The Infeasibility of Enriching Lithium by Centrifuging

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Last revision: 2011-11-29

Molten salt and salt cooled reactors using a lithium salt in the primary loop require lithium highly enriched in the ^{7}Li isotope. Natural lithium is about 92.5% ^{7}Li and 7.5% ^{6}Li These reactors require lithium which is at least 99.95% ^{7}Li. This is needed to avoid the large neutron capture cross-section of ^{6}Li.

Given the almost 13% difference in density between ^{6}Li and ^{7}li, centrifuging would appear to be an obvious approach. Lithium melts at 181C, so this could be done at moderate temperatures.

The problem is that almost all liquid-liquid centrifuges work with immiscible fluids. ^{6}Li and ^{7}Li are totally miscible. So we have to go up against the Second Law, balancing the tendency of the isotopes to mix back together versus the gravitational forces separating them. Nonetheless with modern 100,000 RPM centrifuges capable of tangential velocities of 500 m/s perhaps this is feasible. Unfortunately, our investigation of this avenue indicates it is not.

Tsori and Liebler have derived the following governing equation for the profile of the volume fraction, $\phi(r)$ of one component of a binary system as a function of radial position in a centrifuge, r.[1]

$$\frac{k_B T}{v_o}\left(ln(\frac{\phi}{1-\phi}) + \chi(1-2\phi)\right) - 0.5 * \Delta\rho(\omega r)^2 - \mu = 0$$

In our case, ϕ is the volume fraction of ^{7}Li. k_B is the Boltzmann constant, T is the liquid temperature Kelvin, and v_o is the volume of the lithium atom. χ is the Flory parameter which we assume is zero (athermal mixing). $\Delta\rho$ is the difference in densities between ^{6}Li and ^{7}Li, and ω is the rotational velocity in radians per second.

The first term is the free energy of mixing per m^3; the second term is the gravitation separation energy per m^3. μ is a Lagrange multipler required to force the average composition in the centrifuge to the original, ϕ_0. Assuming χ is zero, Tsori and Liebler show that μ is given by

$$\mu = \frac{k_B T}{v_o} ln(\frac{\phi_0}{1-\phi_0}) - 0.25 * \Delta\rho\omega^2(R_{max}^2 + R_{min}^2)$$

where R_{max} and R_{min} are the outer and inner radius of the centrifuge.

A Perl script implementing this model is attached. It assumes a 100,000 RPM centrifuge with an outer radius of 0.05 m and an inner radius of 0.02 m. This corresponds to a tangential velocity at the outer radius of 524 m/s, about the best currently achievable with maraging steel in uranium gas centrifuges. The model optimistically assumes we can extract the composition at the outer radius and pass it on to the next stage.

Table 1 shows the results.

lithium radius=1.5200e-10			lithium volume=1.4710e-29		
lithium K=4.5800e+02			mixing factor=4.2986e+08		
$\Delta\rho$= 73.0 kg/m3			Initial Li-7 fraction= 0.925		
Inner radius= 0.02			Outer radius= 0.05		
Tangential velocity at outer radius= 523.6					
Stage	ϕ outer	μ	ctf energy	mix energy	error
1	0.92568	1.0741e+09	1.0012e+07	1.0842e+09	-2.7935e+02
5	0.92832	1.0910e+09	1.0012e+07	1.1010e+09	3.1867e+02
10	0.93151	1.1120e+09	1.0012e+07	1.1220e+09	-3.2570e+02
20	0.93749	1.1540e+09	1.0012e+07	1.1640e+09	1.8167e+00
50	0.95264	1.2802e+09	1.0012e+07	1.2902e+09	-2.7858e+02
100	0.97042	1.4904e+09	1.0012e+07	1.5005e+09	-3.9291e+02
200	0.98867	1.9109e+09	1.0012e+07	1.9210e+09	-7.8489e+02
500	0.99939	3.1726e+09	1.0012e+07	3.1826e+09	1.4563e+04
1000	0.99995	4.2513e+09	1.0012e+07	4.2571e+09	-4.2052e+06

Table 1: ^{7}Li enrichment as a function of number of stages

The second column is the ^{7}Li volume fraction leaving the stage. The fifth column is the re-mixing energy density. It is of the order of 2e9 J/m3. The separation energy density of this centrifuge is two orders of magnitude lower. Thus the concentration gradients that can be maintained are very small and rapidly become smaller as ϕ approaches 1.0 due to the $ln(\frac{\phi}{1-\phi})$ term. To put it another way, the average composition, represented by μ, must be almost the same as the composition at the outer wall. Accoring to this model, it would take over 500 stages to get to 99.95% ^{7}Li and nearly 1000 stages to get to 99.995%.

This simple model is almost certainly, highly biased in favor of centrifuging. It assumes:

[1] Tsori, Y. and Leibler, L., Phase Separation of miscible fluids in a centrifuge, C. R. Physique 8 (2007), pages 955-960.

1. We can extract only the liquid at the outside wall. Even if this could be done, the volume throughput of the cascade would be nil.
2. The centrifuge is essentially self-loaded. This may be true for gas centrifuges, but is certainly not true for a centrifuge filled with a liquid whose density is about $500 kg/m^3$.

Lithium enrichment by centrifuging appears to be a non-starter.

```
#!/usr/bin/perl -W
# cascade.pl
# this code does a cascade of centrifuges
# assuming one could pull out only the outermost fraction
# composition profile as a function of r
# assumes mixing is athermal, so Flory parameter is zero
# 2011-11-29 djw1 put in better atomic radius, decreased mixing energy by a factor of 3
#
use Math::Trig;
$UC{k}        = 1.38065e-23;  # Boltzman constant J K-1

# estimate volume of lithium atom in m3
$li_rad = 152.0e-12; # wiki says 152 pm
$li_vol = (4.0 / 3.0) * pi * $li_rad**3;
$li_K = 185 + 273; # just above melting point of 180.5C
$mix_factor = $UC{k} * $li_K / $li_vol;
printf("lithium: radius=%9.4e  vol=%9.4e K=%.3f  mix_factor=%9.4e\n",
        $li_rad, $li_vol, $li_K, $mix_factor);

# estimate density difference
$li7_atomic_mass = 7.01600;
$li6_atomic_mass = 6.01512;
$li_rho = 512.0;  # at melting point
$del_rho = (1.0 - $li6_atomic_mass/ $li7_atomic_mass) * $li_rho;
printf("Delta rho = %9.5f\n", $del_rho);

$phi_zero = 0.925; # initial Li-7 fraction
printf("phi zero = %6.4f\n", $phi_zero);

# centrifuge parameters, set to get a tangential velocity of about 500 m/s
$r_min = 0.02;
$r_max = 0.05;
printf("r_min = %6.4f r_max=%6.4f\n", $r_min, $r_max);
$rpm =  100000;
$omega = (2.0 * pi / 60.0) * $rpm;
$tan_v = $omega * $r_max;
printf("rpm= %7.0f tan_v=%8.2f\n", $rpm, $tan_v);
$ctf_energy = 0.5 * $del_rho * ($omega * $r_max)**2;     # separating energy per m3
printf("ctf_energy= %9.5e\n", $ctf_energy);

$nstages = 5000.0;
#$nstages = 200.0;

$texfile = "tbl_cascade.tex";
open(TEX, ">$texfile") or die "cant open $texfile: $!\n";
printf(TEX "\\begin{tabular}{rrr rrr}\n");
printf(TEX "\\multicolumn{3}{l}{lithium radius=%8.4e}", $li_rad);
printf(TEX "&\\multicolumn{3}{l}{lithium volume=%8.4e}\\\\\n", $li_vol);
printf(TEX "\\multicolumn{3}{l}{lithium K=%8.4e}", $li_K);
printf(TEX "&\\multicolumn{3}{l}{mixing factor=%8.4e}\\\\\n",  $mix_factor);
printf(TEX "\\multicolumn{3}{l}{\$\\Delta\\rho\$=%8.1f kg/m3}", $del_rho);
printf(TEX "&\\multicolumn{3}{l}{Initial Li-7 fraction=%6.3f}\\\\\n", $phi_zero);
printf(TEX "\\multicolumn{3}{l}{Inner radius=%6.2f}", $r_min);
printf(TEX "&\\multicolumn{3}{l}{Outer radius=%6.2f}\\\\\n", $r_max);
printf(TEX "\\multicolumn{6}{l}{Tangential velocity at outer radius=%6.1f}\\\\\n", $tan_v);
printf(TEX "Stage&\$\\phi\$ outer&    \$\\mu\$    &ctf energy&mix energy&  error  \\\\\n");
printf("Stage|  phi   |    mu    |ctf_energy|mix_energy|  error  |\n");

for ($stage = 1; $stage <= $nstages; $stage++) {
#
# we have to find phi(r_max) which solves Tsori equation
# we use brute force, dont go past 99.995%
#
    $best_error = 1.0e20;
    $mu = $mix_factor * log ($phi_zero / (1.0 - $phi_zero))
                - 0.25 * $del_rho * $omega**2 * ($r_max**2 + $r_min**2);
    for ($phi = $phi_zero - 0.01; $phi <= 0.99995; $phi += 0.0000001) {
        $mix_energy = $mix_factor * log($phi / (1.0 - $phi)); # mixing energy per m3

        $error = $mix_energy - $ctf_energy - $mu;
        if (abs($error) < abs($best_error)) {
            $best_error = $error;
            $best_phi = $phi;
            $best_mix_energy = $mix_energy;
        }
    }
    if ($stage == 1    || $stage == 5    || $stage == 10  ||
        $stage == 20   || $stage == 50   || $stage == 100  ||
        $stage == 200  || $stage == 500  || ($stage % 1000 == 0))  {
        printf("%5d|%8.5f|%8.4e|%10.4e|%10.4e|%10.4e|\n",
            $stage, $best_phi, $mu, $ctf_energy, $best_mix_energy, $best_error);
        printf(TEX "%5d&%8.5f&%8.4e&%10.4e&%10.4e&%10.4e\\\\\n",
            $stage, $best_phi, $mu, $ctf_energy, $best_mix_energy, $best_error);
    }
    $phi_zero = $best_phi;
}
printf(TEX "\\end{tabular}\n");
```