Self-burial of radioactive waste

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The problem of the ''self-burial'' of radioactive waste into melting rock is solved for a spherical container of finite thickness. The mathematical model constructed, unlike the existing ones, takes into account the thermal losses to the solid rock and to the melt behind the container, as well as the reverse evolution of heat upon solidification of the melt. A calculation for the particular case of self-burial in granite shows that consideration of these factors significantly increases the maximum permissible radius at which the container will remain in the solid state and slows the burial rate. © *1999 American Institute of Physics.* [S1063-7842(99)02211-4]

One of the promising methods for the final disposal of radioactive waste is ''self-burial.'' Due to the evolution of heat accompanying radioactive decay, a container with radioactive waste melts the surrounding rock and sinks into it under the action of its own weight. The increase in the amount of heat expended on melting the rock during the accelerated downward migration in the initial stage ensures that the process will pass to a steady state. The problem of the steady migration of a spherical heat source was treated in Ref. 1 under the assumption that the temperature of its surface is uniform. As was shown in Ref. 2, this condition does not correspond to reality. The temperature reaches a minimum at the lower critical point and a maximum at the upper, diametrically opposite point. It also increases with increasing radius. Therefore, to keep a container in the solid state, its radius must not exceed a maximum permissible value at which the surface temperature reaches the melting point of the container. The dependence of the limiting radius and the corresponding maximum burial rate on the thickness and thermal conductivity of the container was investigated in Ref. 3. It was assumed in those studies that the heat flux in the direction opposite to the direction of motion can be neglected and that the heat flux in the direction of motion is completely expended on melting the medium. The reverse evolution of heat upon solidification of the melt behind the heat source was not taken into account. The purpose of the present work is to solve the problem of the self-burial of radioactive waste in a spherical container of finite thickness without these assumptions.

The stationary axisymmetric distributions of the temperature in the radioactive waste (T^i) and in the container wall (T^c) satisfy the equations

$$\frac{\partial}{\partial r}\left(r^2\frac{\partial T^i}{\partial r}\right)+\frac{\partial}{\partial \xi}\left[(1-\xi^2)\frac{\partial T^i}{\partial \xi}\right]=-\frac{q}{k_i}r^2,\quad r<R_i,$$

$$\frac{\partial}{\partial r}\left(r^2\frac{\partial T^c}{\partial r}\right)+\frac{\partial}{\partial \xi}\left[(1-\xi^2)\frac{\partial T^c}{\partial \xi}\right]=0,\quad R_i<r<R. \qquad (1)$$

Their solution with allowance for the conditions of continuity of the temperature and the heat flux at the inner surface of the wall can be written in the form[3]

$$T^i(r,\xi)=T_m+\frac{4}{3}\frac{h\nu}{c_p}\left[\frac{1}{8}\left(1-\frac{r^{*2}}{\zeta^2}\right)+\frac{k_i}{4k_c}(1-\zeta)+\sum_{n=0}^{\infty}\frac{C_n}{\Omega_n}r^{*n}P_n(\xi)\right],$$

$$T^c(r,\xi)=T_m+\frac{4}{3}\frac{h\nu}{c_p}\left\{\frac{k_i\zeta}{4k_c}\left(\frac{1}{r^*}-1\right)+\sum_{n=0}^{\infty}\frac{C_n}{(2n+1)\Omega_n}\left[\left(n+1+n\frac{k_i}{k_c}\right)r^{*n}+n\left(1-\frac{k_i}{k_c}\right)\zeta^{2n+1}r^{*-n-1}\right]P_n(\xi)\right\}. \qquad (2)$$

Hence follow the expression for Stefan's number

$$S(\xi)=\frac{c_p}{h}[T_w(\xi)-T_m]=\frac{4}{3}\nu\sum_{n=0}^{\infty}C_nP_n(\xi),$$

$$T_w(\xi)=T^c(R,\xi) \qquad (3)$$

and the expression for the heat flux from the source to the surrounding medium

$$-k_c\frac{\partial T^c}{\partial r}(R,\xi)=\frac{4}{3}\frac{h\nu k_i}{c_pR}\left[\frac{\zeta}{4}-\sum_{n=1}^{\infty}n\Gamma_nC_nP_n(\xi)\right]. \qquad (4)$$

Here

$$r^*=\frac{r}{R},\quad \xi=\cos\Theta,\quad \nu=\frac{c_pqR_i^2}{hk_i},\quad h=h_m+c_pT_m,$$

$$\Gamma_n=\frac{1}{\Omega_n}\left[1+\frac{n+1}{2n+1}\left(\frac{k_c}{k_i}-1\right)(1-\zeta^{2n+1})\right],$$

$$\Omega_n=1-\frac{n}{2n+1}\left(1-\frac{k_i}{k_c}\right)(1-\zeta^{2n+1}),\quad \zeta=\frac{R_i}{R},$$

C_n are arbitrary constants; r and Θ are spherical coordinates; $P_n(\xi)$ are Legendre polynomials; q is the heat output power of the radioactive waste; k_i and k_c are the thermal conductivities of the radioactive waste and the container; c_p, T_m, and h_m are the specific heat, melting point, and heat of the phase transition of the medium; R_i and R are the inner and outer radii of the container; and their ratio ζ is chosen from considerations of mechanical strength and is henceforth considered fixed for different values of R.

The region of the melt in front of the heat source ($\xi > 0$) forms a thin layer, in which flow is described by the methods of lubrication theory. In the reference frame associated with the source the velocity field and the pressure are specified by the equations[2]

$$v_\Theta = V\frac{y}{\delta^*}\left[1+\frac{3}{\delta^*}\left(1-\frac{y}{\delta^*}\right)\right]\sin\Theta,$$

$$v_r = -V\left(\frac{y}{\delta^*}\right)^2\left\{\left(3-2\frac{y}{\delta^*}\right)\cos\Theta + \frac{d\delta^*}{d\Theta}\left[\frac{1}{2}+\frac{3}{\delta^*}\left(1-\frac{y}{\delta^*}\right)\right]\sin\Theta\right\},$$

$$p = p_0 + 6\frac{\eta V}{R}\int_\Theta^{\pi/2}\frac{\sin\Theta}{\delta^{*3}}d\Theta,\quad y = r^*-1,\quad \delta^* = \frac{\delta}{R}, \tag{5}$$

where δ is the thickness of the layer, η is the viscosity coefficient, and V is the burial rate.

In the region of the melt behind the source ($\xi < 0$) the burial rate and pressure are assumed to be constant and equal to V and p_0, respectively. The tangential stresses on the surface of the source are small compared with the pressure. Therefore, the drag force of the melt equals

$$F = 2\pi R^2\int_{-1}^{1} p\xi\, d\xi = 6\pi\eta RVJ,$$

$$J = \int_0^1 (1-\xi^2)\,\delta^{*-3}\,d\xi. \tag{6}$$

Equating the difference between the weight and the buoyant force to this expression for the drag, we obtain the equation

$$VJ = \frac{2}{9}\frac{g}{\eta}R^2(\rho_1-\rho),\quad \rho_1 = \rho_c + \zeta^3(\rho_i-\rho_c), \tag{7}$$

where ρ_i, ρ_c, and ρ are the densities of the radioactive waste, the container material, and the medium, and g is the acceleration of gravity.

The main contribution to the integral J is made by a small vicinity about the point $\xi = 1$. Now setting

$$\delta^*(\xi) = \delta^*(1) - \delta^{*\prime}(1)(1-\xi)$$

and taking δ^* as the integration variable, we obtain up to the leading term

$$J = \frac{1}{\delta^*(1)[\delta^{*\prime}(1)]^2}. \tag{8}$$

The axisymmetric distribution of the temperature in the solid phase T^s satisfies the equation

$$-V\left(\xi\frac{\partial T}{\partial r}+\frac{1-\xi^2}{r}\frac{\partial T}{\partial \xi}\right) = \frac{a}{r^2}\left\{\frac{\partial}{\partial r}\left(r^2\frac{\partial T}{\partial r}\right)+\frac{\partial}{\partial \xi}\left[(1-\xi^2)\frac{\partial T}{\partial \xi}\right]\right\}. \tag{9}$$

According to the assumption made above, this equation is also valid for the distribution of the temperature T^f in the melt at $\xi < 0$. Its solution has the form

$$T^s(r,\xi) = \frac{1}{\sqrt{r^*}}\exp\left(-\frac{\beta}{2}r^*\xi\right)\times\sum_{n=0}^{\infty}E_nK_{n+\frac{1}{2}}\left(\frac{\beta}{2}r^*\right)P_n(\xi),$$

$$r > R+\delta,$$

$$T^f(r,\xi) = T_m + \frac{1}{\sqrt{r^*}}\exp\left(-\frac{\beta}{2}r^*\xi\right)\sum_{n=0}^{\infty}\left[F_nK_{n+\frac{1}{2}}\left(\frac{\beta}{2}r^*\right)+G_nI_{n+\frac{1}{2}}\left(\frac{\beta}{2}r^*\right)\right]P_n(\xi),$$

$$R < r < R+\delta,\quad \xi < 0, \tag{10}$$

where $K_{n+\frac{1}{2}}(x)$ and $I_{n+\frac{1}{2}}(x)$ are Bessel functions of imaginary argument, $\beta = VR/\mathfrak{a}$, and $\mathfrak{a} = k/\rho c_p$.

The density, specific heat, and thermal conductivity of the medium in the solid and liquid phases are assumed to be identical. The arbitrary constants E_n, F_n, and G_n are specified by the boundary conditions

$$T^s(R+\delta,\xi) = T^f(R+\delta,\xi) = T_m,$$

$$T^f(R,\xi) = T^c(R,\xi). \tag{11}$$

Confining ourselves to the leading terms of the asymptotic expressions of the Bessel functions

$$K_{n+\frac{1}{2}}(x) = \left(\frac{\pi}{2x}\right)^{1/2}\exp(-x),$$

$$I_{n+\frac{1}{2}}(x) = \frac{1}{(2\pi x)^{1/2}}[\exp(x)-(-1)^n\exp(-x)],$$

we have

$$T^s(r,\xi) = T_m\frac{1+\delta^*}{r^*}\exp\left[-\frac{\beta}{2}(r^*-1-\delta^*)(1+\xi)\right],$$

$$r > R+\delta,$$

$$T^f(r,\xi) = T_m + \frac{h}{c_p}\frac{S(\xi)}{r^*}\frac{1-\exp[-\beta(1+\delta^*-r^*)]}{1-\exp(-\beta\delta^*)}\times\exp\left[-\frac{\beta}{2}(r^*-1)(1+\xi)\right],$$

$$R < r < R+\delta,\quad \xi < 0. \tag{12}$$

From the boundary condition

$$-k\frac{\partial T^f}{\partial r}(R+\delta,\xi)=\pm h_m\rho V\xi-k\frac{\partial T^s}{\partial r}(R+\delta,\xi), \tag{13}$$

where $\pm h_m\rho V\xi$ is the quantity of heat spent on melting the medium in front of the heat source and given back upon solidification behind it, we obtain the equation of the phase boundary at $\xi<0$

$$(1+\delta^*)[D+(3D-1)\xi]+\frac{2D}{\beta}$$
$$=\frac{S(\xi)}{1-\exp(-\beta\delta^*)}\exp\left[-\frac{\beta}{2}\delta^*(1+\xi)\right],$$
$$D=\frac{c_pT_m}{2h} \tag{14}$$

and the expression for the heat flux from the melt into the solid phase at $\xi>0$

$$-k\frac{\partial T^f}{\partial r}(R+\delta,\xi)=\frac{kh}{c_pR}[(1-D)\beta\xi+D(\beta+2)]. \tag{15}$$

The distribution of the temperature in the melt in front of the heat source is found by a parametric method of boundary-layer theory. Integrating the heat conduction equation

$$v_r\frac{\partial T^f}{\partial y}+v_\Theta\frac{\partial T^f}{\partial\Theta}=\frac{\mathfrak{a}}{R}\frac{\partial^2T^f}{\partial y^2}$$

over the thickness of the layer and taking into account the continuity equation

$$\sin\Theta\frac{\partial v_r}{\partial y}+\frac{\partial}{\partial\Theta}(v_\Theta\sin\Theta)=0,$$

we obtain the integral relation

$$\left[\int_0^{\delta^*}v_\Theta T^f\sin\Theta\,dy\right]'+VT_m[\xi-(1-\xi^2)\delta^{*\prime}]$$
$$=-\frac{a}{R}\left[\frac{\partial T^f}{\partial y}(\delta,\xi)-\frac{\partial T^f}{\partial y}(0,\xi)\right]. \tag{16}$$

We approximate T^f by a trinomial, which is quadratic with respect to y and whose coefficients are defined by the conditions (11) and (15),

$$T^f(y,\xi)=T_m+\frac{h}{c_p}\left(1-\frac{y}{\delta^*}\right)$$
$$\times\left\{S+y\left[D(\beta+2)+(1-D)\beta\xi-\frac{S}{\delta^*}\right]\right\}. \tag{17}$$

The relation (16) with allowance for (5) and (17) gives the differential equation for δ

$$\frac{1-\xi^2}{20}\left[D(\beta-8)+(1-D)\beta\xi+\frac{5}{6}S\right]\delta^{*\prime}=\varphi-\frac{S}{\beta\delta^*},$$

$$\varphi=D\left(1+\frac{2}{\beta}\right)+(1-D)\xi-\frac{3}{40}[(1-\xi^2)S]', \tag{18}$$

whence it follows that up to the leading term we have

$$\delta^*=\frac{S}{\beta\varphi}. \tag{19}$$

The prime sign denotes a derivative with respect to ξ. Using (8) and (19), we bring Eq. (7) into the form

$$S(1)=\frac{9}{2}\frac{\gamma\beta(\beta+2D)^3}{[S'(1)]},\quad \gamma=\frac{\mathfrak{a}\eta}{gR^3(\rho_1-\rho)}. \tag{20}$$

From (12) and (14) we find

$$-k\frac{\partial T^f}{\partial r}(R,\xi)=\begin{cases}\dfrac{kh\beta}{c_pR}\left\{D\left(1+\dfrac{2}{\beta}\right)+(1-D)\xi-\dfrac{3}{20}[(1-\xi^2)S]'\right\}, & \xi\geqslant0\\[2ex] \dfrac{kh\beta}{c_pR}\left\{\left(\dfrac{1+\xi}{2}+\dfrac{1}{\beta}\right)S+\left[(1+\delta^*)(D+3D\xi-\xi)+\dfrac{2D}{\beta}\right]\times\exp\left[-\dfrac{\beta}{2}\delta^*(1-\xi)\right]\right\}, & \xi<0.\end{cases} \tag{21}$$

To determine the constants C_n we have the boundary condition

$$k_c\frac{\partial T^c}{\partial r}(R,\xi)=k\frac{\partial T^f}{\partial r}(R\xi). \tag{22}$$

In the limiting case of $\beta=0$ and $\delta=0$, it follows from (14) and (19) that $S=0$ and $C_n=0$, i.e., we obtain the stationary solution for an immobile container with a surface temperature T_m. The heat flux (21) in all directions reduces to kT_m/R. According to (4) and (22), the radius of such a container equals

$$R_0=\left(\frac{3kT_m}{q\zeta^3}\right)^{1/2}. \tag{23}$$

Thus, self-burial is possible under the condition $R>R_0$. As R is increased, the exponential functions in (21) rapidly decrease; therefore, we shall henceforth neglect the corresponding terms. The resulting error in the determination of the burial rate is not more than V_0, i.e., the value obtained for $R=R_0$. Multiplying (22) by $P_n(\xi)$ and integrating over ξ from -1 to $+1$, we obtain the infinite system of equations

$$\frac{4n\chi}{(2n+1)\beta}C_n+\sum_{k=0}^{\infty}C_k\left\{\frac{3}{10}P_k(0)P_n(0)+\frac{n}{2n+1}\left[\frac{3}{10}(n+1)-(-1)^{k+n}\right]\Psi_{k,n-1}-\frac{n+1}{2n+1}\left[\frac{3}{10}n+(-1)^{k+n}\right]\Psi_{k,n+1}+(-1)^{k+n}\left(1+\frac{2}{\beta}\right)\Psi_{k,n}\right\}=\frac{\chi}{2}\frac{\zeta}{\beta}\int_{-1}^{1}P_n(\xi)\,d\xi-\frac{3}{2\nu}Q_n,$$

$$Q_n=D\left(1+\frac{2}{\beta}\right)\Psi_{0,n}+(1-D)\Psi_{1,n},$$

$$\Psi_{k,n}=\int_0^1 P_k(\xi)P_n(\xi)\,d\xi,\quad \chi=\frac{k_i}{k},\quad n=0,1,\ldots\,. \tag{24}$$

We confine ourselves to a finite number N of the constants C_n and the first N equations. For $N=2$ we have

$$C_0=\frac{3}{\nu}\frac{\Delta_0}{\Delta},\quad C_1=-\frac{3}{\nu}\frac{\Delta_1}{\Delta};$$

$$S(1)=\frac{4}{3}\nu(C_0+C_1),\quad S'(1)=\frac{4}{3}\nu C_1;$$

$$\Delta_0=\frac{2}{3}\chi\zeta\nu\left(1+2\chi\Gamma_1+\frac{19}{80}\beta\right)-\frac{1}{2}(7+8\chi\Gamma_1)D-\left[1+\frac{59}{40}D+\chi\Gamma_1(1+D)\right]\beta-\frac{1}{480}(97+77D)\beta^2,$$

$$\Delta_1=\chi\zeta\nu\left(-1+\frac{1}{30}\beta\right)+6D+\frac{1}{20}(35+47D)\beta+\frac{1}{40}(15+7D)\beta^2,$$

$$\Delta=1+8\chi\Gamma_1+\frac{1}{20}(43+64\chi\Gamma_1)\beta+\frac{119}{300}\beta^2. \tag{25}$$

The parameter β is found from the equation

$$\Delta_0-\Delta_1=\frac{\Delta}{4}S(1), \tag{26}$$

where, according to (20),

$$S(1)=\frac{9\chi}{32}\left(\frac{\Delta}{\Delta_1}\right)^2\beta(\beta+2D)^3. \tag{27}$$

Since $S(1)\ll 1$, Eq. (26) reduces to a quadratic equation. When $R>R_0$, it has one positive root

$$\beta(\nu)=\frac{1}{277+161D}\{[36L^2+(277+161D)M]^{1/2}-6L\},$$

$$L=110+153D+40\chi\Gamma_1(1+D)-5\chi\zeta\nu,$$

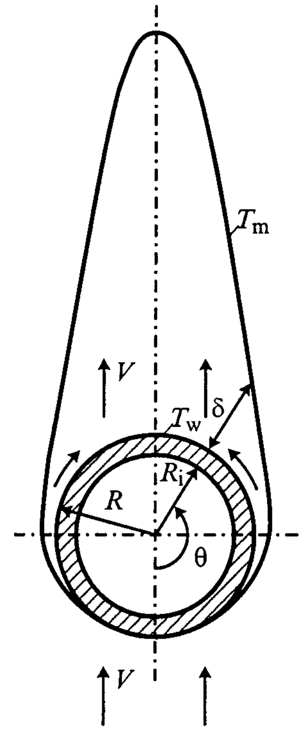

FIG. 1. Physical model of the geometry of the problem.

$$M=\frac{1}{3}\chi\zeta\nu(5+4\chi\Gamma_1)-\frac{1}{2}(19+8\chi\Gamma_1)D, \tag{28}$$

which specifies the dependence of the burial rate on the radius and heat output power of the heat source, as well as the physical characteristics of the medium. According to (2), the maximum temperature is achieved on the line $\xi=-1$ at the point

$$r_m^*=-4\frac{\zeta^2}{\Omega_1}C_1; \tag{29}$$

and equals

$$T^i(r_m^*,-1)=T_m+\frac{h\nu}{6c_p}\left[1+2\frac{k_i}{k_c}(1-\zeta)+\frac{r_m^*}{\zeta}(r_m^*+2\Omega_1)\right]. \tag{30}$$

The container has its highest temperature at the upper critical point on the inner surface of the wall:

$$T^c(R_i,-1)=T_m+\frac{4}{3}\frac{h\nu}{c_p}\left[\frac{k_i}{4k_c}(1-\zeta)+\left(1+\frac{\zeta}{\Omega_1}\right)C_0\right]. \tag{31}$$

Equating it to the melting point of the container T_*, we find the maximum permissible value of ν:

$$\nu_*=\frac{3}{G}\left\{\left(1+\frac{\zeta}{\Omega_1}\right)[876D+(277+161D)\beta_*]-S_*(445+464\chi\Gamma_1)\right\},$$

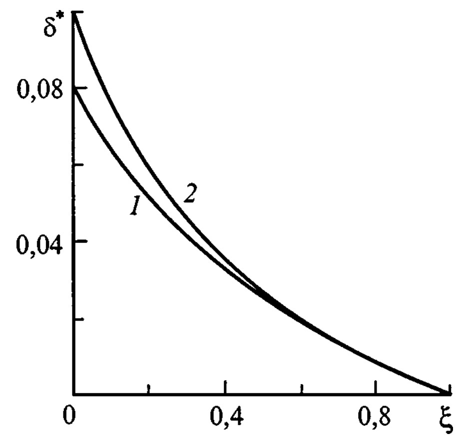

FIG. 2. Thickness of the melt zone at $\xi>0$.

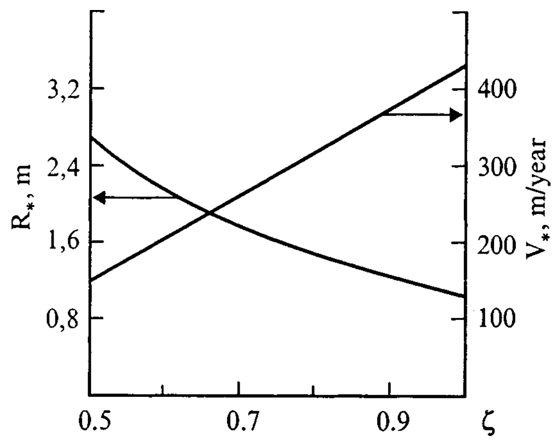

FIG. 4. Plots of the dependence of the maximum radius and burial rate on ζ.

$$G=180\chi\zeta\left(1+\frac{\zeta}{\Omega_1}\right)-\frac{k_i}{k_c}(445+464\chi\Gamma_1)(1-\zeta),$$

$$S_*=\frac{c_p}{h}(T_*-T_m). \tag{32}$$

Substituting it into (26), we obtain a quadratic equation for the corresponding value $\beta=\beta_*$. According to the definitions of these parameters, the maximum radius and burial rate are found using the formulas

$$R_*=\frac{1}{\zeta}\left(\frac{hk_i}{c_pq}\nu_*\right)^{1/2}, \quad V_*=\frac{a}{R_*}\beta_*. \tag{33}$$

When $N>2$, the problem requires a numerical solution.

Let us consider the case of the self-burial of radioactive waste housed in a container composed of the high-temperature ceramic NbC in granite when $q=130\,000$ W/m^3. We take the following values for the physical constants (in SI units):[4] $\rho=2700$, $c_p=1301$, $k=3.013$, $h_m=585\,800$, $T_m=1200\,°\mathrm{C}$, $\eta=10$; $\rho_c=7820$, $k_c=44$, $T_*=3480\,°\mathrm{C}$; $\rho_i=7800$, $k_i=36$.

For $\zeta=0.9$ we obtain $R_*=1.221$ m, $V_*=376.28$ m/year, $S(1)=0.855\times10^{-4}$, $S'(1)=-0.684$, $\delta^*(1)=0.359\times10^{-5}$, and $\delta^*(-1)=4.012$.

At the point $\xi=0$ the expressions (14) and (19) give fairly close values: $\delta^*(-0)=0.092$ and $\delta^*(+0)=0.099$. At $\xi=1$ the container is in direct contact with the solid medium and has a temperature exceeding its melting point by 0.14 °C. The maximum temperature within the container at the point $r_m^*=0.639$, $\xi=-1$ equals 3540.96 °C. The melt zone and the computational model of flow are shown in Fig. 1. In Fig. 2 curve *1* corresponds to the boundary of the melt zone in the region $\xi>0$ defined by Eq. (18), and curve *2* corresponds to the approximate formula (19). In the range $0.5<\xi\leqslant1$ they essentially coincide. The maximum difference between them at $\xi=0$ amounts to 0.019. Since the motion of the container depends mainly on the conditions in the vicinity of $\xi=1$, the accuracy of formula (19) is fully satisfactory. Formula (6) with allowance for (19) gives $J=139.04\times10^6$, and the value obtained from the approximate formula (8) is 0.05% higher. According to (23), $R_0=0.338$ m corresponds to a zero burial rate. Figure 3 shows a plot of $V(R)$ specified by formula (28). For the radius R_0 it gives $V_0=68.39$ m/year, which represents the maximum error caused by neglect of the exponential term in (21). As ζ is increased from 0.5 to 1, the radius R_* decreases from 2.683 to 1.025 m, and V_* increases roughly according to a linear law from 150.08 to 429.94 m/year (Fig. 4). A comparison with the results in Ref. 2 for $\zeta=1$ shows that consideration of the heat flux and the reverse evolution of heat upon solidification of the melt at $\xi<0$ increases R_* by 0.298 m and diminishes V_* by 44 m/year. The melt zone behind the container becomes 1.5 times longer.

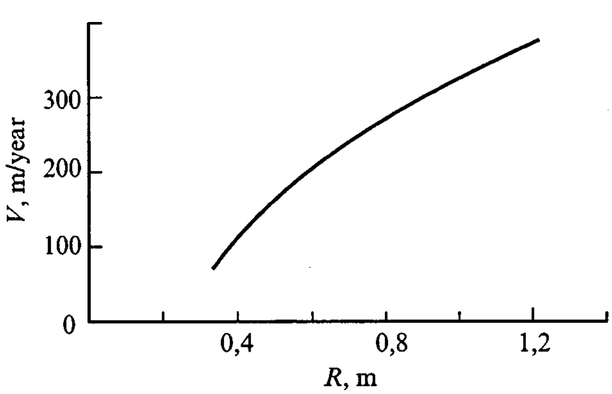

FIG. 3. Plot of the dependence of the burial rate on the container radius for $\zeta=0.9$.

The results obtained depend weakly on the choice of the value of N. For example, for $N=10$ the values of R_* and V_* increase by 1.53% and 0.51%, respectively, and for $N=100$ they increase by 1.62% and 0.56%.

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