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## Counter-current motion in counter-current chromatography \*

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#### ABSTRACT

After the CCC2012 meeting, I have received an e-mail regarding the terminology of "Countercurrent Chromatography". It stated that the term "Countercurrent" is a misnomer, because its stationary phase is motionless in the column and that the method should be renamed as liquid-liquid separations or centrifugal separations. However, it was found that these names are already used for various other techniques as found via Google search. The term "Countercurrent Chromatography" was originally made after two preparative methods of Countercurrent distribution and liquid Chromatography, both having no countercurrent motion in the column. However, it is surprising to find that this F1 hybrid method "Countercurrent Chromatography" can clearly exhibit countercurrent motion within the separation column in both hydrodynamic and hydrostatic equilibrium systems. This justifies that "Countercurrent Chromatography" is a proper term for this chromatographic method.

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#### 1. Introduction

After CCC2012 meeting, I have received an e-mail from Prof. Berthod stating "The technique denomination: countercurrent chromatography (CCC) is confusing by itself and needs some explanations. In normal CCC use, there is no countercurrent circulation of any fluid. The stationary phase is liquid but motionless, most often maintained steady by centrifugal fields. The liquid mobile phase is flown through the liquid stationary phase as in any other chromatographic techniques. The technique naming was coined in the early 1970s by its inventor, Yoichiro Ito, as a reference to the Craig's countercurrent distribution method [1]. Unfortunately, the countercurrent term is a misnomer. The term chromatography is correct but it frightens process chemists who do not like the cost and complexity supposedly attached to any preparative chromatographic techniques. Consequently, it is proposed to call the purification methods using liquid-liquid exchanges between a mobile and a stationary phase: liquid-Liquid Separations (LLS)". Since I named countercurrent chromatography, I am compelled to address my comments on this issue. In order to justify the new name for CCC suggested by the above letter, I first, consulted through Google

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search which will furnish the common usage of terms in various fields including science. The results are shown in Table 1.

In this table Google search items are each evaluated by the number of matches, the number of CCC listed in the first 10 matches and its major contents. The first search on liquid-liquid separation, that was suggested as a new term in the above letter, yielded over 10 million matches, but it does not contain CCC in the first 10 hits. Besides, this name is already given to a totally different technique for separating two liquid phases from emulsified liquid, and the method is extensively used in oil industries and dairy factories. Over one hundred of instruments have been developed for this purpose. Second, the centrifugal separation, that was suggested as an alternative new name for CCC, gave 15 million matches including no CCC in the first 10 matches. It contained various other centrifugal techniques. Next, liquid-liquid chromatography or liquid partition chromatography showed 15 – 1 million matches but without CCC in the first 10 matches. It contained mostly liquid chromatography with solid support. This clearly indicates that "liquid-liquid chromatography uses always solid support" is the common understanding. When the term "without solid support" was added to these terms, the name CCC starts to appear in the first 10 matches. And the search on countercurrent chromatography contained CCC in all of the first 10 matches. From the results of these Google searches, it is certain that changing the CCC name to liquid-liquid separations or centrifugal separations would produce tremendous confusion in the scientific community.

As mentioned in the above letter, the term "countercurrent chromatography" was coined in our Science publication in 1970

Table 1 Google search.

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Google search terms	No. of matches	Occurrences of CCC in first 10 matches	Most frequently matched topic(s)
Liquid–liquid separation	10,500,000	0	Separation of two phases
Centrifugal separation	15,000,000	0	Centrifugal technologies
Liquid chromatography	10,100,000	0	LC with solid support
Liquid-liquid chromatography	15,600,000	0	LC with solid support
Liquid partition chromatography	970,000	0	LC with solid support
Liquid–liquid chromatography without solid support	5,400,000	3	CCC, GC and LC
Liquid–liquid partition chromatography without solid support	244,000	6	Mostly CCC
Countercurrent distribution	170,000	5	CCC and CCD
chromatography Countercurrent chromatography	199,000	10	All CCC

[2]. At that time there were two major preparative separation techniques, namely, countercurrent distribution method (CCD) [1] and liquid chromatography. Then, we developed a new hybrid method which could share their merits with these two methods while eliminating their problems: CCC is more efficient than CCD while it provides higher yields by avoiding irreversible adsorption of

#### 2. Seal-free flow-through centrifuge systems

Fig. 1 shows a series of rotary-seal-free centrifuge systems developed for performing CCC [3]. In all these centrifuge systems a bundle of flow tubes from the cylindrical column holder supported at the upper end of the centrifuge axis is free from twisting so that the solvent can be continuously eluted through the rotating column without the conventional rotary seal device which may become a potential source of complications such as leakage and contamination. These systems are divided into three categories according to the motion of the column holder, i.e., synchronous (left column), non-planetary (middle column) and non-synchronous (right column). Among those the synchronous systems (left column) are mostly used for performing countercurrent chromatography. Therefore, two typical synchronous systems, type-I (left, top) and type-I (left, bottom) synchronous systems are chosen to illustrate countercurrent motion of the two liquid phases. In the type-I system (left, top), the column holder synchronously counter-rotates about its own axis while revolving around the centrifugal axis of the centrifuge whereas in the type-I system the column synchronously rotates in the same direction to the revolution and this planetary motion is used for high-speed CCC. As illustrated in Fig. 3, these two types of planetary motions produce countercurrent motion of the two phases in the coiled column but with totally different hydrodynamic distribution patterns.



Fig. 1. A series of rotary-seal-free centrifuge systems developed for performing CCC.

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