Title: Modelling a countercurrent liquid centrifuge and its application for high-throughput isotope separation

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Abstract: Large quantities of enriched natural isotopes have allowed for major advancements in every field of science upon entering the nuclear age. However, most elements are incompatible with high throughput methods like gas centrifugation and distillation since they do not have ambient gaseous molecules. Here, the recently uncovered method of liquid phase centrifugation is assessed in its capability for high throughput separation for all remaining elements. This is achieved by modelling a countercurrent liquid centrifuge and understanding its capabilities within a cascade. It is shown that there are remarkable similarities in performance and process engineering between a liquid and gas centrifuge cascade. Finally, the generality of a hypothetical cascade is shown by comparing the throughputs of multiple elements when used interchangeably in the same cascade.

Main Text:

1) **Introduction:**

Enriched isotopes have been, and are, essential to uncovering the mechanisms of science and for building new machines and devices. In particular, the large-scale separation of certain isotopes such as ^{235}U, ^{2}H, ^{13}C, ^{15}N, and ^{18}O have markedly accelerated science and technology. However, the remarkable technologies which allow for these high-throughput, low-cost separations are simply incompatible with most elements. Instead, electromagnetic isotope separation (EMIS) can

only produce isotopes at the 1 mg to 1 kg scale annually for most isotopes. These limitations hinder science in known and unknown ways, for example ^{48}Ca and ^{46}Ti.

Here, the recent method of liquid phase centrifugation is modelled and analyzed as a potential technology to facilitate the kg-ton separation of essentially all isotopes. This is done by understanding the behavior of a theoretical countercurrent liquid centrifuge which could continuously separate an incoming stream of multi-isotope fluid. In general, it is found that there are remarkable similarities between theoretical liquid centrifuges and existing gas centrifuges.

2) Equations and model

The movement of ions, atoms, and molecules inside a spinning centrifuge is governed by equations (1) and (2).

The diffusion, centrifuge, and advection terms are practically identical to the counterpart gas equations. The main difference here is electrostatic term to account for the interaction of ions, though this is zero for neutral species like pure liquids (H_2O and $TiCl_4$). The advection term now requires Navier-Stokes for incompressible fluids, rendering the equations analytically intractable, even in the steady-state. As such the equations require discretization and computation solutions.

… Full discussion here on the scheme. Including further discussions and details for L and F.

… Figure 1 is a schematic of the discretization scheme for a finite volume element within the centrifuge.

One advantage of solving the equations computationally is that this allows for the full transient results, thus allowing for the determination of the equilibrium time of machines. As far as the authors are aware, this is the first example of a full transient model of a countercurrent isotope centrifuge (liquid or gas) available in open literature. In general, the broad similarities between the governing equations for liquid and gas cases may lead to predictable similar results for the overall behavior of these systems.

Despite the similarities, there still several large fundamental differences which are likely to be impactful: 1) Fluid properties, including ρ, D, and ν. 2) Inducing the countercurrent. 3) Extracting the streams. 4) Wall stresses.

3) Results

Before discussing the results, it is worth mentioning that published analytical solutions exist for the steady-state gas case. These are discussed in more detail in the S.I... This means that these can be used as both a comparison and a validation for some of the expected trends of the state-state behavior.

Firstly, there are two broad cases to analyze: Pure liquids and dissolved salts. The pure liquid case will be assessed here since the results are easier to generalize across elements and molecules, although the counterpart analysis is presented in the S.I. for a selected salt ($CaCl_2$), and the generalization of this is discussed.

A centrifuge of height $Z = 100$ cm $r_i = 2.5$ cm, $r_o = 7.5$ cm, and $\omega = 60$ kRPM ($v = 471$ ms^{-1}) is used for all simulations where these parameters are not themselves varied. For L, $r_1 = 5.05$ cm and $r_2 = 7.45$ cm. For F, the source is at (4.5, 50) cm, and the two sinks are (7.25, 50 ± 45) cm.

Figure 2a shows α vs Z for F = 0, L = X, and v = 300, 400, and 500 ms^{-1}.

In real system the peripheral velocity is limited by the strength of the rotor walls and stresses induced within them as a result of the centrifugation.

Figure 2b shows α vs L for F = 0, Z = 0.5, 0.6, ..., 1.5 m. (Or k = 1, 2, 3, ...)

Figure 2c shows α vs F for Z = 1 m, L = 1, 2, ..., 10. (Or k = 1, 2, 3, ...)

Figure 2d shows δU vs F for L = 1, 2, ..., 10. (Or k = 1, 2, 3, ...)

The plot of Fig. 2d agrees well with the gas centrifuge case, and even has very similar values for the optimal ratio between the countercurrent and mass flow speeds (k). This means that F can be varied with predictable and small changes in δU. This is necessary for cascades and high-volume production. It can be noted that a value of F = 10 g/hr is equivalent to 87.6 kg/year per machine. Hundreds or thousands of such machines can assembled into a cascade to achieve high degrees of enrichment and throughput. Moreover, the results account for many liquids at once, such as water, $TiCl_4$, VCl_4, and $FE(CO)_5$.

Figure 3a shows α vs F for the optimal L and k.

Figure 3b shows α vs τ_{95} vs Z for optimal L, and faster L and F.

Figure 3d shows Flow and Machines vs Stage Number for 5% to 95% enrichment at n = 3, N_w = 1%, and p = 1 g/hr.

The results above are for a liquid and centrifuge with presumed typical values, but it is important to understand how all results vary as a function of other parameters: Diffusivity, D) High D allows for higher F (with same k), and therefore more throughput. Density, ρ) High ρ means greater throughput, but also more stress on the centrifuge walls and therefore a reduced speed is likely necessary. Rotational velocity, ω) Increased α and δU, but also increase the wall stresses. Centrifuge size, for same v) Reducing the radius reduces the equilibrium time by r^2, and volume/mass of the machine by r^3, and reduces the F and δU by r. Temperature, T) Lower temperature increases α, δU, and E, but this typically lowers D and increases ρ. Feed material price, $/kg) Cheaper feed materials mean N_w can be increased resulting in fewer centrifuges needed in the overall cascade. Mass difference, ΔM) Higher ΔM increases α and δU. The dependence for δU goes as $(\Delta M)^2$.

4) Discussion

In general, there are striking process engineering similarities to gas centrifuges. An interesting similarity to consider is that in both cases δU is proportional is $D\rho$. This means that for the same dimension of machine a direct comparison of the two methods can be made. In general, the throughput for gas phases is 1-2 orders of magnitude higher, which facilities Uranium enrichment on the scale of thousands of tons per year. Fortunately, tens of kilograms annually would be orders of magnitude above the global supply for practically all isotopes.

One key advantage of centrifugation in general is that it is element independent and its effectiveness depends on ΔM only, this is not the case for chemical exchange or numerous other methods. This means that the same cascade can be used for multiple elements or liquids, even simultaneously like H and O or Ti and Cl. For example, such a cascade could be used to meet new market demand in real time, instead of lagging behind by many years due to development costs and timelines.

In general, the gas technology is more mature, but the liquid case is more general although lower throughput, which should quell any nuclear proliferation concerns about the technology.

Figure 4a shows N_p vs Stage number for different ΔM.

Figure 4b shows a range of outputs and yields for numerous elements using the same cascade. This demonstrates the broad generality of such a cascade if built.

This could also lead to several unknown benefits due to the abundance of highly enriched isotopes. For example, advanced light-weight and high-strength spaceflight materials made from alloys of ^{46}Ti or ^{54}Fe, ^{6}Li in fueling fusion reactors, ^{192}Os as the densest stable isotope and ambient material, or ^{48}Ca for uncovering new physics.

5) Conclusion

Overall enriched isotopes have led to major advancements in science and technology. New isotopes would certainly lead to new known and unknown benefits for Earth's progress. Liquid centrifugation appears as a strong candidate to allow this due to its interchangeable uses across the periodic table. Moreover, the similarities to the gas case helps skill transfer and investment/development confidence. A single cascade could have huge positive impacts in its ability to supply new materials for all uses.

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Data and materials availability: All data are available in the manuscript or the supplementary materials. Information requests should be directed to the corresponding author.

Supplementary Information:

S1- Materials

Supplementary Text

Figs. S1 to SX

Tables S1 to SX

References