

Reports

Xenon-Iodine Dating: Sharp Isochronism in Chondrites

Abstract. *Measurements of the accumulation of Xe¹²⁹ from radioactive decay of extinct I¹²⁹ in meteorites show that the I¹²⁹/I¹²⁷ ratio in high-temperature minerals in diverse chondrites was 10⁻⁴ at the time of cooling. The uniformity in the ratio indicates that the minerals cooled simultaneously within 1 or 2 million years.*

Short-lived radionuclides, now extinct, which were present in the early solar system provide possible radioactive clocks which are unusually sensitive to small differences in time, occurring long ago. Among these nuclides is I¹²⁹ which decays with a 17 m.y. (million-year) half-life to Xe¹²⁹, which is detectable in meteorites by sensitive mass spectrometers (1-3). Chondrites constitute the most important class of stone meteorites; they are so-named because of the spherical silicate inclusions, or chondrules, they contain. We show here that the requirements for a successful dating method are fulfilled by the I¹²⁹ incorporated in high-temperature minerals in the chondrites. We infer that these minerals cooled simultaneously in diverse chondrites within a few million years, 4.7 billion years ago (4).

If a chondrite which has been irradiated by neutrons in an atomic pile is heated, there are, in the traces of xenon expelled in the heating, excess amounts of Xe¹²⁹ from a natural source and of Xe¹²⁸ implanted through the neutron irradiation by the reaction I¹²⁷(n,γβ)Xe¹²⁸. Proof that the excess Xe¹²⁹ is at iodine sites in the meteorite can be obtained by comparison of the thermal release patterns of the two isotopes (5). An especially simple way to search for the correlation is to plot the ratio Xe¹²⁹/Xe¹³² against Xe¹²⁸/Xe¹³² for the various fractions released in a stepwise heating (1 hour at progressively higher temperatures) of the irradiated sample (6). For a sample in which radiogenic (7) xenon (Xe^{129*}) resides only at iodine sites, in fixed proportion to the artificially implanted xenon (Xe^{128*}), that is, where Xe^{129*}/Xe^{128*} = K = a constant, and where other-

wise isotopes 128, 129, and 132 are present only in a single trapped component (subscript T) of uniform isotopic composition, one finds easily that

$$\frac{Xe^{129}}{Xe^{132}} = \left[\left(\frac{Xe^{129}}{Xe^{132}} \right)_T - K \left(\frac{Xe^{128}}{Xe^{132}} \right)_T \right] + K \left(\frac{Xe^{128}}{Xe^{132}} \right)$$

so that the curve obtained when the ratios are graphed is a straight line of slope K, passing through the point (Xe¹²⁸/Xe¹³²)_T, (Xe¹²⁹/Xe¹³²)_T. The quantity

$$K = \left(\frac{Xe^{129*}}{I^{127}} \right) \cdot \left(\frac{I^{127}}{Xe^{128*}} \right) = \left(\frac{I^{129}}{I^{127}} \right)_0 \cdot \left(\frac{I^{127}}{Xe^{128*}} \right)$$

also depends, through the second factor, on the magnitude of the neutron irradiation, which can be ascertained best by examining the xenon from a monitoring sample of iodine with the same neutron irradiation. With this factor known, the slope of the line gives (I¹²⁹/I¹²⁷)₀ which is of cosmological interest. The subscript zero refers to the value of the I¹²⁹/I¹²⁷ ratio at the time when the meteorite began to retain xenon.

Our colleagues at Berkeley have published curves of the above type (2, 3, 8, 9). A feature common to almost all such plots is that, in a stepwise heating, the highest temperature points indeed define a line, but lower temperature points fail to fall on this line by virtue of "too little" Xe^{129*}. These facts can be understood if some iodine occurs in the meteorite's high-temperature minerals which have bound xenon there very tightly and which are the last to break down in a stepwise heating, while other iodine resides in low-temperature

minerals which have lost xenon. Values of (I¹²⁹/I¹²⁷)₀ evaluated from the high-temperature data have been similar, as emphasized by Merrihue and Turner (10). But what has not been appreciated until now, when additional meteorites have been studied and a new monitor measurement has been completed, is that, for all chondrites studied at Berkeley, the high-temperature points fall on a single line, if the amounts of Xe^{128*} are corrected to account for differences in neutron irradiation of the samples. In other words, there seems to be a universal I-Xe isochron for high-temperature, iodine-bearing phases in chondrites. These phases occur in the chondrules in part, and possibly totally, because one of our samples is an individual chondrule.

Ten chondritic samples have been run at Berkeley in two separate pile irradiations (labeled α and γ—irradiation β was a technical failure); data for all appear in Table 1 and are plotted in Fig. 1. All data for release temperatures > 1100°C have been plotted (filled circles). For some samples, the line is "joined," starting at a lower temperature T_J. In these cases, points for all temperatures between T_J and 1100°C, inclusive, have been plotted as well (open circles). In the figure, the points for each irradiation fall very close to a single straight line. One exception must be noted: The points (sample "5") for a chip from the meteorite Bruderheim conform poorly to the line for irradiation α. But we know (11) that this meteorite was heated 500 million years ago to the extent that it lost 90 percent of its radiogenic Ar⁴⁰; we can expect that even some of the tightly bound radiogenic xenon was lost in this heating, which would account for the effect seen. A single chondrule pried out of Bruderheim (sample "B") falls on the isochron, nevertheless.

Both lines in Fig. 1 were fitted by the method of least squares (12) and by assigning weights for both x and y coordinates which corresponded to constant fractional errors in the measured isotope ratios. Sample "5" was ignored in the fitting. For samples "B" and "E" there is a detectable effect upon the Xe¹³⁴/Xe¹³² and Xe¹³⁶/Xe¹³² ratios by fission xenon generated in the neutron irradiation. Here, then, it was possible to correct the coordinates for pile-produced Xe¹³². The corrected points (not shown) differ only slightly from the uncorrected points shown in Fig. 1, but define a slightly better straight line for

irradiation γ . For the uncorrected points, $dy/dx = 2.843 \pm 0.055$; for the corrected points, $dy/dx = 2.899 \pm 0.049$. The errors quoted are standard deviations deduced from the least-squares fit (the quantity σ_b in reference 12). We have used the "corrected" line in the following calculations.

If the lines for irradiation α and irradiation β represent the same isochron, rotated because of different neutron exposures, the slopes of the lines should be in the same ratio as the quan-

ties in I^{127}/Xe^{128} determined for the iodine monitors. The ratio of the slopes γ/α is $(2.899 \pm 0.049)/(1.457 \pm 0.015) = 1.990 \pm 0.037$. For irradiation γ , Turner (3) determined the quantity (I^{127}/Xe^{128}) for the monitor by isotopic dilution in two samples of irradiated potassium iodide. His average value was $(2.75 \pm 0.15) \times 10^4$. The calibration constant for irradiation α has been in doubt ever since 1961, when determinations on two separate samples gave discordant values. Since other samples

of the monitor material have been carefully preserved, we have been able to resolve this problem with a new determination by better techniques; the new value is $(1.336 \pm 0.046) \times 10^4$, which agrees with one of the 1961 values within 6 percent. The ratio of the calibration constants γ/α is $(2.75 \pm 0.15)/(1.336 \pm 0.046) = 2.06 \pm 0.13$, which agrees with the ratio of slopes (1.990 ± 0.037) within experimental error.

In our model the lines for irradiation α and γ should intersect at the composition of the trapped component. It is an oversimplification to suppose that the high-temperature xenon fractions contain a unique trapped component. Whereas trapped xenon in chondrites has proved to be homogeneous in isotopic composition when the concentration of trapped gas is substantial (13), the aforesaid high-temperature minerals are unlikely sites for trapped gas. The "trapped" gas here is most likely a composite sample of authentic trapped xenon, atmospheric xenon from the extraction apparatus, and spallation xenon from cosmic-ray action on trace elements in the stone (14). We expect, then, that the ratios (Xe^{128}/Xe^{132}) and (Xe^{129}/Xe^{132}) for the intersection point will somewhat exceed the atmospheric ratios of 0.0714 and 0.983, respectively. The intersection at the point (0.111 ± 0.013) , (1.053 ± 0.034) is thus entirely reasonable.

Since the slopes and the intersection of the two lines in Fig. 1 are consistent with simultaneity for the cooling of high-temperature minerals in the chondrites, it is interesting to ask how sharply this simultaneity is defined. If we assume that I^{129} and I^{127} were uniformly mixed everywhere in these systems just before the first minerals cooled, the fraction I^{129}/I^{127} decayed thereafter with the 17 m.y. half-life of I^{129} . For two samples, then, $(I^{129}/I^{127})_1/(I^{129}/I^{127})_2 = \exp(\Delta t/\tau)$ where Δt is the time interval between the cooling of sample 1 and sample 2, respectively, and τ is the 25 m.y. mean life of I^{129} . On this basis $(I^{129}/I^{127})_\alpha/(I^{129}/I^{127})_\gamma = 1.035 \pm 0.070$ and $\Delta t = 0.8 \pm 1.6$ m.y. If we ask how the values of $(I^{129}/I^{127})_0$ for the nine individual meteorite samples (we omit sample "No. 5" in the discussion) are distributed about an average, we find that the mean deviation in this quantity is 10 percent, corresponding to a mean simultaneity of 2.5 m.y. Estimates of the sharpness of the simultaneity thus range from 0 to

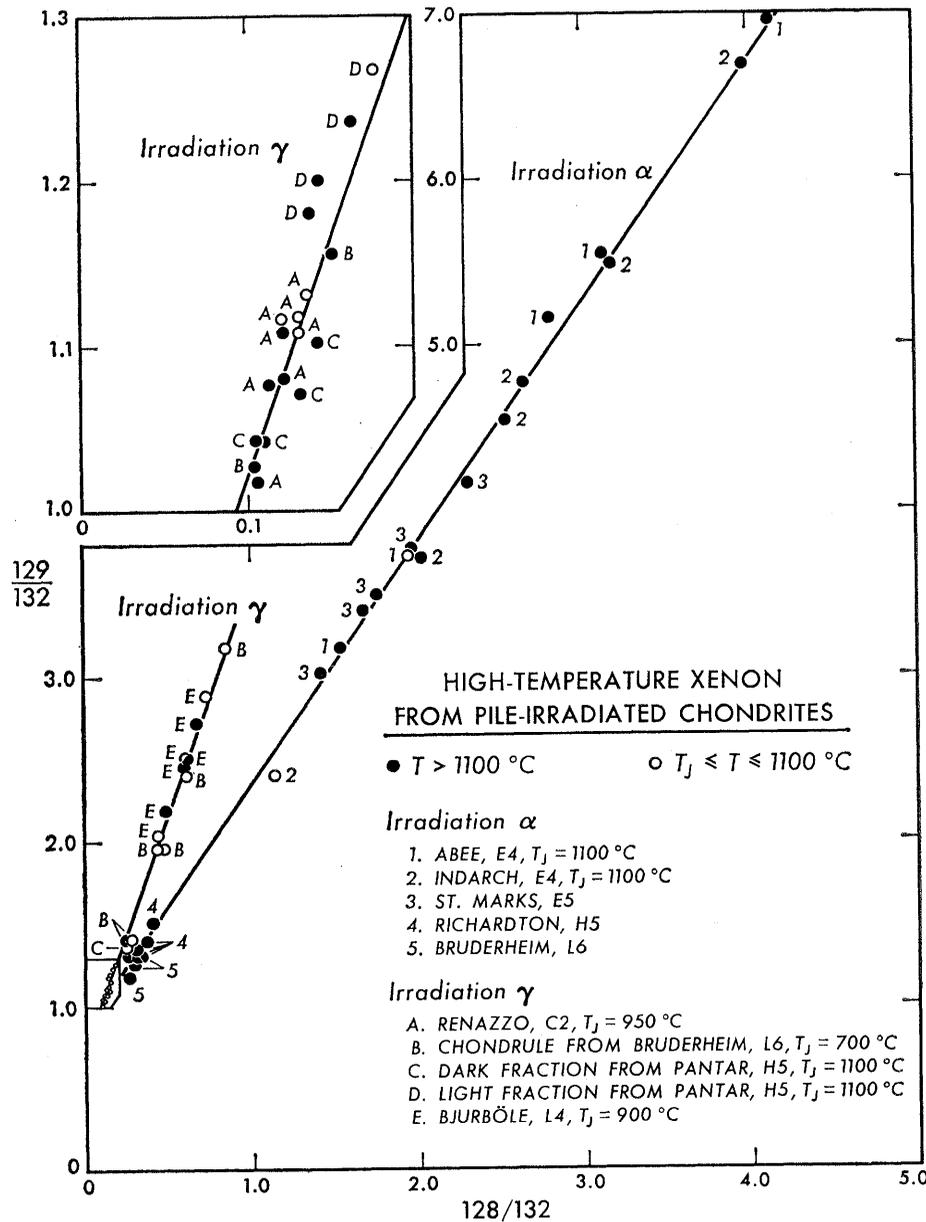


Fig. 1. The ratio of xenon isotopes (Xe^{129}/Xe^{132}) plotted against the ratio of isotopes (Xe^{128}/Xe^{132}) for traces of xenon expelled at the highest temperatures in a stepwise heating of various meteorite samples. All data above $1100^\circ C$ have been plotted; lower-temperature data are also plotted when the line is joined at a lower temperature T_j . Symbols after the meteorite name are classifications by Van Schmus and Wood (see text). The meteorites were all pile-irradiated before analysis, in two batches. For high-temperature xenon, the excess Xe^{129} from radioactive decay of extinct I^{129} is at the same sites in the meteorite as, and in fixed proportion with, the excess Xe^{128} produced in the pile by neutron absorption in I^{127} . Calculations show that the two lines would have coincided if the neutron irradiations in α and γ had been the same.

2.5 m.y. This sharpness is impressive when we consider that the cooling of the minerals took place 4.7 billion years ago. For irradiations α and γ , respectively, the quantity $(I^{129/127})_0$ is $(1.091 \pm 0.039) \times 10^{-4}$ and $(1.054 \pm 0.060) \times 10^{-4}$.

The origin of I^{129} in meteorites is an unsettled question. It could have been derived from the r -process in galactic synthesis along with I^{127} (15), or it could have been derived from spallation reactions or neutron absorption later in the history of the solar system (16), or it could have been derived from combinations of these sources. The universal association between I^{129} and I^{127} which we have found in this work for high-temperature minerals in chondrites does not answer this question, except as it shows that the production had to precede the formation of the minerals so that iodine mixing could occur. There has been considerable interest in a possible association between Xe^{129} and Te in meteorites. Such an association could result from the reaction $Te^{128}(n,\gamma\beta)I^{129}$ in neutron irradiation of material from the primitive solar system with subsequent xenon loss but only limited fractionation between iodine and tellurium before the formation of the meteorite. Correlations between Xe^{129*} and Xe^{131*} from $Te^{130}(n,\gamma\beta\beta)Xe^{131}$ in the pile could possibly demonstrate such an association, but in our experiment, as Merrihue has pointed out (9), conditions are very unfavorable for detection of such a correlation because Xe^{131*} is produced by pile neutrons on barium as well as on tellurium, and in the high-temperature minerals the barium source of Xe^{131*} could be more important. In any case scrutiny of our data (Table 1) for a correlation between Xe^{129*} and Xe^{131*} shows nothing like a universal correlation. There is no correlation in some samples (Abee, Indarch, Richardton, Pantar light fraction), there is a much weaker correlation than for Xe^{128*} in others (St. Marks, Bruderheim, Renazzo, Bruderheim chondrule, Pantar dark fraction), and a comparable correlation in one (Bjurböle); but none of the correlations for individual samples are shared by the others. It seems likely that these Xe^{131*} correlations, when found, are due to an association between barium or tellurium and iodine in the high-temperature minerals.

The chondrites in this study are of diverse types. Figure 1 lists the meteorite classifications according to a

scheme by Van Schmus and Wood (17). Represented are enstatite chondrites (E), a carbonaceous chondrite (C), and ordinary chondrites from both high (H) and low (L) iron groups. In addition, a considerable spread in inferred metamorphism (numerical scale, 2 to 6) is sampled. One of the samples is an individual chondrule from an ordinary chondrite. This sample and Bjurböle,

Table 1. Isotopic data for the high-temperature fractions of xenon gas evolved in stepwise heating of the meteorite samples. The first five meteorites received neutron irradiation α . The others received neutron irradiation γ . Heatings were usually for 1 hour each at temperatures increasing in steps of 100°C from 200°C. Precision of the ratio determinations is typically one percent.

| Temperature (°C) | Ratio | | |
|------------------------------|-------------------------------------|-----------------------------|-----------------------------|
| | $\frac{Xe^{128}}{Xe^{132}}$ | $\frac{Xe^{129}}{Xe^{132}}$ | $\frac{Xe^{131}}{Xe^{132}}$ |
| | <i>Abee*</i> | | |
| 1100 | 1.95 | 3.72 | 1.02 |
| 1200 | 3.13 | 5.55 | 0.991 |
| 1300 | 4.14 | 6.96 | .986 |
| 1400 | 2.81 | 5.16 | .943 |
| 1500 | 1.54 | 3.17 | .861 |
| | <i>Indarch†</i> | | |
| 1100 | 1.138 | 2.401 | 0.921 |
| 1200 | 2.027 | 3.714 | .930 |
| 1300 | 2.651 | 4.767 | .073 |
| 1400 | 2.544 | 4.541 | .947 |
| 1500 | 3.176 | 5.489 | .995 |
| 1600 | 3.985 | 6.689 | .989 |
| | <i>St. Marks‡</i> | | |
| 1200 | 2.310 | 4.161 | 1.071 |
| 1300 | 1.966 | 3.770 | 0.941 |
| 1400 | 1.758 | 3.493 | .909 |
| 1500 | 1.421 | 3.025 | .861 |
| 1600 | 1.678 | 3.404 | .889 |
| | <i>Richardton‡</i> | | |
| 1200 | 0.310 | 1.310 | 1.145 |
| 1300 | .400 | 1.51 | 1.067 |
| 1400 | .265 | 1.31 | 0.960 |
| 1500 | .305 | 1.35 | .911 |
| 1600 | .370 | 1.40 | .916 |
| | <i>Bruderheim§</i> | | |
| 1200 | 0.329 | 1.311 | 2.13 |
| 1300 | .292 | 1.259 | 1.54 |
| 1400 | .260 | 1.179 | 1.19 |
| | <i>Renazzo</i> | | |
| [See reference (8) for data] | | | |
| | <i>Bruderheim chondrule IBC-21¶</i> | | |
| 700 | 0.2799 | 1.408 | 1.552 |
| 800 | .4768 | 1.961 | 2.271 |
| 900 | .4358 | 1.960 | 1.794 |
| 1000 | .8524 | 3.173 | 2.668 |
| 1100 | .6035 | 2.401 | 1.842 |
| 1200 | .2468 | 1.410 | 1.096 |
| 1300 | .1513 | 1.156 | 0.861 |
| 1400 | .1032 | 1.026 | .811 |

Pantar, dark fraction
[See reference (3) for data]

Pantar, light fraction
[See reference (3) for data]

Bjurböle
[See reference (3) for data]

* Data in modified form plotted in references (2), (5), and (6). †This work. ‡Data in modified form plotted in reference (2). §Data in modified form tabulated in reference (9) and plotted in reference (2). ¶Data tabulated and plotted in modified form in reference (9).

which is a loosely cemented meteorite with a high proportion of chondrules, gave very similar results in irradiation γ . One of the meteorites (Pantar) is a gas-rich chondrite in which we have studied both the gas-rich dark fraction and the light fraction. The group is so diverse that it is difficult to find properties shared by these meteorites other than the property of containing chondrules. For this and other reasons, we think it likely that the high-temperature minerals under scrutiny are in the chondrules.

It is almost universally agreed that chondrules solidified from a silicate melt, that is, that they are of a high-temperature origin. The occurrence of glass in some chondrules in the less-metamorphosed stones is evidence of a quenched state for these chondrules (18). Rapid quenching could certainly provide the sharp simultaneity observed. Xenon-iodine studies of both chondrules and matrix minerals could settle questions about the location of the isochronous minerals.

Thus among the high-temperature minerals in the chondrites there are systems which meet the requirements for xenon-iodine dating, namely an initially uniform mix of I^{129} and I^{127} in well-defined proportions. Whether other systems for this dating method occur in nature remains to be seen.

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Rapid Rotation of the Solar Interior

Abstract. *A model proposed for the sun apparently can account for the maintenance of photospheric rotation against deceleration by the solar wind. The rotational period is about half a day near the top of the radiative mass, and 1.6 times shorter at the center.*

The solar wind produces a net flow in the radial (r) direction of the azimuthal (φ) component of angular momentum, τ_{φ}^r . According to independent estimates (1-3), this flux of angular momentum is approximately 1×10^8 g sec $^{-2}$ when averaged over the photosphere. The corresponding torque would be sufficient to halve the present angular momentum of the sun in 4×10^9 years, if the sun were in uniform rotation. Below the hydrogen convection zone (HCZ), however, the processes known to be available for the transport of angular momentum would all appear to be of low efficiency (4). Because the angular momentum of the HCZ alone has a half-life of less than 1×10^8 years, it is necessary to examine the processes that could support its continued rotation against the decelerating torque of the solar wind.

Several authors (1, 5, 6) have suggested that throughout most of the solar interior the rotational period is possibly much shorter than it is at the photosphere. Some evidence is also available for rapid rotation in the interiors of certain other main-sequence stars of solar type (7). If the angular velocity actually is large in the solar interior, then the radiation flowing from it could carry enough angular momentum to replace the losses in the solar wind. In a part of a star where the angular velocity is ω , Jeans showed that there is a radiative flux of angular

momentum that may be written approximately

$$(\tau_{\varphi}^r)_{\text{rad}} = (\omega r^2 \sin^2 \theta) F/c^2 \quad (1)$$

where θ is the polar angle, and F is the usual radiative flux of energy (8, 9). Outside of the energy-generating region at the center, the Jeans flux of angular momentum is therefore

$$\langle (\tau_{\varphi}^r)_{\text{rad}} \rangle = \frac{\omega L}{6\pi c^2} \quad (2)$$

when averaged over the sphere.

The arguments of Dicke (1), Deutsch (7), and Plaskett (6) led them to characterize the solar interior with a rotational period of the order of half a day. Let us adopt this value for $r/r_0 = 0.8$, at a level near the top of the radiative mass. The corresponding rotational velocity at this level is 100 km per second. We then find that $\langle (\tau_{\varphi}^r)_{\text{rad}} \rangle = 3 \times 10^7$ g sec $^{-2}$. This is to be compared with the solar-wind flux which, when reduced to $r/r_0 = 0.8$, is 2×10^8 g sec $^{-2}$.

The uncertainties of both calculations admit the conclusion that the HCZ can retain the slow angular velocity now seen at the photosphere, provided that the Jeans flux of angular momentum can be transferred from the radiation field to the matter near the base of the HCZ. This transfer can be effected by the radiative viscosity acting in an angular-velocity gradient in the outer part of the radiative mass. When the viscous transport of angular momentum is averaged over the sphere, it yields (10)

$$\langle (\tau_{\varphi}^r)_{\text{vis}} \rangle = \frac{2}{3} \eta r^2 (d\omega/dr) \quad (3)$$

The coefficient of radiative viscosity is (8, 11)

$$\eta_R = 4\sigma T^4/15 \kappa \rho c \quad (4)$$

At $r/r_0 = 0.8$ in the Schwarzschild-Weymann model of the sun (12), this is $\eta_R = 0.46$ g cm $^{-1}$ sec $^{-1}$, which is an order of magnitude larger than the coefficient of molecular viscosity. If we introduce this value into Eq. 3, and set $\langle (\tau_{\varphi}^r)_{\text{vis}} \rangle$ equal to the flux "observed" by Brandt and by Weber and Davis, we find that the requisite gradient is $d\omega/dr = 1 \times 10^{-13}$ sec $^{-1}$ cm $^{-1}$. The thickness of the transition zone that supports this gradient is then approximately

$$\delta r = \omega / |d\omega/dr| \quad (5)$$

evaluated at $r/r_0 = 0.8$. This turns out to be 1×10^4 km. Let us take $r = r_2$ at the inner boundary of the zone, and r_1 at the outer boundary.

We may expect that the outflow of angular momentum in the solar wind

would increase in proportion to ω_0^n , where $n \cong 1$, according to Weber and Davis (3). On the other hand, the radiative torque on the HCZ evidently depends only on the difference, $\omega_2 - \omega_0$, between the angular velocities at the inner boundary of the transition zone and at the photosphere. With $\omega_0/\omega_2 \ll 1$, the radiative torque is relatively insensitive to changes in ω_0 . The actual angular velocity of the HCZ therefore represents a true dynamical equilibrium between the opposing torques associated with the Jeans flux and the solar wind.

In view of the fact that the Jeans flux of angular momentum maintains the requisite flow of angular momentum into the base of the transition zone, there need be no appreciable gradient of angular velocity below this level. A model that seems to satisfy the requirements is that of Roxburgh (13), in which the angular velocity is a function $\omega(r)$ that rises gently from its value ω_2 at the top of the radiative mass to a maximum $\omega_c \cong 1.6 \omega_2$ at the center of the star. Roxburgh's model is characterized by vanishing meridional circulation, but it incorporates a toroidal magnetic field of the type first described by Biermann (14), which can arise from the battery effect of the centrifugal force on the free electrons. Everywhere in Roxburgh's model the gravitational force is large compared to the centrifugal force, and the centrifugal force is large compared to the magnetic body force. Over intervals of order 5×10^9 years, the magnetic field has insufficient time to reach its steady state (15). However, Roxburgh has also shown (16) that $\omega(r)$ can achieve its steady state within this time scale, provided that the angular momentum of the sun is high enough so that $r_2\omega_2 \cong 60$ km per second in the steady state—a condition probably satisfied.

Turning back to the transition zone near the top of the radiative mass, we may approximate its behavior by neglecting effects of compressibility and viscosity gradients, and idealizing the problem as follows: to find the steady motion of a uniform viscous liquid that is confined to the shell between two concentric spherical surfaces, the inner surface in rotation with constant angular velocity ω_2 , and the outer surface in rotation on the same axis with constant angular velocity $\omega_1 < \omega_2$. If Φ is the gravitational potential, the equation of motion for this problem is

$$\rho \mathbf{v} \cdot \nabla \mathbf{v} = -\rho \nabla \Phi - \nabla p + \eta \nabla^2 \mathbf{v} \quad (6)$$