Problem VI.5 ... gadolinium sphere

9 points; průměr 3,00; řešilo 34 studentů

What is the smallest amount of gadolinium 148 *needed to put together to cause local melting from the heat generated by its nuclear decay? Assume that only α decays take place and the material is at room temperature in the air.*

Karel was thinking about elements, but Matěj Rz. changed that.

The task combines multiple phenomena

1. the nuclear decay,
2. the interaction of radiation with matter,
3. the heat transfer.

Theory

The theory concerning nuclear decay and the interaction of radiation with matter needed for this problem is explained in problem T.5 in the Fyziklani 2021.[1] The nucleus of ^{148}Gd is unstable and decays. Since it is a heavy nucleus, it α decays[2]

$$^{148}_{64}\mathrm{Gd} \longrightarrow {}^{4}_{2}\mathrm{He} + {}^{144}_{62}\mathrm{Sm}\,.$$

The decay releases a significant amount of energy (in the literature also known as "Q-value") – in our case 3 271 keV per 1 decay.[3]

In nuclear decay, it should be mentioned that the daughter nucleus[4] can also decay. We do not need to deal with it because the problem assignment says the sphere is made of ^{148}Gd. If we were to consider the time dependence, the problem would get more complicated. We have the simpler case – ^{148}Gd decays only by alpha[5] decaying and the daughter ^{144}Sm is stable.[6] In case that we would have a cascade of successive decays

$$^{\mathrm{A}_1}_{\mathrm{Z}_1}\mathrm{X}_1 \longrightarrow {}^{\mathrm{A}_2}_{\mathrm{Z}_2}\mathrm{X}_2 \longrightarrow {}^{\mathrm{A}_3}_{\mathrm{Z}_3}\mathrm{X}_3 \longrightarrow {}^{\mathrm{A}_n}_{\mathrm{Z}_n}\mathrm{X}_n\,,$$

we would get a system of differential equations, called Bateman equation.[7] By solving them, we would get the time dependence of the number of nuclei $N_i(t)$, or rather their activities $A_i(t)$ and eventually their heat power over time.

The α particles are heavy and charged, so they interact intensely with matter, thus stopping quickly. Specifically, we are referring to the electromagnetic interaction of the α particle (with a charge $+4\,\mathrm{e}$) with charged parts of the surrounding matter (nuclei, electrons), which leads to the ionization of the surrounding environment. To give you an idea of how fast the α particle interacts, consider that its mean free path in the air is bare 20 cm or (according to the well-known fact[8]) can be stopped by a sheet of paper. Because of this, we can assume that the newly formed α particles will stop already in the material (Gd) and transfer all its energy to it. The sum of the energies of all the decays then leads to a non-negligible thermal power, which heats

[1] `https://fyziklani.cz/download/2021/solutions.pdf`
[2] `http://nucleardata.nuclear.lu.se/toi/nuclide.asp?iZA=640148`
[3] `http://nucleardata.nuclear.lu.se/toi/nuclide.asp?iZA=640148`
[4] "maternal nucleus" is original (on the left side of the equation), "daughter" is newly formed (on the right side of the equation)
[5] `http://nucleardata.nuclear.lu.se/toi/nuclide.asp?iZA=640148`
[6] `http://nucleardata.nuclear.lu.se/toi/nuclide.asp?iZA=620144`
[7] `https://en.wikipedia.org/wiki/Bateman_equation`
[8] `https://commons.wikimedia.org/wiki/File:Alfa_beta_gamma_neutron_radiation.svg`

the material[9] The heat power to its volume P_V can then be derived directly from the definition of

$$P_V = \frac{Q\Lambda N_\mathrm{A}\rho}{M}\,, \tag{1}$$

where Q is the Q-value, ρ is the density, M is the molar mass, $\Lambda = (\ln 2)/T$ is the decay constant and N_A is Avogadro constant. The product ΛN_A has the meaning of the activity of 1 mole of material, that is, how many nuclei decay in 1 s in 1 mole of material.

The heat transfer is realized by

- conduction,
- convection, *radiation.*

Conduction is realized by collisions of atoms or oscillations of the crystal lattice[10] – that is the reason why it is typically more pronounced in solids, especially metals. It also exists in liquids; however, it is usually negligible due to the larger interatomic distances. For a general description of conduction, we have the heat conduction equation

$$a\,\nabla^2 T + \frac{P_V}{\rho c_p} = \frac{\mathrm{d}T}{\mathrm{d}t}\,, \tag{2}$$

where $a = \lambda/\rho c_p$ is the thermal diffusivity (λ is the thermal conductivity, c_p is the specific heat capacity), T is the temperature, ρ is the density of the material, and t is the time. The equation describes the heat balance of the system. The input variables are the material's properties and geometry, and the solution yields the temperature profile, i.e. its dependence $T(x, y, z, t)$.

Convection is accomplished by moving mass – i.e., atoms actually change their equilibrium position, often moving along complicated trajectories. That is the reason why we speak about convection in the context of fluids. According to the action of external forces, we distinguish forced convection, where the fluid flow is driven by an external force (e.g. work of a pump), or natural, which works by buoyancy forces generated due to the thermal expansion of the fluid– a hot fluid has less density, so it rises upwards due to buoyant forces.

In terms of the amount of heat transferred over a one-unit area, forced convection is more pronounced. Natural convection is, however, more significant on an absolute scale, as it is also realized in large systems (such as the Earth's crust or stars).

Radiative heat transfer is accomplished directly by the release of photons. Each object with a non-zero (absolute) temperature radiates. Radiance (the amount of energy transferred through radiation per 1 s over one-unit area) is given by equation

$$M_e = \sigma T^4\,, \tag{3}$$

where $\sigma = 5.67\cdot 10^{-8}\,\mathrm{W{\cdot}m^{-2}{\cdot}K^{-4}}$ is the Stefan–Boltzmann constant[11] The constant σ is small, but the term T^4 grows rather quickly, so it is dominant for high-temperature radiation. Since all objects radiate, we must subtract the energy from the radiated energy received by absorbing radiation from the surrounding environment.

$$M_{e,\mathrm{ef}} = M_{e,\mathrm{surf}} - M_{e,\mathrm{out}} = \sigma\left(T_\mathrm{surf}^4 - T_\mathrm{out}^4\right)\,.$$

[9] Not quite true – particles formed near the edge of the material will be emitted outward, escaping, and transferring their energy to the surrounding air. Since a certain width can confine this layer, its significance decreases with the thickness of the material.

[10] For this reason, we can say that thermal excitations propagate at the speed of sound because the mechanism is the same.

[11] The derivation is beyond the scope of this problem. For the curious participants, let us reveal the formula was obtained by integrating Planck's radiation law over all wavelengths and across the solid angle.

The equation above still needs correction for reflectivity – not whole radiation is absorbed – some of it gets reflected. The absorbance α expresses a proportion of the absorbed and reflected energy

$$M_{e,\mathrm{ef}} = M_{e,\mathrm{surf}} - \alpha M_{e,\mathrm{out}} = \sigma\left(T_{\mathrm{surf}}^4 - \alpha T_{\mathrm{out}}^4\right) .$$

Calculation

To melt the material, we need the temperature in the hottest place to be higher than its melting temperature. From the table[12] we can see the melting temperature $T_{\mathrm{melt}} = 1\,585\,\mathrm{K}$. First, let us consider what geometry will achieve the highest temperature in a given amount of material at the center. This is obtained by

- minimizing the area, because by reducing the area, we reduce the heat transfer, so the object will have to heat up more to establish equilibrium,
- maximizing the distance of "the innermost point" from the edge – because the temperature increases away from the surface.

Without proof, let us use the fact that the properties above are satisfied by a sphere. Since the sphere is solid, the heat transfer will be mainly by conduction – so let us compute the heat conduction equation (2) in sphere geometry. We want to solve for the stationary state (i.e., the state that occurs after a sufficiently long time when the system is in equilibrium and does not change over time). Therefore, the right-hand side of the equation is zero

$$a\nabla^2 T + \frac{P_V}{\rho c_p} = 0 . \tag{4}$$

For further simplification, we can divide the whole equation by a

$$\nabla^2 T + \frac{P_V}{\lambda} = 0 . \tag{5}$$

The Laplace operator in the sphere geometry (assuming the problem is dependent only on the radial coordinate[13]) is

$$\nabla_{\mathrm{sph}}^2 = \frac{\partial^2}{\partial r^2} + \frac{2}{r}\frac{\partial}{\partial r} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial}{\partial r}\right) .$$

By substituting (5) we get

$$\begin{aligned} \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial T}{\partial r}\right) + \frac{P_V}{\lambda} &= 0 ,\\ \frac{\partial}{\partial r}\left(r^2\frac{\partial T}{\partial r}\right) &= -\frac{P_V}{\lambda}r^2 ,\\ r^2\frac{\partial T}{\partial r} &= -\frac{P_V}{3\lambda}r^3 + C_1 . \end{aligned}$$

As a boundary condition, let us choose that there is a maximum in the core of the sphere, i.e.,

$$\frac{\partial T}{\partial r}(r=0) = 0 = C_1 .$$

[12] `https://www.webelements.com/gadolinium/thermochemistry.html`
[13] Which is not entirely true here, see discussion.

We will continue by integrating both sides

$$
\begin{aligned}
r^2 \frac{\partial T}{\partial r} &= -\frac{P_V}{3\lambda} r^3 \,, \\
\frac{\partial T}{\partial r} &= -\frac{P_V}{3\lambda} r \,, \\
T(r) &= -\frac{P_V}{6\lambda} r^2 + C_2 \,.
\end{aligned}
$$

Finally, we apply the boundary condition to the known temperature in the core $T(r=0) = T_{\mathrm{in}}$

$$T(r) = T_{\mathrm{in}} - \frac{P_V}{6\lambda} r^2 \,,$$

for the temperature at the edge is the following true

$$T(R) = T_{\mathrm{in}} - \frac{P_V}{6\lambda} R^2 \,. \tag{6}$$

We will determine the temperature at the edge from the condition that the entire heat output of the sphere must be dissipated. Calculating natural convection would be complicated, so we will start by assuming that all heat is transferred to the surrounding environment by the radiation and that the absorption is 1, i.e., everything that hits the sphere is absorbed. Then

$$P_V V = SM = S\sigma \left(T_{\mathrm{surf}}^4 - T_{\mathrm{out}}^4 \right) \,. \tag{7}$$

Substituting for the volume and the surface of the sphere as well as the temperature profile in the sphere we get

$$T_{\mathrm{in}} = \frac{P_V}{6\lambda} R^2 + \sqrt[4]{\frac{P_V}{3\sigma} R + T_{\mathrm{out}}^4} \,. \tag{8}$$

Explicitly expressing R so that $T_{\mathrm{in}} = T_{\mathrm{melt}}$ would be challenging, so we can help ourselves by plotting the graph of $T_{\mathrm{in}}(R)$ and finding where $T_{\mathrm{in}} = T_{\mathrm{melt}}$. Afterward, we will use $T_{1/2} = 74.6\,\mathrm{y}$ (from the Nuclear Data Search[14]) in order to obtain the decay constant Λ, then $Q = 3271.21\,\mathrm{keV}$ (the same source), $N_{\mathrm{A}} = 6.022 \cdot 10^{+23}\,\mathrm{mol^{-1}}$ (Avogadro constant), $\rho = 7900\,\mathrm{kg{\cdot}m^{-3}}$ (from Wikipedia[15]) and $M = 0.148\,\mathrm{kg{\cdot}mol^{-1}}$ (we could not find anything more precise). We will substitute to the formula (1) and get P_V. Furthermore, $T_{\mathrm{out}} = 293.15\,\mathrm{K}$ (from the problem assignment) and $\lambda = 10.6\,\mathrm{W{\cdot}m^{-1}K^{-1}}$ (also from Wikipedia) substituting to the formula (8) will give us dependence, which is shown in figure 1. Using a popular numerical technique or by brute force, we find

$$R = 7.01\,\mathrm{cm} \,,$$

which implies $m = 11.4\,\mathrm{kg}$.

[14] http://nucleardata.nuclear.lu.se/toi/nuclide.asp?iZA=640148
[15] https://en.wikipedia.org/wiki/Gadolinium

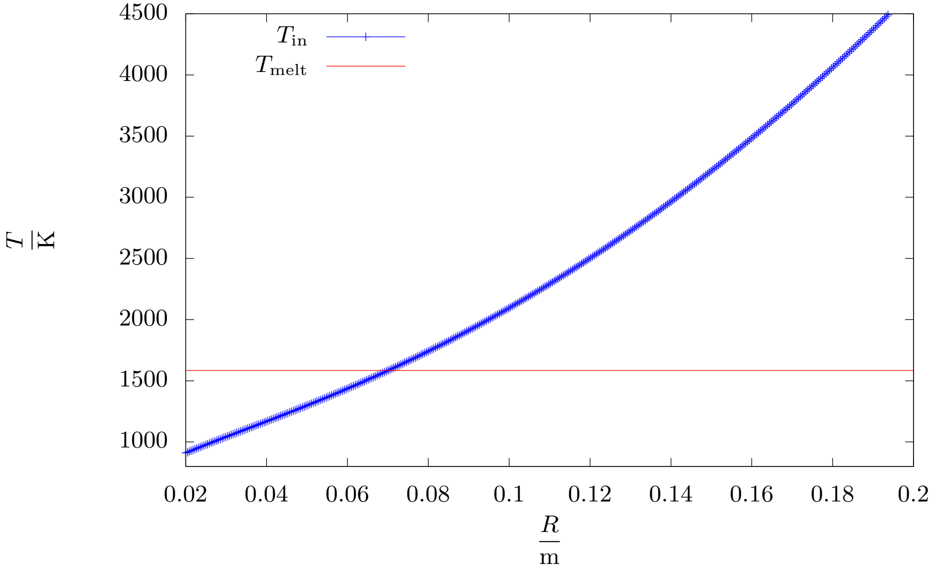

Fig. 1: Dependence of the temperature T_{in} at the core of the sphere on the radius, assuming that heat is transferred to the surroundings only by radiation.

Influence of natural convection

Let us evaluate the effect of natural convection – we have neglected it in the calculations above; however, we have not proven it in any way. In the balance equation (7) we will add a term for natural convection q_{conv}

$$P_V V = S\left(\sigma\left(T_{\mathrm{surf}}^4 - T_{\mathrm{out}}^4\right) + q_{\mathrm{conv}}\right)\,. \tag{9}$$

We need to determine the term q_{conv}. For heat transfer between two media, we generally use Newton's law

$$q_{\mathrm{conv}} = \alpha\left(T_{\mathrm{surf}} - T_{\infty}\right)\,,$$

where T_{∞} is "temperature far enough in the surrounding medium", since the fluid in the near vicinity of the hot sphere will have a higher temperature. We need to find the coefficient α – this is determined by the type and shape of the surface and type of the flow – which is affected by the wall temperature. All in all, we have a problem with too many unknowns. In general, α increases with the difference $T_{\mathrm{surf}} - T_{\infty}$, because higher wall temperature causes a greater difference in the density of the surrounding air, resulting in a more intense natural flow. The exact analytical calculation does not exist. We can compute the numerical solution of the Navier-Stokes equations with heat transfer, which the most modern CFD software are capable of,[16] or use similarity theory and empirical correlations. Let us show the estimation using similarity

[16] CFD is from *Computational Fluid Dynamics* – typically based on the finite volume method. Examples of codes used are ANSYS Fluent, Siemens StarCCM+ or OpenFOAM.

theory (let us forgive the theorists). To calculate the α we can use the Nusselt number defined as

$$Nu = \frac{\alpha L}{\lambda}\,,$$

where L is the characteristic dimension –in the case of a sphere, the diameter. We obtained the Nusselt number Nu from the correlation[17]

$$Nu = \left(2^{0,816} + 0,15 Ra_D^{0,277}\right)^{1/0,816}\,,$$

where Ra_D is the Rayleigh number, defined as

$$Ra_D = \frac{g\beta}{\nu a}\left(T_{\mathrm{surf}} - T_{\mathrm{out}}\right) D^3\,,$$

where g is the free-fall acceleration, β is the volumetric coefficient of thermal expansion, ν is the kinematic viscosity, and D is the characteristic dimension – the diameter. After all the substitutions, we will get

$$\sigma\left(T_{\mathrm{surf}}^4 - T_{\mathrm{out}}^4\right) + \frac{\lambda}{2R}\left(T_{\mathrm{surf}} - T_{\mathrm{out}}\right)\cdot$$
$$\cdot\left(2^{0,816} + 0,15\left(\frac{g\beta}{\nu a}\left(T_{\mathrm{surf}} - T_{\mathrm{out}}\right)\cdot 8R^3\right)^{0,277}\right)^{1/0,816} - \frac{1}{3}P_V R = 0\,, \tag{10}$$

here we should also substitute from equation (6) for the conversion T_{surf} for T_{in}, finally express T_{in} in dependence on R and find out where $T_{\mathrm{in}} > T_{\mathrm{melt}}$ – or the other way around, put $T_{\mathrm{in}} = T_{\mathrm{melt}}$ and solve for R. The equation is already very ugly; let us put $T_{\mathrm{in}} = T_{\mathrm{melt}}$, by using the computer, we will calculate the left side of the equation (10) for various R and find when it equals zero. Thus, we will get

$$R = 12.42\,\mathrm{cm}\,,$$

which corresponds to a mass of 63.34 kg. Let us keep in mind that the relationships are approximate, and we should always be concerned with their conditions of validity.

In summary, we have shown that the original assumption about dominant radiation was wrong. We can calculate, out of curiosity, that the surface temperature of the sphere is 374 K, which is not much – the difference between the surface layer of the sphere and the inside is over 1 200 K, which is due to a combination of the volumetric heat source and poor thermal conductivity (as an example, steels have approximately 8 – 60 $\mathrm{W{\cdot}m^{-1}{\cdot}K^{-1}}$, aluminum has over 200 $\mathrm{W{\cdot}m^{-1}{\cdot}K^{-1}}$; in the contrast, insulating materials such as polystyrene 0.3 $\mathrm{W{\cdot}m^{-1}{\cdot}K^{-1}}$).

Finally, let us comment on the assumption from the problem assignment that no fission occurs. We cannot say that with absolute certainty – it is just that we do not have enough data for fission, because the isotope ^{148}Gd is neither commonly found in nature nor commonly used (Gd is often used as a burnup absorber in nuclear reactors, but the ^{155}Gd and ^{157}Gd are mainly applied there).

The rarity of this isotope also makes the task unrealistic– it is hardly imaginable that an α emitter could be found anywhere in such large quantities. If anything, it would be found in very small amounts. The problem was inspired by radioisotope thermoelectric generators, where α emitters are actually used to generate heat– e.g., the isotope ^{238}Pu in the MMRTG[18] on the Curiosity rover.

[17] `https://thermopedia.com/content/786/`

[18] `https://en.wikipedia.org/wiki/Multi-mission_radioisotope_thermoelectric_generator`

Addendum – nuclear data

We took data from "various" sometimes even too open sources. For the FYKOS problem assignment, this is not a problem, but in academic papers, the above sources might not be considered "sufficiently credible". Furthermore, the more complicated calculations would require even more data – different half-lives, the types, and energy of the decay products; in the case of neutronic calculations, we would need effective cross-sections. For the reasons above, it is necessary to have all the data from one source and ideally verified by someone.

That is why ENDFs (*Evaluated Nuclear Data File*) exist. For individual isotopes, there are ENDF files containing different data – effective cross-sections, the decay products, and the nuclear parameters,... there are entire libraries of ENDF files such as the European JEFF library or the US ENDF/B library. The libraries are typically considered a trusted source.[19]

Let us use this problem to show how to search the nuclear data. One of the available tools is JANIS.[20] On the main page, select the button "Browse", then "Radioactive data". Choose a library – e.g., JEFF-3.3. Then "Radioactive decay data", "Gd", "^{148}Gd". It will open up a blank visualizer window, and at the bottom of the table, click on the list of "Decay data" and in the row "Discrete Alpha" select T. The new tab should open in the visualizer window with a table with a single line 3 182 800 eV. We used a little different value. We could use a similar procedure for other data for a particular isotope and its decay and reactions.

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[19] They still vary in the amount of data and generally provide slightly different results. Academics then have their favorites, so "suitability" of the data used for the calculation can be the subject of long debates.

[20] JANIS is only for viewing and visualizing nuclear data; it is not a library. It is available at https://oecd-nea.org/janisweb/