotation described above. Thus the planetary drive ratio of a J-type centrifuge is 1, and is –1 for an I-type, planetary drive ratio is defined in Section 2.

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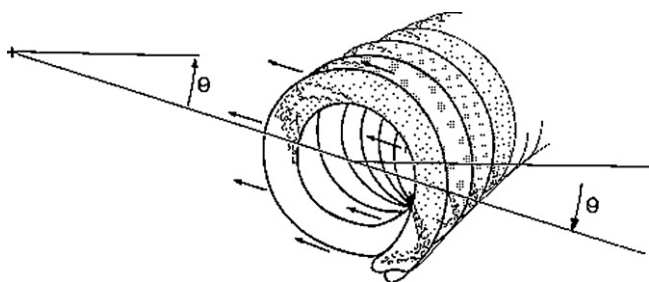


Fig. 1. Cascade mixing in an I-type centrifuge [4].

### 1.1.2. Cascade mixing and the I-type centrifuge

As Sutherland et al. [4] reported, cascade mixing occurs in an I-type centrifuge as shown in Fig. 1 regardless of  $\beta$ -value. For I-type motion the resultant acceleration vectors all have the same magnitude and direction as shown by the arrows in Fig. 1. However, these acceleration vectors do continuously change direction relative to the axis of the column every  $180^\circ$ . It is these continuous changes of the direction of the acceleration that cause cascade mixing.

### 1.1.3. Hydrodynamic processes in the J-type centrifuge

For a J-type centrifuge, it has been shown that hydrodynamic behaviour of the immiscible solvents is divided into three distinct regions by two critical  $\beta$ -values. These values are related to the motion of the bobbin relative to the rotor, i.e. the planetary drive ratio. For a J-type centrifuge these critical values are 0.25 and 0.5 [2,3]. These values divide the hydrodynamics of the J-type centrifuge into three regions with  $\beta$ -value ranges of 0–0.25, 0.25–0.5 and above 0.5 [2].

Between 0 and 0.25 the resultant acceleration vectors continuously change direction relative to the axis of the column for each  $180^\circ$  of rotation [3]. This is similar to the situation that creates cascade mixing in an I-type centrifuge. Thus in the J-type centrifuge, a form of cascade mixing occurs in this  $\beta$ -value range.

For a  $\beta$ -value of 0.25 the radial acceleration at the proximal key node is zero. For  $\beta$ -values above 0.25, the radial acceleration increases proportionally but always points in the same direction relative to the tubing. This allows the upper and lower phases of the solvent system to form distinct layers within the column [3]. Thus in a J-type centrifuge, cascade mixing does not occur above a  $\beta$ -value of 0.25. In fact wave mixing occurs as shown in Fig. 2 and described by both Sutherland et al. [4] and Conway [3]. Zones of mixing and settling travel along the column coincident with the low (proximal key node) and high (distal key node) accelerations caused by the column's epi-cyclic motion [4]. The mixing zones are coincident with the lower accelerations

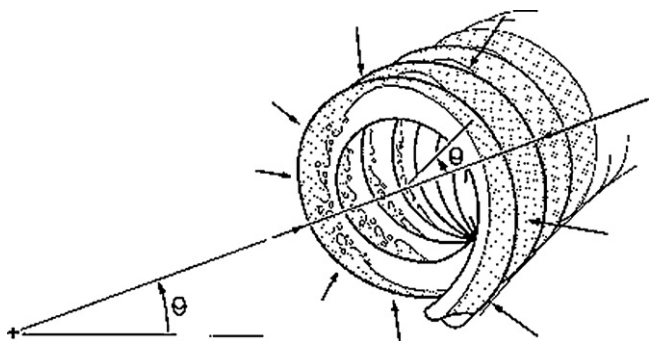


Fig. 2. Wave mixing in a J-type centrifuge [4].

at the proximal key node and take the form of waves [4]. The settling zones are coincident with the high accelerations at the distal key node and take the form of a smooth interfacial area [3].

For  $\beta$ -values between 0.25 and 0.5 the hydrodynamic (unilateral) distribution of solvent systems is not consistent. Polar phase systems distribute in the opposite manner to that of intermediate and non-polar phase systems [1,2]. The Archimedean screw effect states that the rotation of the coiled column transports (screws) the contents of the column towards the head end of a column [8]. This assumes that the transportation process is consistent throughout the column, i.e. the screwing effect always causes migration of the column contents towards one end of the column. It has been shown that below a  $\beta$ -value of 0.5, the direction of the tangential velocity changes and this is denoted in Fig. 3 by becoming negative each side of the proximal key node (angular position  $180^\circ$ ) as shown in Eq. (10) from [2]. This change in direction of the tangential velocity causes the Archimedean screw effect to reverse its direction as and when the tangential velocity does. This also means that the head and tail ends of the column switch ends whenever the tangential velocity changes its direction. Hence for  $\beta$ -values below 0.5 the Archimedean screw effect does not consistently push the contents of the column towards one end of the column. In fact the Archimedean screw effect alternates the end of the column to which it pushes the contents of the column. This partially explains why hydrophilic (polar) phase systems exhibit the opposite unilateral distribution (hydrodynamic behaviour) between  $\beta$ -values of 0.25 and 0.5 compared to intermediate and hydrophobic (non-polar) solvent systems [1,2].

At a  $\beta$ -value of 0.5 the radial and tangential velocities at the proximal key node are zero [2]. Thus this part of the bobbin/column is momentarily stationary (i.e. not moving) while the rest of the bobbin/column is still moving.

For  $\beta$ -values above 0.5 all solvent phase systems have the same hydrodynamic distribution with the lower (denser) phase migrating to the tail end of a column while the upper (less dense) phase moves to the head [1,2]. Above a  $\beta$ -value of 0.5 the tangential velocity does not switch direction, see Fig. 3, and hence the Archimedean screw effect is consistent and upper phase always migrates towards the head end of a column and vice versa. This explains the consistent hydrodynamic behaviour (unilateral distribution) of all phase systems when  $\beta$ -value is greater than 0.5.

## 1.2. Photographic evidence of wave mixing

At the 1984 Pittsburgh Conference Conway and Ito presented photographic studies, of the mixing that occurs in J-type centrifuges for  $\beta$ -values between 0.5 and 0.9 [1,9]. At the 1985 Pittsburgh Conference, Sutherland et al. extended the range of these photographic studies for  $\beta$ -values between 0.35 and 0.82 [7,10]. Unfortunately, neither of the Pittsburgh Conference presentations was published and the only images available in the public domain are those shown in page 191 of Ref. [3] which were for  $\beta$ -values greater than 0.5. Conway states, "macro-photographs show a wave motion at the interface of the mixing zone with little, if any, generation of droplets". It would have been interesting to see if droplets could be seen in the photographs presented by Sutherland for  $\beta$ -values between 0.35 and 0.5. However, evidence that there are differences in the effectiveness of the wave mixing above and below a  $\beta$ -value of 0.5 is presented below and shows that the wave mixing in the  $\beta$ -value range 0.25–0.5 is more effective than that above 0.5.

Now that scientific journals can print colour images it may be time to publish all the stroboscopic images presented by Sutherland and Conway. It may also be interesting to repeat these studies on a

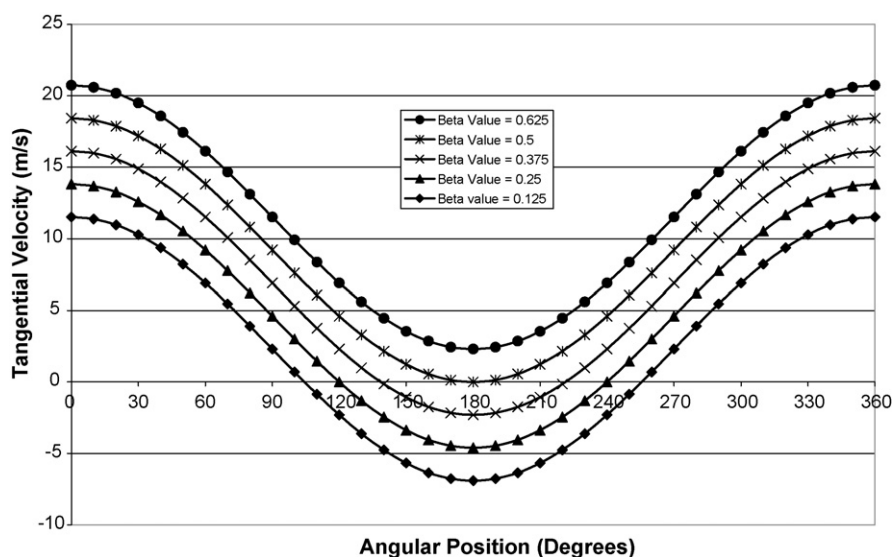


Fig. 3. Variation of the tangential velocity with  $\beta$ -value for a J-type centrifuge for a 110 mm rotor radius and a rotational velocity of 800 rpm.

suitable non-synchronous centrifuge knowing the critical  $\beta$ -values and observe the different types of mixing, wave and cascade, and to see if there is an observable difference in the mixing waves above and below the critical  $\beta$ -value  $\beta_1$ , see Section 2.

#### 1.2.1. Chromatographic efficiency in a J-type centrifuge

Table 1 from Sandlin and Ito [11] compares results for both “short” and “long” columns at  $\beta$ -values of 0.25 and 0.50. A “short” column was wound with a  $\beta$ -value of 0.25 and a “long” column was wound with a  $\beta$ -value range of 0.25–0.30. Also a “short” column was wound with a  $\beta$ -value of 0.50 and a “long” column was wound with a  $\beta$ -value range of 0.50–0.55.

Comparing the results for the “short” columns wound at 0.25 and 0.5 showed that the chromatographic efficiency of the 0.25  $\beta$ -value columns was the greater. These chromatographic efficiencies were measured in terms of both the number of theoretical plates and resolution.

In Table 1 of Ref. [11] the higher resolution of the 0.25  $\beta$ -value short column cannot be attributed to stationary phase retention. For reverse phase  $S_f = 84.1\%$  ( $R_S = 2.1$ ) for the 0.25  $\beta$ -value column and for the 0.5  $\beta$ -value column the  $S_f = 84.6\%$  ( $R_S = 0.98$ ).

In normal phase, even correcting<sup>1</sup> for the reduced stationary phase retention in the 0.5  $\beta$ -value column ( $S_f = 51.3\%$ ,  $R_S = 0.97$ ) does not account for the higher resolution of the 0.25  $\beta$ -value column ( $S_f = 77.5\%$ ,  $R_S = 2.1$ ).

Comparison of the chromatographic efficiencies of both the “long” columns shows that the 0.25  $\beta$ -value column also has the greater efficiency in terms of resolution and the number of theoretical plates, despite the 0.5  $\beta$ -value column having a higher stationary phase retention for each separation in both normal and reverse phase.

These higher chromatographic efficiencies must have been due to more effective mixing and hence higher mass transfer rates in the 0.25  $\beta$ -value columns. The mixing in the  $\beta$ -value range 0.25–0.5 must be more effective than above 0.5 and may be partially due to

the change in direction of the tangential velocity just either side of the proximal key node as discussed earlier. Stroboscopic photography could show evidence of such enhanced wave mixing within the  $\beta$ -value range 0.25–0.5.

#### 1.3. Kinematic motion and critical $\beta$ -values

The kinematic analysis of the J-type centrifuge motion led to the concept of critical  $\beta$ -values. Originally it was assumed that the I-type centrifuge had no critical  $\beta$ -values. However, the kinematic analysis of non-synchronous centrifuges shows that the I-type also has 2 critical  $\beta$ -values. Both of these values tend towards infinity (see Section 2). This explains why the I-type centrifuges’ motion and mixing process are not influenced by  $\beta$ -value.

#### 1.4. Non-synchronous centrifuges

Both I-type and J-type type centrifuges produce useful separations. However, the fixed planetary drive ratios of these centrifuges may not be optimal for a particular class/family of molecules to be separated or solvent system to be retained within the column. Thus there is a need to adjust this drive ratio to suit particular families of similar molecules and solvent systems normally used to separate them. However, as explained above,  $\beta$ -value plays an important role in determining stationary phase retention and chromatographic efficiency [1,2,11,12] in a J-type centrifuge. The critical  $\beta$ -values must also be taken into account when investigating the hydrodynamic behaviour of non-synchronous centrifuges. The problem with critical  $\beta$ -values is that they change depending upon the planetary drive ratio, i.e. the relative rotational velocities of the rotor and bobbins. The following section provides two equations to calculate both critical  $\beta$ -values based upon the relative rotational velocities of the rotor and bobbins.

## 2. Theory

In this section, the kinematics of a point  $P$  on the inside wall of a column that is undergoing non-synchronous motion are stud-

<sup>1</sup> The correction assumes that the peak widths remain constant for both the short columns but the difference between peak retention times is proportional to the stationary phase retention ( $S_f$ ).

ied. The column contains the mobile and stationary phases that as fluids are able to move relative to the point  $P$ . Thus the motion of the point  $P$  although it greatly influences the motion of the mobile and stationary phases it is not the actual motion of these fluids. However the kinematics of the point  $P$  is a close approximation to the motion of these fluids as demonstrated by previous kinematic analysis of the movement of I-type and J-type centrifuges [2–4,7] and can provide useful insights to the construction and use of coil planet centrifuges.

### 2.1. Planetary drive ratio

The planetary drive ratio ( $P_{Dr}$ ) is the rotational velocity of the column/bobbin relative to the rotor ( $u$ ) compared to the rotational velocity of the rotor ( $\omega$ ).

$$P_{Dr} = \frac{u}{\omega}$$

### 2.2. Derivation of the kinematic equations of motion for non-synchronous planetary centrifuges

From Fig. 4, resolve for the displacement of point  $P$  in the  $x$ -direction taking the centre of rotation as the origin. Then differentiate to obtain formulas for velocity and acceleration in the  $x$ -direction remembering that  $\theta = \omega t$  and  $\lambda = ut$ .

Displacement  $x$ -direction:

$$\begin{aligned} x &= R \cos(\theta) + r \cos(\lambda + \theta) \\ x &= R \cos(\omega t) + r \cos((u + \omega)t) \end{aligned} \quad (1)$$

Velocity  $x$ -direction:

$$V_x = \frac{dx}{dt} = -R\omega \sin(\omega t) - r(u + \omega) \sin((u + \omega)t) \quad (2)$$

Acceleration  $x$ -direction:

$$A_x = \frac{d^2x}{dt^2} = -R\omega^2 \cos(\omega t) - r(u + \omega)^2 \cos((u + \omega)t) \quad (3)$$

From Fig. 4, resolve for the displacement of point  $P$  in the  $y$ -direction taking the centre of rotation as the origin. Then differentiate to obtain formulas for velocity and acceleration in the  $y$ -direction.

Displacement  $y$ -direction:

$$\begin{aligned} y &= R \sin(\theta) + r \sin(\lambda + \theta) \\ y &= R \sin(\omega t) + r \sin((u + \omega)t) \end{aligned} \quad (4)$$

Velocity  $y$ -direction:

$$V_y = \frac{dy}{dt} = R\omega \cos(\omega t) + r(u + \omega) \cos((u + \omega)t) \quad (5)$$

Acceleration  $y$ -direction:

$$A_y = \frac{d^2y}{dt^2} = -R\omega^2 \sin(\omega t) - r(u + \omega)^2 \sin((u + \omega)t) \quad (6)$$

These equations represent the kinematic motion of point  $P$  in space relative to the origin, which is the main rotary axis of a non-synchronous planetary centrifuge. The negative signs in front of the above equations mean that the directions of the vector quantities are in the opposite direction to that shown in Fig. 4.

### 2.3. Radial and tangential velocities

From Fig. 5 resolving in the radial and tangential directions gives:

$$\begin{aligned} \text{Radial Velocity} &= V_x \cos(\lambda + \theta) + V_y \sin(\lambda + \theta) \\ \text{Radial Velocity } (V_R) &= V_x \cos((u + \omega)t) + V_y \sin((u + \omega)t) \end{aligned} \quad (7)$$

$$\begin{aligned} \text{Tangential Velocity} &= -V_x \sin(\lambda + \theta) + V_y \cos(\lambda + \theta) \\ \text{Tangential Velocity } (V_T) &= -V_x \sin((u + \omega)t) + V_y \cos((u + \omega)t) \end{aligned} \quad (8)$$

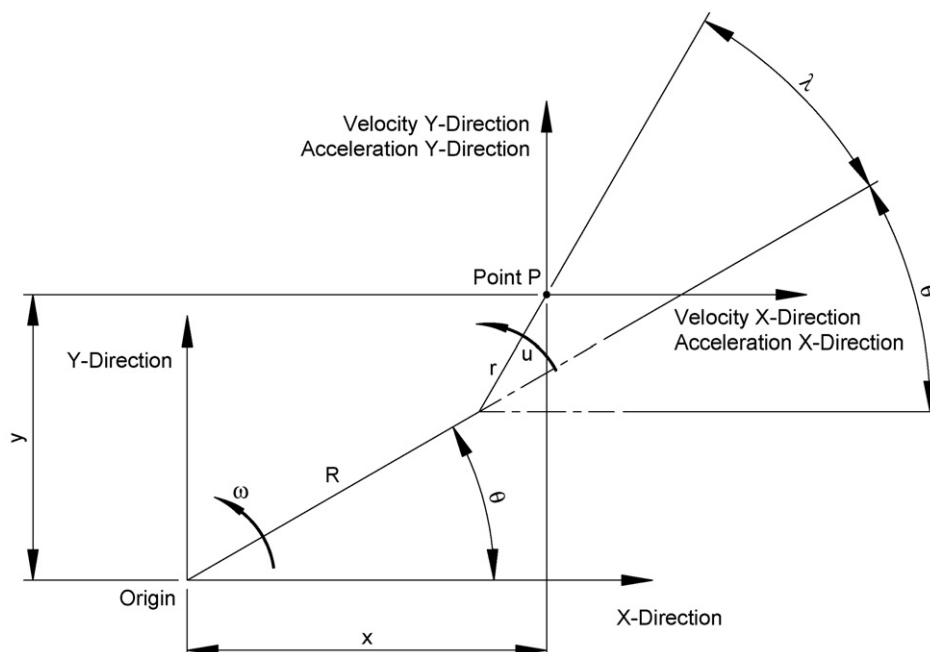


Fig. 4. The free-body diagram for a non-synchronous planetary centrifuge.

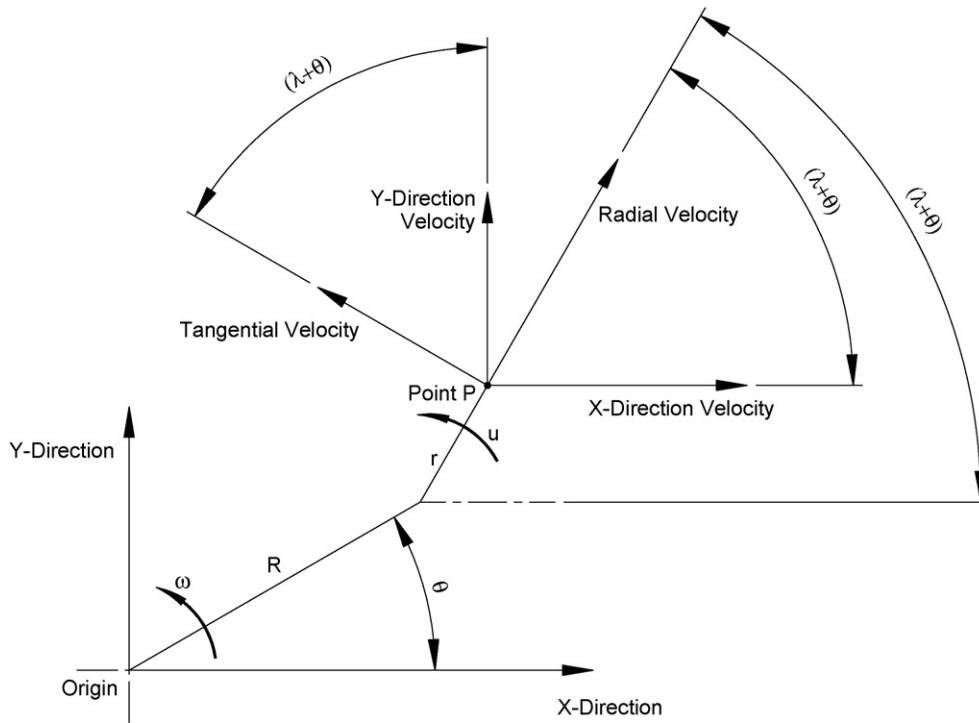


Fig. 5. The free-body diagram of a non-synchronous planetary centrifuge with the radial and tangential velocity vectors added.

2.3.1. Radial velocity

Substituting in Eq. (7) for  $V_x$  from Eq. (2) and for  $V_y$  from Eq. (5) gives:

$$V_R = -(R\omega \sin(\omega t) + r(u + \omega) \sin((u + \omega)t)) \cos((u + \omega)t) + (R\omega \cos(\omega t) + r(u + \omega) \cos((u + \omega)t)) \sin((u + \omega)t)$$

$$V_R = -R\omega \sin(\omega t) \cos((u + \omega)t) - r(u + \omega) \sin((u + \omega)t) \cos((u + \omega)t) + R\omega \cos(\omega t) \sin((u + \omega)t) + r(u + \omega) \cos((u + \omega)t) \sin((u + \omega)t)$$

Which reduces to:

$$V_T = (R\omega \sin(\omega t) + r(u + \omega) \sin((u + \omega)t)) \sin((u + \omega)t) + (R\omega \cos(\omega t) + r(u + \omega) \cos((u + \omega)t)) \cos((u + \omega)t)$$

$$V_T = R\omega [\sin(\omega t) \sin((u + \omega)t) + \cos(\omega t) \cos((u + \omega)t)] + r(u + \omega) [\sin^2((u + \omega)t) + \cos^2((u + \omega)t)]$$

$$V_R = R\omega [\cos(\omega t) \sin((u + \omega)t) - \sin(\omega t) \cos((u + \omega)t)]$$

Using  $\sin(A - B) = \sin(A) \cos(B) - \cos(A) \sin(B)$  where  $A = (u + \omega)t$  and  $B = \omega t$

Gives :  $V_R = R\omega \sin(ut)$  (9)

Since there is a no negative sign at the front of Eq. (9) the direction of  $V_R$  is the same as shown in Fig. 5. Eq. (9) also shows that the radial velocity will be zero whenever  $\sin(ut) = 0$  which is when  $ut = 0^\circ$  and  $180^\circ$ , i.e. the proximal and distal key nodes. This equation also shows that the  $V_R$  does not vary with  $\beta$ -value.

For a J-type centrifuge where  $u = \omega$ , Eq. (9) becomes:

J-type  $V_R = R\omega \sin(\omega t)$

Which agrees with the findings of Wood [2].

For an I-type centrifuge, the rotational velocity ( $u$ ) of the bobbin has the same magnitude as the rotational velocity ( $\omega$ ) of the rotor but is in the opposite direction, hence in Eq. (9)  $u$  is replaced by  $-\omega$  so Eq. (9) becomes:

I-type  $V_R = R\omega \sin(-\omega t)$

Since  $-\sin(\theta) = \sin(-\theta)$

I-type  $V_R = -R\omega \sin(\omega t)$  (10)

2.3.2. Tangential velocity

Substituting in Eq. (8) for  $V_x$  from Eq. (2) and for  $V_y$  from Eq. (5) gives:

But  $\sin^2((u + \omega)t) + \cos^2((u + \omega)t) = 1$ .  
Therefore  $V_T = R\omega [\sin(\omega t) \sin((u + \omega)t) + \cos(\omega t) \cos((u + \omega)t)] + r(u + \omega)$ .

Using  $\cos(A - B) = \sin(A) \sin(B) + \cos(A) \cos(B)$  where  $A = (u + \omega)t$  and  $B = \omega t$  gives:

$V_T = R\omega \cos(ut) + r(u + \omega)$  (11)

But  $\beta = r/R$  hence  $r = R\beta$ .

Therefore  $V_T = R\omega \cos(ut) + R\beta(u + \omega)$

$V_T = R[\omega \cos(ut) + \beta(u + \omega)]$  (12)

For a J-type centrifuge where  $u = \omega$ , Eq. (12) becomes:

J-type  $V_T = R\omega [\cos(\omega t) + 2\beta]$  (13)

Which agrees with the findings of Wood [2].

For an I-type centrifuge where  $u = -\omega$ , Eq. (12) becomes:

I-type  $V_T = R\omega \cos(-\omega t)$

Since  $\cos(\theta) = \cos(-\theta)$ :

I-type  $V_T = R\omega \cos(\omega t)$  (14)

2.3.3. First critical  $\beta$ -value

Eq. (12) shows that the tangential velocity is sinusoidal but with an offset that depends upon the variables  $\omega$ ,  $\beta$ -value and  $u$ . A critical  $\beta$ -value will occur when  $V_T = 0$ , let this be known as the first

critical  $\beta$ -value ( $\beta_1$ ). This can only occur when  $\cos(ut) = -1$  cancelling the offset created by the variables  $\omega$ ,  $\beta$ -value and  $u$ . So Eq. (12) becomes:

$$0 = R[\beta_1(u + \omega) - \omega]$$

For the right hand side of the above equation to =0, either  $R$  or the contents of the brackets must =0. However the rotor radius  $R \neq 0$ :

$$\text{Therefore } 0 = \beta_1(u + \omega) - \omega$$

$$\omega = \beta_1(u + \omega)$$

$$\beta_1 = \frac{\omega}{u + \omega} \tag{15}$$

Eq. (15) represents  $\beta_1$  in terms of the angular velocities of the rotor and the bobbin. Eq. (15) can also be applied to a J-type centrifuge where  $u = \omega$ . Thus  $\beta_1$  for a J-type centrifuge =0.5, which agrees with the findings of Wood [2].

Also for an I-type centrifuge where the bobbin has the same rotational speed but in the opposite direction to the rotor,  $u = -\omega$ . Substituting in Eq. (15) for  $u$  shows that the first critical  $\beta$ -value for an I-type centrifuge tends towards infinity.

Dividing the numerator and denominator of Eq. (15) by  $\omega$  gives:

$$\beta_1 = \frac{1}{(u/\omega) + 1}$$

Since  $P_{Dr} = u/\omega$  then:

$$\beta_1 = \frac{1}{P_{Dr} + 1} \tag{16}$$

2.4. Radial and tangential accelerations

From Fig. 6 resolving in the radial and tangential directions.

$$\text{Radial Acceleration } (A_R) = A_X \cos(\lambda + \theta) + A_Y \sin(\lambda + \theta) \tag{17}$$

$$A_R = A_X \cos((u + \omega)t) + A_Y \sin((u + \omega)t)$$

$$\text{Tangential Acceleration } (A_T) = -A_X \sin(\lambda + \theta) + A_Y \cos(\lambda + \theta) \tag{18}$$

$$A_T = -A_X \sin((u + \omega)t) + A_Y \cos((u + \omega)t)$$

2.4.1. Radial acceleration

Substituting in Eq. (17) for  $A_X$  from Eq. (3) and for  $A_Y$  from Eq. (6) gives:

$$A_R = -(R\omega^2 \cos(\omega t) + r(u + \omega)^2 \cos((u + \omega)t)) \cos((u + \omega)t) + (-R\omega^2 \sin(\omega t) - r(u + \omega)^2 \sin((u + \omega)t)) \sin((u + \omega)t)$$

$$A_R = -R\omega^2 \cos(\omega t) \cos((u + \omega)t) - r(u + \omega)^2 \cos^2((u + \omega)t) - R\omega^2 \sin(\omega t) \sin((u + \omega)t) - r(u + \omega)^2 \sin^2((u + \omega)t)$$

But  $\cos^2((u + \omega)t) + \sin^2((u + \omega)t) = 1$

$$\text{Therefore } A_R = -R\omega^2 \cos(\omega t) \cos((u + \omega)t) - R\omega^2 \sin(\omega t) \sin((u + \omega)t) - r(u + \omega)^2$$

Using  $\cos(A - B) = \cos(A)\cos(B) + \sin(A)\sin(B)$  where  $A = (u + \omega)t$  and  $B = \omega t$ .

$$\text{Therefore } \cos(ut) = \cos((u + \omega)t) \cos(\omega t) + \sin((u + \omega)t) \sin(\omega t)$$

$$\text{Therefore } A_R = -(R\omega^2 \cos(ut) + r(u + \omega)^2) \tag{19}$$

Since there is a negative sign, the direction of the  $A_R$  is inwards due to the vector being in the opposite direction of that in Fig. 6.

Since  $\beta = r/R \Rightarrow r = R\beta$ .

$$\text{Therefore } A_R = -(R\omega^2 \cos(ut) + R\beta(u + \omega)^2) \tag{20}$$

$$A_R = -R(\omega^2 \cos(ut) + \beta(u + \omega)^2)$$

For a J-type centrifuge where  $u = \omega$ , Eq. (18) becomes:

$$\text{J-type } A_R = -R\omega^2(\cos(\omega t) + 4\beta) \tag{21}$$

Which agrees with the findings of Wood [13].

For an I-type centrifuge the rotational velocity ( $u$ ) of the bobbin has the same magnitude as the rotational velocity ( $\omega$ ) of the rotor but is in the opposite direction, hence in Eq. (20),  $u$  is replaced by  $-\omega$  and Eq. (19) becomes:

$$\text{I-Type } A_R = -R\omega^2 \cos(-\omega t)$$

Since  $\cos(\theta) = \cos(-\theta)$ :

$$\text{I-Type } A_R = -R\omega^2 \cos(\omega t) \tag{22}$$

Which agrees with the findings of Wood [13].

2.4.2. Second critical  $\beta$ -value

Eq. (20) shows that the radial acceleration is sinusoidal but with an offset that depends upon the variables  $\omega$ ,  $\beta$ -value and  $u$ . A critical  $\beta$ -value will occur when  $A_R = 0$ , let this be known as the second critical  $\beta$ -value ( $\beta_2$ ). This can only occur when  $\cos(ut) = -1$  cancelling the offset created by the variables  $\omega$ ,  $\beta$ -value and  $u$ . So Eq. (20) becomes:

$$0 = -R(-\omega^2 + \beta_2(u + \omega)^2)$$

For the right hand side of the above equation to =0 either  $R$  or the contents within the brackets must =0. However the rotor radius  $R \neq 0$ , therefore:

$$0 = -\omega^2 + \beta_2(u + \omega)^2$$

$$\beta_2 = \frac{\omega^2}{(u + \omega)^2} \tag{23}$$

Eq. (23) represents  $\beta_2$  in terms of the angular velocities of the rotor and bobbin. It can be applied to a J-type centrifuge where  $u = \omega$ . Thus  $\beta_2 = 0.25$ , which agrees with that given in Refs. [13–15].

Also for an I-type centrifuge where the bobbin has the same rotational speed but in the opposite direction to the rotor, i.e.  $u = -\omega$ . Substituting in Eq. (23) for  $u$  shows that  $\beta_2$  for an I-type centrifuge also tends towards infinity.

Dividing the numerator and denominator of Eq. (23) by  $\omega^2$  gives:

$$\beta_2 = \frac{1}{(u + \omega)^2 / \omega^2}$$

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$$\beta_2 = \frac{1}{(u^2 + 2u\omega + \omega^2) / \omega^2}$$

$$\beta_2 = \frac{1}{(u^2/\omega^2 + 2u/\omega + 1)}$$

Since  $P_{Dr} = u/\omega$  then

$$\beta_2 = \frac{1}{(P_{Dr} + 2P_{Dr} + 1)^2}$$

$$\beta_2 = \frac{1}{(P_{Dr} + 1)^2} \tag{24}$$

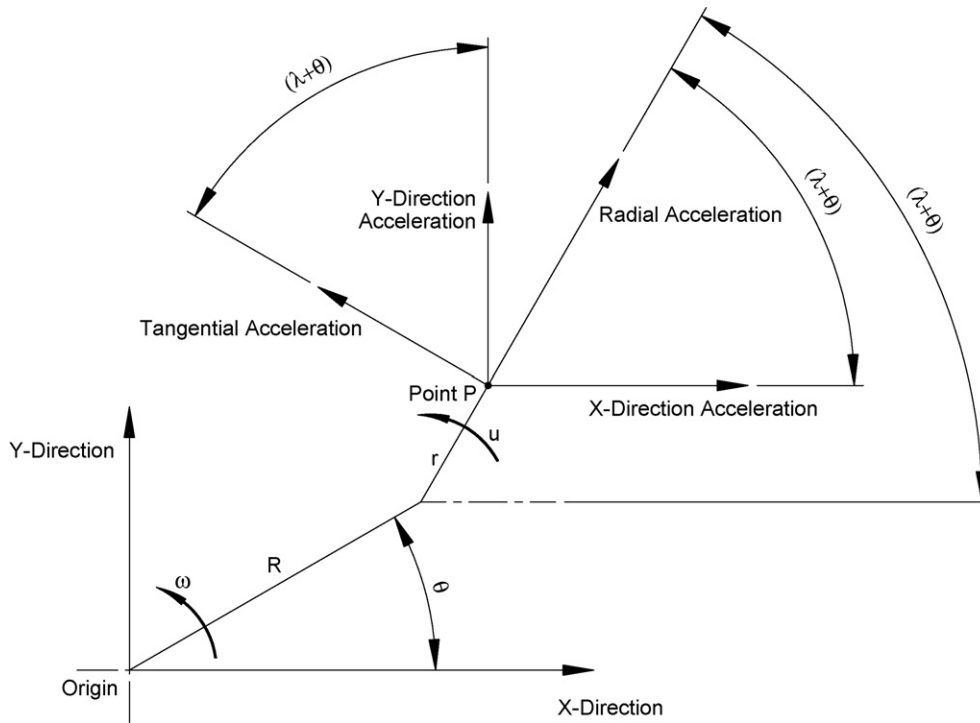


Fig. 6. The free-body diagram of a non-synchronous centrifuge with the radial and tangential acceleration vectors added.

2.4.3. Tangential acceleration

Substituting in Eq. (18) for  $A_x$  from Eq. (3) and for  $A_y$  from Eq (6) gives:

$$A_T = (R\omega^2 \cos(\omega t) + r(u + \omega)^2 \cos((u + \omega)t)) \sin((u + \omega)t) - (R\omega^2 \sin(\omega t) + r(u + \omega)^2 \sin((u + \omega)t)) \cos((u + \omega)t)$$

$$A_T = R\omega^2 \cos(\omega t) \sin((u + \omega)t) + r(u + \omega)^2 \cos((u + \omega)t) \sin((u + \omega)t) - R\omega^2 \sin(\omega t) \cos((u + \omega)t) - r(u + \omega)^2 \sin((u + \omega)t) \cos((u + \omega)t)$$

$$A_T = R\omega^2 [\sin((u + \omega)t) \cos(\omega t) - \cos((u + \omega)t) \sin(\omega t)]$$

Using  $\sin(A - B) = \sin(A) \cos(B) - \cos(A) \sin(B)$  where  $A = (u + \omega)t$  and  $B = \omega t$

$$\text{Therefore } \sin(ut) = \sin((u + \omega)t) \cos(\omega t) - \cos((u + \omega)t) \sin(\omega t)$$

$$\text{Therefore } A_T = R\omega^2 \sin(ut) \tag{25}$$

Since there is no negative sign in front of Eq. (25), the direction of  $A_T$  is in the same as shown in Fig. 6. Also as there is no bobbin radius ( $r$ ) term in Eq. (25),  $A_T$  is not influenced by the bobbin radius and consequently  $\beta$ -value.

For a J-type centrifuge, where  $u = \omega$ , Eq. (25) becomes:

$$\text{J-type } A_T = R\omega^2 \sin(\omega t) \tag{26}$$

Which agrees with the findings of Wood [13].

For an I-type centrifuge, the rotational velocity ( $u$ ) of the bobbin has the same magnitude as the rotational velocity ( $\omega$ ) as the rotor but is in the opposite direction, hence in Eq. (25)  $u$  can be replaced by  $-\omega$  and Eq. (25) becomes:

$$\text{I-type } A_T = R\omega^2 \sin(-\omega t)$$

$$\text{Since } -\sin(\theta) = \sin(-\theta):$$

$$\text{I-type } A_T = -R\omega^2 \sin(\omega t) \tag{27}$$

Which agrees with the findings of Wood [13].

3. Discussion

3.1. Archimedean screw effect and the I-type centrifuge

To date the Archimedean screw effect has been determined by only considering the rotation of a column about the column's axis. However, coil planet centrifuges rotated about 2 axes: the column's axis and the solar axis, the origin in Figs. 4–6. When determining the head and tail ends of a column the combined affect of rotation about these axes must be considered. Within the field of fluid mechanics coil planet centrifuges are considered as a rotodynamic machines. Analysing the fluid mechanics within such machines relies upon knowing the velocities of both the fluids and the channels through which the fluids flow. Therefore understanding the tangential velocity allows the head and tail ends of the column to be determined and thus the direction in which the upper and lower phases will flow.

Eq. (14) represents the tangential velocity for an I-type centrifuge, which takes the form of a cosine wave with a period of 1 revolution and amplitude  $R\omega$  and is shown in Fig. 7.

Considering Fig. 7, it can be seen that for half a revolution of the centrifuge, the tangential velocity is positive and for the other half it is negative. With respect to the Archimedean screw effect this means that for half the revolution the head is at one end of the column and for the other half of the revolution the head is at the other. Hence for half the time the upper and lower phases are pumped towards one end of the column and for the other half towards the

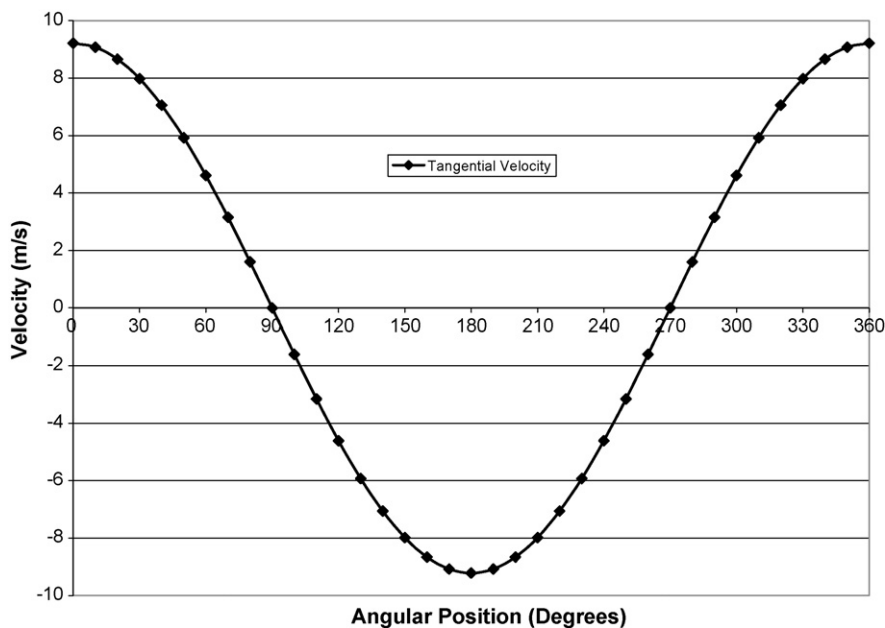


Fig. 7. Variation of the tangential velocity of an I-type centrifuge with angular position with a 110 mm rotor radius at a rotational speed of 800 rpm.

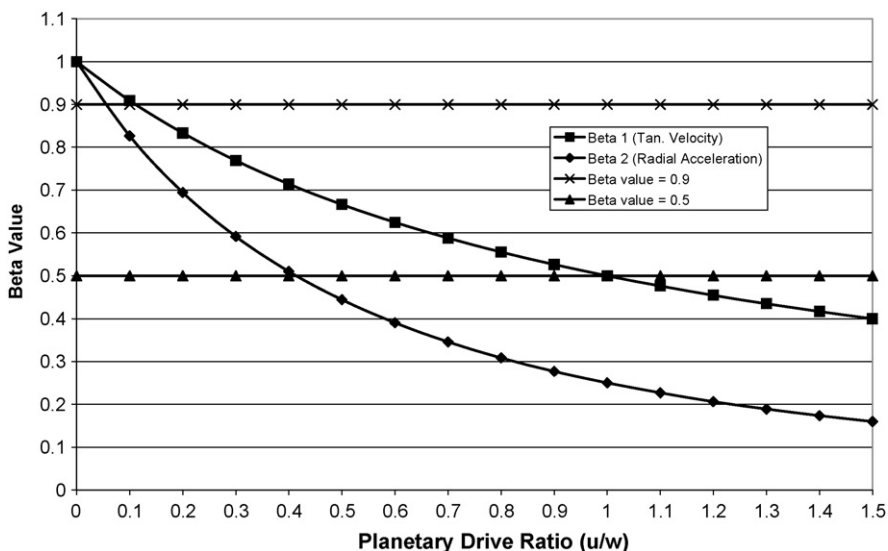


Fig. 8. The critical  $\beta$ -values for planetary drive ratios between 0 and 1.5.

other. In a helical column this would equate to an even (50:50) unilateral distribution of the upper and lower phases in each loop of the column (coil), when there is no flow of mobile phase. This may explain why the maximum stationary phase retention in an I-type centrifuge appears to be 50% [6,7] when there is no flow of the mobile phase.

### 3.2. The importance of critical $\beta$ -values ( $\beta_1$ and $\beta_2$ )

For both I-type and J-type centrifuges, critical  $\beta$ -values help to explain the type of mixing and the hydrodynamic distribution of upper and lower phases observed. For I-type devices, both critical  $\beta$ -values tend to infinity, which creates the conditions for cascade mixing to occur. Also, for a J-type centrifuge the critical  $\beta$ -values are 0.25 ( $\beta_2$ ) and 0.5 ( $\beta_1$ ). The lower  $\beta$ -value ( $\beta_2$ ) indicates the onset of wave mixing and the higher value ( $\beta_1$ ) shows when the unilateral distribution of the solvent phases becomes the same for every solvent system [2]. For a non-synchronous centrifuge it would be

extremely helpful to know the values of these critical  $\beta$ -values in order to understand how to perform a particular separation or understand results of stationary phase retention and resolution studies. Figs. 8 and 9 were both created using Eqs. (16) and (24). These figures are shown separately so that the scales of the vertical axis allow easy interpretation of the data.

Fig. 8 could be used to determine how to perform separations for positive planetary drive ratios. For consistent hydrodynamic behaviour<sup>2</sup> of all phase systems, the minimum  $\beta$ -value for a column is determined by the  $\beta_1$  curve that is determined by the tangential velocity. The  $\beta_2$  curve denotes the division between cascade and wave mixing as derived from the radial acceleration. At values below this curve, cascade mixing occurs and above it, wave mixing.

<sup>2</sup> The upper phase moves towards the head end of the column and the lower phase to the tail. Thus an upper mobile phase should be pumped from the tail to the head and a lower mobile phase is pumped in the opposite direction.



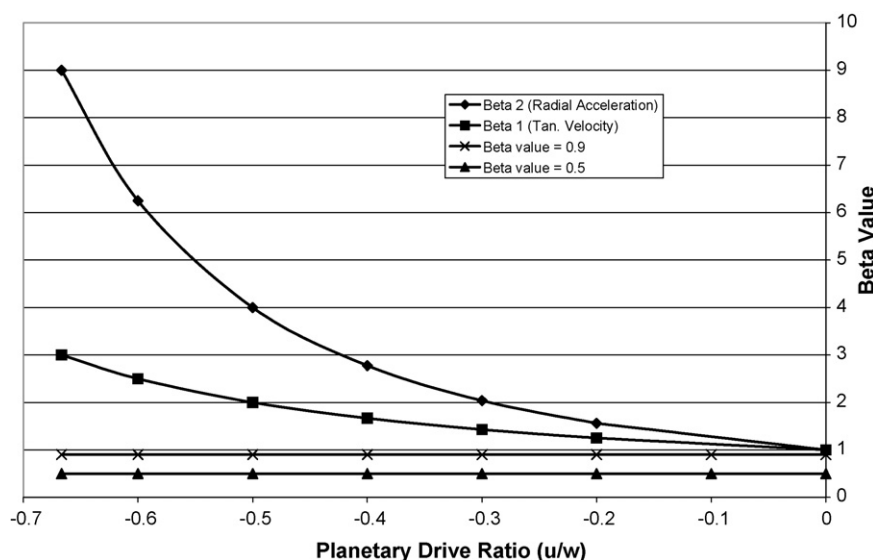


Fig. 9. Critical  $\beta$ -values for planetary drive ratios between 0 and  $-2/3$ .

If a non-synchronous centrifuge had a column with a  $\beta$ -value range of 0.5–0.9 then the results from both stationary phase retention studies and separations could be interpreted using Fig. 8 when the column was rotated in the same direction as the rotor. The horizontal lines denote the  $\beta$ -value range of 0.5–0.9 of such a column. Using Fig. 8 the following hypotheses can be made for a column with a  $\beta$ -value range of 0.5–0.9:

1. For planetary drive ratios equal to or greater than 1, the hydrodynamic behaviour of all phase systems will be the same (see footnote 2) and wave mixing will occur.
2. For planetary ratios between 0.4 and 1, wave mixing will occur but the hydrodynamic behaviour of hydrophilic phase systems will differ from that of intermediate and hydrophobic solvents systems. This is akin to the behaviour experienced with J-type centrifuges in the  $\beta$ -value range of 0.25–0.5.
3. Between planetary drive ratios of 0.05 and 0.4, a form of cascade mixing occurs, similar to that found in J-type centrifuges between  $\beta$ -values of 0 and 0.25.
4. Below a planetary drive ratio of 0.05, I-type cascade mixing occurs.

Fig. 9 shows how the critical  $\beta$ -values change with the planetary drive ratio when the column is rotated in the opposite direction to the rotor. The horizontal lines in Fig. 9 denote the  $\beta$ -value range of 0.5–0.9 for such a column. This figure demonstrates that the  $\beta$ -value range for multiple column (self-balancing) centrifuges will always be below both of the critical values. This means that cascade mixing will occur for all negative planetary drive ratios. Thus for a column with a  $\beta$ -value range of 0.5–0.9 only I-type cascade mixing would occur.

#### 4. Conclusions

This paper demonstrates the importance of understanding the influences that critical  $\beta$ -values have, for I-type, J-type and non-synchronous centrifuges, on:

- The unilateral (hydrodynamic) distribution of the upper and lower phases.
- The stationary phase retention.
- The type and effectiveness of the mixing and settling processes.

Hence the user of any coil planet centrifuge needs to understand where the critical  $\beta$ -values are in relation to the  $\beta$ -value range of the columns. Such knowledge will allow coil planet centrifuges to be used effectively to produce the best possible separations.

The Archimedean screw effect has been used to determine the head and tail ends of a column traditionally based solely upon the rotation of a column about its own axis. However, to truly determine the head and tail ends of a column requires an analysis of rotation about the column's axis and the main axis of the centrifuge. This has been achieved by determining the equation for the tangential velocity, Eq. (12), for all coil planet centrifuges. Thus it is the tangential velocity that determines the head and tail ends of a column and in turn the unilateral distribution of the upper and lower phases.

The tangential velocity, Archimedean screw effect, in J-type centrifuges does not always pump towards one end of a column [2]. For  $\beta$ -values between 0.25 and 0.5 the screw effect pushes the contents of the column towards one end of the column when the tangential velocity is positive and towards the other end when negative. For  $\beta$ -values above 0.5, see Fig. 3, the tangential velocity is always positive and the lower phase of any phase system moves towards the tail and the upper phase towards the head [2].

The tangential velocity of an I-type centrifuge spends half the rotation in the clockwise direction and the other half in the anti-clockwise direction. Thus the maximum stationary phase retention of 50% is caused by the reversal of the tangential velocity every  $180^\circ$  of rotation.

It is hypothesised that the types of mixing seen on most types of coil planet centrifuge (non-synchronous centrifuge and J-type) can be divided into three distinct regions. The first region is below a  $\beta$ -value as determined by  $\beta_2$  (Eq. (24)), which creates cascade mixing as in an I-type centrifuge. The next region is between the  $\beta$ -values  $\beta_2$  and  $\beta_1$  (Eq. (16)), currently the best description is enhanced wave mixing. The third region of mixing is above the critical  $\beta$ -value  $\beta_1$  where wave mixing occurs [3,4] as observed in a J-type centrifuge.

Stroboscopic photography is required to confirm the types of mixing that occur in each of the three regions created by the critical  $\beta$ -values, cascade mixing below  $\beta_2$  and wave mixing above  $\beta_2$ . Such photography would also demonstrate whether there is an observable difference in the mixing waves above and below  $\beta_1$ . To this end it is strongly recommended that a cantilever non-synchronous

centrifuge be built that allows such photography and can replicate the motion of all planetary drive ratios between  $-1$  (I-type) and  $1$  (J-type).

It is also recommended that Head and Tail studies similar to those in [2] are performed on a non-synchronous centrifuge fitted with two helical columns, each column to have a different  $\beta$ -value. The planetary drive ratio of this centrifuge would also replicate the motion of all planetary drive ratios between  $-1$  (I-type) and  $1$  (J-type). This would allow the unilateral distribution of hydrophobic, intermediate and hydrophilic solvent systems to be studied when each column is used within one of the three  $\beta$ -value ranges determined by  $\beta_1$  and  $\beta_2$ .

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### Glossary

*$\beta$ -value*: is defined as: the ratio of the radius ( $r$ ) of a point on a bobbin to the rotor radius ( $R$ ).

*Critical  $\beta$ -value*: provides a significant change in both the unilateral distribution of the upper and lower phases and the mixing and settling of the phases.

*Distal key node*: is the position when point  $P$  is furthest away from the main axis of the centrifuge or the origin in Figs. 4–6. In Figs. 3 and 7 the  $0^\circ$  and  $360^\circ$  positions along the horizontal axis represent the distal key node.

*Head end*: of a column is the end to which a bubble or a bead would move towards as the column was rotated in planetary motion.

*Hydrophilic (polar) phase systems*: generally have the following physical characteristics: low density differences between phases, low interfacial tension and higher viscosity phases.

*Hydrophobic (non-polar) phase systems*: generally have the following physical characteristics: high-density differences between phases, high interfacial tension and low viscosity phases.

*Intermediate phase systems*: generally have the physical characteristics between those of hydrophobic and hydrophilic phase systems.

*Proximal key node*: is the position when point  $P$  is closest to the main axis of the centrifuge or the origin in Figs. 4–6. In Figs. 3 and 7 the  $180^\circ$  position along the horizontal axis represents the proximal key node.

*Tail end*: of a column is the end from which a bubble or a bead would move away from as the column was rotated in planetary motion. The tail end is at the opposite end to the column to the head end.