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Supplementary_Movie_2_Time_lapse_salt_flower_macroscopic_24hours.mp4

Supplementary_Movie_3_Time_lapse_salt_flower_microscopic_2days.mp4

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INTERFACE

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Research



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Onggi's permeability to carbon dioxide accelerates kimchi fermentation

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Since ancient times, Korean chefs have fermented foods in an onggi, a traditional earthenware vessel. The porous structure of the onggi mimics the loose soil where lactic acid bacteria is naturally found. This permeability has been purported to facilitate the growth of lactic acid bacteria, but the details of the process remain poorly understood. In this combined experimental and theoretical study, we ferment salted Napa cabbage in onggi and hermetic glassware and measure the time course of carbon dioxide concentration, which is a signature of fermentation. We present a mathematical model for carbon dioxide generation rate during fermentation using the onggi's gas permeability as a free parameter. Our model provides a good fit for the data, and we conclude that porous walls help the onggi to 'exhale' carbon dioxide, lowering internal levels to those favoured by lactic acid bacteria. The positive pressure inside the onggi and the constant outflow through its walls act as a safety valve for bacteria growth by blocking the entry of external contaminants without mechanical components. We hope this study draws attention to the work of traditional artisans and inspires energy-efficient methods for fermenting and storing food products.

1. Introduction

According to ancient texts, Koreans have enjoyed fermented foods since the Goguryeo Kingdom (37 BC–AD 668) [1]. Before the advent of refrigeration, Korean chefs used handmade clay vessels called onggi to ferment their foods. The onggi was designed without modern knowledge of chemistry, microbiology or fluid mechanics. Onggi-shaped vessels are depicted in numerous historical records, such as the tomb mural in figure 1*a* from Anak Tomb No. 3, dating back to AD 357 [2]. Many recent studies purport that fermentation in onggi can help preserve and increase the nutrition of fermented foods [3,4]. While modern mass fermentation is performed in controlled conditions such as metal vessels, there remain a number of artisans who continue to make onggi by hand. The goal of this study is to present a mechanical explanation for the ability of onggi to aid in fermentation. We hope that this study will increase the visibility of onggi makers and inspire energy-efficient ways to preserve and ferment foods.

Onggi have traditionally been used to make a variety of fermented food such as kimchi (fermented spicy cabbage, figure 1*b*), ganjang (soya sauce), gochujang (red pepper paste) and doenjang (soya bean paste), all of which have become essential parts of Korean cuisine. The addition of salt to these vegetable products, which already contain natural bacteria, prevents spoilage while promoting the proliferation of salt-loving lactic acid bacteria [5,6]. In fact, the signature sour taste of these foods is due to the lactic acid produced by probiotic bacteria including the rod-shaped *Lactobacillus* species (10 μ m in length) and oval-shaped *Leuconostoc* species (1 μ m in length) [7]. Research on onggi has primarily focused on accelerating the growth of lactic acid bacteria while suppressing the growth of other bacterial strains. Kimchi fermented in onggi over four weeks had 100 times higher lactic acid bacteria counts than kimchi

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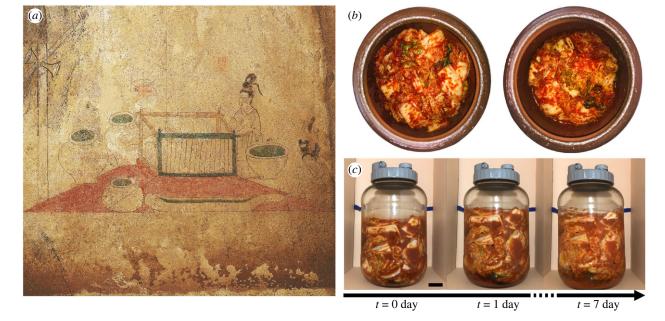


Figure 1. (*a*) Vessels resembling onggi on a mural at Anak Tomb No. 3 from Goguryeo Kingdom (37 BC–AD 668). Image courtesy of the Northeast Asian History Foundation. (*b*) Top view of kimchi inside an onggi before (left) and after (right) fermentation. (*c*) Side view of kimchi fermented in a glass jar for 7 days at 30°C. Scale bar 30 mm. The process of interest is the lactic acid fermentation driven by two species of anaerobic bacteria, *Lactobacillus* and *Leuconostoc*. A dramatic volume increase and the formation of carbon dioxide bubbles were observed within 1 day.

fermented in plastic and steel containers [3]. Moreover, onggi fermentation slowed the growth of foul-tasting aerobic bacteria by a factor of 100. Onggi also increases the acidity and antioxidant activity of kimchi [8,9].

While the improved lactic acid bacteria growth in onggi is exciting, a missing link in this field is a connection between the material properties of the onggi and the growth of bacteria. In this study, we apply the methods of fluid mechanics to determine this connection. We hypothesize that onggi influences fermentation by the transmission of gas through its porous walls. Onggi has numerous gaspermeable micro-pores ranging from 1 to 100 µm in size [10,11]. These pores increase the transmission of oxygen and carbon dioxide with respect to most modern food containers made from polyethylene and glass [3,12]. Many researchers assume that the gas permeability of onggi causes the high bacterial count in fermented food like kimchi, but a clear mechanism has yet to be given. Previous studies only measured the carbon dioxide of an initially 109 gas-filled vessel without living bacteria, or only considered 110 the growth of bacteria without measuring gas generation 111 [3]. In our work, we examined the interaction of fermenting 112 cabbage, the carbon dioxide it generates, and the 113 permeability of its walls. We linked these three subjects 114 using a mathematical model that accounts for the onggi's 115 permeability to carbon dioxide.

116 Critical to the onggi's function is its permeability, the 117 capacity of a porous material to allow fluids to flow through 118 it. By virtue of its liquid permeability k, onggi generates 'salt 119 flowers', salt crystals that appear on its outside when fer-120 menting salty foods. As we will show, the onggi's gas 121 permeability k_{α} enables it to transmit the carbon dioxide gen-122 erated by fermentation. In general, permeability depends on 123 the shape and size of interconnected pores in the material. 124 It is an intrinsic property of porous materials and has tra-125 ditionally been used to show how water travels through 126 filters, gravel and aquifers. Permeability is defined in units of area and refers to the area of open space in a cross-section that is perpendicular to the direction of flowing fluid [13,14].

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2. Material and methods

2.1. Onggi construction and characterization

In the summer of 2021, we purchased a large onggi from Jeju Onggi Village on Jeju Island, Korea. The onggi had a volume of 4600 ml with a mouth radius of 10 cm and a height of 20 cm. One constraint was that the vessel had to be tall and wide enough to fit our airborne carbon dioxide sensors without touching the cabbage. The onggi was manufactured by a traditional process which we describe here [11]. Raw mud containing water, silt and clay was pressed and slapped by hand to homogenize it. Pebbles were picked out and the material was first formed into long clay rods. Then, the pottery was shaped on a spinning wheel and left for about a day under ambient conditions to dry out. Finally, onggi slowly sintered inside a kiln at around 1200°C for a day and slowly cools down. While some onggi are glazed, ours was not.

We characterized the pore structure with both a scanning electron microscope (Hitachi SU-8010 SEM) and computed tomography (Scanco CT-50). The sample for SEM imaging was prepared using a rotary saw with a diamond wafering blade (PELCO Precision Low-Speed Saw) to obtain a smooth surface. The sample for the CT scan was prepared by breaking it with a hammer. A small piece was mounted on a 6 mm diameter sample holder and imaged under high-resolution settings (55 kV of energy, 145 μ A of intensity, 1 s of sample time and 2 μ m voxel size). A threshold masking was set to lower and upper limits of 5183 HU (Hounsfield units) and 10 000 HU, respectively. The porosity was computed by sampling multiple cylinders (diameter 600 μ m and length 1000 μ m) within the boundary of the outer surface.

2.2. Kimchi fermentation time-lapse

For the time-lapse video of fermentation (figure 1*c*; electronic supplementary material, movie S1), we relied on a traditional



Figure 2. (*a*) Time-lapse of salt flower formation on the outer surface of an onggi for 24 h. Scale bar 50 mm. (*b*) Onggi surface covered with salt crystals after 3 days. Scale bar 50 mm. (*c*) The microscopic time-lapse of salt flower at the outer onggi surface for 8 h. Scale bar 500 μ m. Salt crystals form as the evaporation dries out the water contents and fluid is imbibed to the surface.

kimchi recipe. Throughout the study, Napa cabbage (Brassica rapa 154 subsp. pekinensis) was purchased from local grocery stores, either 155 on Jeju Island or in Atlanta. We used 1 kg of the crispy layers of 156 cabbage for the time-lapse. Each leaf of cabbage was cut into two 157 or three pieces perpendicular to the midrib so that each piece 158 consisted of both the wrinkled lamina and the surrounding 159 veins. The Napa cabbage was immersed in 2 wt% salty water 160 for 6 h and then mixed thoroughly with traditional kimchi sea-161 soning, which for each kg of cabbage consisted of $160\,\mathrm{g}$ of 162 powdered red pepper, 200 g of minced onion, 20 g of anchovy 163 fish sauce, 15 g of minced garlic and 5 g of minced ginger.

Kimchi fermentation could not be easily observed in onggi because of the dark conditions inside the vessel. Thus, we conducted the time-lapse video of kimchi fermentation in a hermetically sealed glass jar. The fermentation process was filmed by a USB camera (Logitech Brio 4K Webcam) for seven days at 3°C, illuminated by a single LED source.

2.3. Salt flower formation

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Two time-lapse videos of salt crystal formation were performed: one in a macroscopic view and another in a microscopic view. In the macroscopic view (figure 2*a*,*b*; electronic supplementary material, movie S2), the onggi was filled with 21 of 15 wt% salty water and observed for 24 h using a USB camera (Logitech Brio 4K Webcam).

For the microscopic film (figure 2*c*; electronic supplementary material, movies S3,4), a rectangular onggi piece was observed for 2 days while the bottom of the sample was immersed in 15 wt% salty water. This per cent saltwater is comparable to the salt concentration in soy sauce (approx. 18%) which may be fermented in onggi [15]. We used a stereo optical microscope (Leica DVM6 Digital Microscope) in the multi-focus setting.

2.4. Water evaporation measurement

To measure the onggi permeability to water, we measured how
 quickly water evaporated from its walls. The onggi was initially
 filled with 2000 g of water and the top was sealed with plastic

stretch wrap and a rubber ring. This set-up ensured that evaporation mostly occurred through the porous walls of the onggi. The time course of the mass was measured by a portable balance (Mettler Toledo PL-E).

2.5. Carbon dioxide measurement

For the carbon dioxide measurements, we applied only saltwater to the cabbage because the kimchi recipe includes ingredients for flavour but salt is the necessary component to stimulate fermentation. We measured the carbon dioxide concentration and estimated the gas production in two containers, a hermetically sealed glass jar and closed-top onggi (figure 4a). For the onggi, we designed and built a custom-fit lid for three types of gas sensors: a pressure sensor, a carbon dioxide sensor, and an oxygen sensor (Vernier Software & Technology). These sensors fit vertically into the onggi mouth using a custom-designed three-dimensional-printed lid (Formlabs Form 3). All purpose sealing film (Bemis, Parafilm) filled any remaining gaps between the devices and the lid. Despite our efforts, our 'hermetically sealed' glass jar leaked, but less so than the permeable onggi, as shown by the factor of two differences in their gas permeability k_g presented in the results section.

We performed three fermentation trials of salted cabbage in a glass jar and an onggi. The glass jar was less than half the volume of the onggi: the glass jar had a volume of 1900 ml, a height of 15 cm and a radius of 10 cm. In each trial, 200 g of cabbage was observed for 2 days. To prepare the salted cabbage for observation, it was first immersed in brine with 2 wt% of salt concentration for 6 h. For all trials, cabbage was drained of water before being placed in the containers for measurement. Unlike traditional kimchi fermentation, there were no extra seasonings in the jars, so the salted cabbage sat without any surrounding liquid, as shown in figure 4*a*.

The onggi was placed in a laboratory oven (Quincy Lab 30GC Convection Lab Oven), controlled by a voltage regulator (Variac TDGC-0.5KM) to maintain darkness and a temperature of 25°C. While the typical temperature for kimchi fermentation is about 10°C [16], we used a higher temperature to expedite

fermentation. Since the containers' pores harbour bacteria [11,17], the containers were sterilized in an autoclave (Steris Amsco Century SV-136H Prevac Steam Sterilizer) before each trial.

3. Mathematical models

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We present mathematical models on the permeability of the onggi to liquids and gases. The former was used to estimate the liquid permeability k and the latter to infer the carbon dioxide generation rate of the lactic acid bacteria. At the end of the section, we enumerate the values of the dimensionless groups used in these models. The dimensionless groups justify the modelling choices made.

3.1. Liquid permeability model

Our model predicts the liquid permeability k of the onggi wall based on the evaporation rate of water from the onggi. This model follows our experiment in which a capped water-filled onggi loses mass at a steady rate. Based on our observations of salt formation on the outside of the onggi (figure 2), we assume this mass loss is due to water that has imbibed through the porous walls and evaporated from the outside.

Based on the flow speeds of water presented in the dimensional analysis section, the flow through the onggi is characterized by incompressibility and a low Reynolds number. We apply Darcy's Law, which is valid for creeping laminar flow at low Reynold's numbers [18,19]. Darcy's Law states that the volumetric flow rate per unit area *q* is proportional to the gradient of pressure, $q \sim \nabla P = \Delta P/d$, where *d* is the onggi wall thickness. In all,

$$q = -\frac{k}{\mu} \frac{\Delta P}{d},\tag{3.1}$$

where the constant of proportionality involves the liquid permeability k of onggi, and the water's dynamic viscosity μ . The outside wall of the onggi is at atmospheric pressure. The two driving pressures of the fluid are the water's hydrostatic pressure and the capillary pressure which tends to imbibe fluid into the hydrophilic wall. The sum of these two pressures may be written

$$|\Delta P| \sim \rho g z + \frac{4\sigma \cos\theta}{d_p},\tag{3.2}$$

where ρ is the density of water, *g* is gravitational acceleration, *z* is the height from the water surface, σ is the surface tension of water, θ is the contact angle of water on the onggi and d_p is the pore diameter which we show using microscopy to be on the order of 10 µm. The first term on the right-hand side is the hydrostatic pressure, and the second term is the capillary pressure, or Laplace pressure. The ratio of the hydrostatic term to the capillary term is related to the modified Bond number, which we calculate to be less than 1, indicating the dominance of capillary pressure. We proceed by considering only capillary pressure.

We assume a cylindrical container with an effective radius R, which is the average of the onggi mouth radius, maximum radius and bottom radius. The evaporation rate, or the total flow rate integrated across the height of the liquid z_r may be written (figure 3c)

$$Q_e \sim 2\pi R \int_0^{z_r} q(z) \,\mathrm{d}z,\tag{3.3}$$

where q can be substituted using equations (3.1) and (3.2). Simplifying equation (3.3), we write the liquid permeability k as

$$k \sim \frac{\mu d}{2\pi R} \frac{Q_e}{(1/2)\rho g z_r^2 + (4\sigma \cos\theta/d_p) z_r)} \sim \frac{\mu dQ_e d_p}{8\pi R z_r \sigma \cos\theta}.$$
 (3.4)

We may solve equation (3.4) by measuring Q_e from the rate of mass loss of the onggi. The rate of mass loss was converted to a volumetric flow rate using water density. If the evaporation rate is measured over a sufficiently small time window, the water height z_r can be considered constant. The model presented applies to liquids passing through the onggi walls. In the next section, we consider the passage of gases.

3.2. Gas permeability model

The goal of this model is to infer the carbon dioxide generation rate \dot{n}_g (mmol h⁻¹) by the bacteria. This model is necessary because the onggi is permeable to carbon dioxide. Figure 4b depicts the variables used in this model. The primary input to our model is the concentration of carbon dioxide in the vessel C_{CO_2} as measured by our sensor. In order to determine the carbon dioxide generation rate \dot{n}_g (mmol h⁻¹), we converted C_{CO_2} to the partial pressure of carbon dioxide in the vessel p_{in} . Note that we used p to denote partial pressure and P to denote the total pressure, which according to Dalton's Law, is the sum of all the partial pressures of the gases (p_{CO_2} , p_{N_2} , p_{O_2} ...). With this notation, p_{in} and p_{out} are the partial pressures of carbon dioxide inside and outside the vessel. Similarly, P_{in} and P₀ denote the total pressures inside and outside the vessel. The total pressure in the vessel P_{in} was measured using our pressure sensor. Pressure P_0 and temperature T_0 outside the vessel were given by ambient conditions, which in this case corresponded to a temperature-controlled oven at atmospheric pressure.

Conservation of mass states that the rate of change of carbon dioxide in the vessel $\dot{n}_{total} \pmod{h^{-1}}$ is the sum of the rates of carbon dioxide outflow \dot{n}_{out} and carbon dioxide generation \dot{n}_g . Note that \dot{n}_{out} is negative, corresponding to carbon dioxide leaving the vessel.

$$\dot{n}_{\text{total}} = \dot{n}_{\text{out}} + \dot{n}_g. \tag{3.5}$$

The rate of outflow is dictated by the gas permeance equation, which is equivalent to Darcy's Law and states that the molar flow rate is proportional to the pressure gradient. In a porous vessel (figure 5b), the gas permeance equation may be written [3]

$$\dot{n}_{\rm out} = \bar{K} \frac{A}{d} (p_{\rm out} - p_{\rm in}), \tag{3.6}$$

where \bar{K} is the gas permeance, A is the vessel surface area, d is the vessel wall thickness and p_{out} and p_{in} are the partial pressures of carbon dioxide outside and inside the vessel, respectively. The partial pressure of carbon dioxide outside the vessel is dictated by atmospheric conditions and is given in table S1 in the electronic supplementary material. Carbon dioxide flows out of the vessel if the outflow of carbon dioxide flows out of the vessel if the outflow of carbon dioxide \dot{n}_{out} is negative. This is the case for an onggi fermenting kimchi where the pressure building up inside the vessel exceeds the pressure outside: p_{in} is higher than p_{out} .

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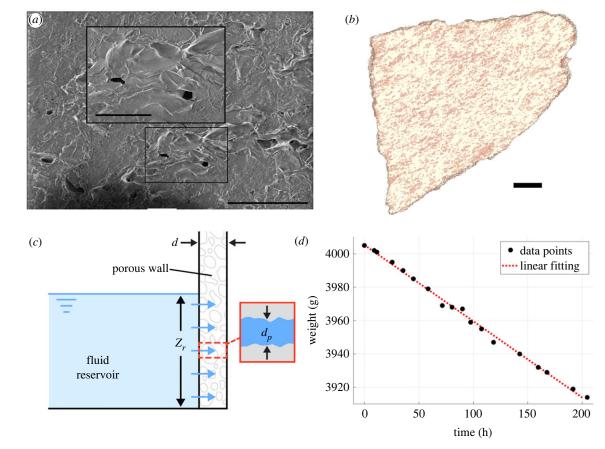


Figure 3. (*a*) Scanning electron microscope view of micro-pores in onggi. Scale bar, 50 μ m. Inset scale bar, 20 μ m. Pore size varies from 1 to 100 μ m diameter with an average of 5 μ m. Additional SEM images can be found in electronic supplementary material, figure S1. (*b*) Three-dimensional model of a 40 μ m-thick onggi slice using computed tomography (CT) scan. Scale bar, 200 μ m. Pores are pink and onggi clay is transparent. (*c*) Schematic of water flow through the porous wall. (*d*) Time course of the weight of the onggi due to evaporation at the onggi surface.

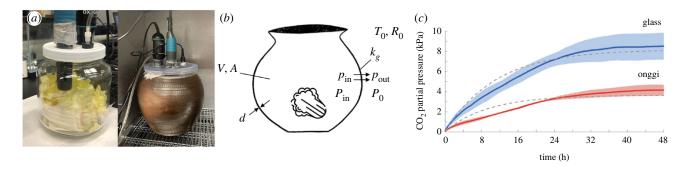


Figure 4. (*a*) Headspace gas set-up for measuring carbon dioxide, oxygen and pressure in a hermetic glass container (left, 1900 ml of volume) and onggi (right, 4600 ml of volume). (*b*) Schematic of carbon dioxide generation. Cabbage and jar icons from Flaticon and Clipart Library, respectively. (*c*) Time course of the carbon dioxide partial pressure for 200 g of salted cabbage stored in glass (in blue) and onggi (in red). Solid lines are from experiments and the dashed lines are from the theoretical model. Raw data are given in electronic supplementary material, figure S2.

The gas permeance of the vessel is defined in terms of the gas permeability and conditions outside the vessel

$$\bar{K} = \frac{P_0}{R_0 T_0} \frac{k_g}{\mu_{\rm CO_2}},\tag{3.7}$$

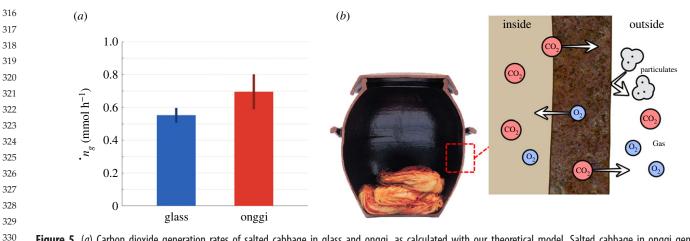
where k_g is the gas permeability of the vessel, P_0 is the ambient pressure, T_0 is ambient temperature, R_0 is the gas constant and μ_{CO_2} is the dynamic viscosity of carbon dioxide at the prescribed pressure and temperature. Note that equation (3.7) is arbitrary in that it neglects the conditions inside the vessel. However, this assumption does not affect our final solution since we will be using k_g as a free parameter. Before we proceed, we provide a brief cautionary note on the use of gas permeance \bar{K} . In the kimchi fermentation literature, authors may inadvertently interchange the terms permeability, permeance and permeation [3,11,20]. In other fields, confusion between these terms is also common [14]; in fact, some researchers define gas permeance as \bar{K}/d , where *d* is the wall thickness. In our study, gas permeance is defined in terms of molecules of gas moving unit length that pass a cross-sectional area per hour at a given pressure on the outside of the wall. Thus, permeance has a unit of mol $\cdot kPa^{-1} \cdot m^{-1} \cdot h^{-1}$, but it is dependent on the intrinsic gas permeability of the material, which has a unit of m² [21–23]. We set the gas permeability k_g as a free parameter 

Figure 5. (a) Carbon dioxide generation rates of salted cabbage in glass and onggi, as calculated with our theoretical model. Salted cabbage in onggi generated 331 25% more carbon dioxide than in glass. (b) Schematic of selective permeability of onggi. The porous onggi wall allows carbon dioxide to exit and oxygen to enter. It 332 also blocks the passage of unwanted particulates larger than the pore size of 5 µm. Onggi cross-section image from Korean Ministry of Culture, Sports and Tourism. 333

then proceed by presenting the relationships between gas permeability and partial pressures of carbon dioxide.

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The partial pressure of carbon dioxide p_{in} inside the vessel is given by the ideal gas law which states that p_{in} is proportional to n_{total} the number of moles of carbon dioxide inside the vessel:

$$p_{\rm in}V = n_{\rm total}R_0T_0, \qquad (3.8)$$

where V is the vessel volume, T_0 is ambient temperature, and 343 R_0 is the ideal gas constant. Note, here we assume that the 344 temperature inside the vessel is the same as that outside, 345 which is supported by our temperature measurements. The 346 partial pressure of carbon dioxide (in kPa) is simply 347 the product of carbon dioxide concentration C_{CO_2} (in ppm), 348 the total pressure (in kPa), and a constant to convert ppm 349 to a proportion: 350

$$p_{\rm in} = C_{\rm CO_2} \cdot (1 \times 10^{-6}) \cdot P_{\rm in}. \tag{3.9}$$

353 Further constants are given in table S1 in the electronic supplementary material.

By assuming p_{out} and \dot{n}_g are constant, we may rewrite equation (3.8) using equations (3.5)-(3.7) in terms of the variable p_{in}

$$\frac{dp_{\rm in}}{dt} = k_g \frac{P_0 A}{\mu_{\rm CO_2} V d} (p_{\rm out} - p_{\rm in}) + \frac{R_o T_0}{V} \dot{n}_g$$
(3.10)

The initial condition of p_{in} is the partial pressure of carbon dioxide in the ambient. This differential equation may be solved to find the time course of the partial pressure of carbon dioxide inside the vessel

$$p_{\rm in} = p_{\rm out} + \beta \left(1 - \exp\left(-\frac{t}{\tau}\right) \right), \tag{3.11}$$

where the time constant τ is

$$r = \frac{\mu_{\rm CO_2} V d}{k_8 P_0 A},$$
 (3.12)

and the plateau value of p_{in} is influenced by the constant

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$$\beta = \frac{\dot{n}_g \mu_{\rm CO_2} R_0 T_0 d}{k_g P_0 A}.$$
(3.13)

377 Thus, the carbon dioxide generation rate \dot{n}_g influences only 378 the steady-state plateau without influencing the time constant. Equation (3.11) is the primary tool that we will use to measure the carbon dioxide generation rate from the time course of carbon dioxide data in the onggi headspace.

We collected six total experimental curves for two separate conditions: salted cabbage in each of the onggi and glass containers. Figure 4c shows the average of three trials per condition. The fitting was conducted in MATLAB and OriginPro based on the least-square method. There exist two free parameters: k_g and \dot{n}_g . Optimal k_g values for each of the glass and onggi containers were obtained using a MATLAB code that minimizes the mean of the R^2 values for the datasets. Then, OriginPro produced the associated \dot{n}_g values with uncertainties.

3.3. Dimensionless groups

Our mathematical modelling relied on dimensionless groups, which we calculate using our experimental results below. These dimensionless groups justify our choices in mathematical modelling. A small Reynolds number supports Darcy's law, and a small Bond number reinforces the capillary force dominance, simplifying the calculations for liquid permeability in equation (3.4).

We begin with characteristic velocities and length scales that are relevant for liquid and airflow through the pores of onggi. Using microscopy, we found the pores in the onggi wall had a characteristic diameter of $d_p = 10 \,\mu\text{m}$. The characteristic velocity U_w for water flow through onggi was obtained by dividing the volumetric flow rate per unit area by the porosity: using equation (3.1), $U_w = q_w/\epsilon \sim 50 \,\mathrm{m \, s^{-1}}$, where we converted porosity converted from percentage to proportion.

To calculate the characteristic velocity of carbon dioxide, we use a combination of equation (3.6), and the time derivative of the ideal gas law applied to the outgoing carbon dioxide \dot{n}_{out} , which is calculated from the experiment

$$P_0 \dot{v} = \dot{n}_{\rm out} R_0 T_0, \tag{3.14}$$

where \dot{v} is the volumetric flow rate of carbon dioxide. This equation, along with the surface area A of the onggi yields the volumetric carbon dioxide flow rate per unit area $q_{\rm gas} = \dot{v}/A$. Onggi's characteristic permeability to carbon dioxide, as measured in experiments was $k_g = 6.12 \times 10^{-15} \text{ m}^2$.

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379 Consequently, using equation (3.1) again, the characteristic 380 velocity of the carbon dioxide was $U_{\text{gas}} = q_{\text{gas}}/\epsilon \sim 1.9 \,\text{m}\,\text{s}^{-1}$. 381

The Reynolds number is defined as

$$\operatorname{Re} = \frac{\mathcal{U}d_p}{\nu},\tag{3.15}$$

where *U* is the velocity of the fluid (either U_w or U_{gas}) and *v* is 385 the kinematic viscosity of the fluid. The Reynolds numbers 386 for the water and carbon dioxide flow are 10.4×10^{-4} and 5.0×10^{-6} , respectively. Both liquid and gas flow are in the creeping regime, which indicates fluid flow is laminar and inertia-free. 390

The driving force of water flow through the onggi walls is capillarity. Capillary pressure in a pore may be estimated as $P_c = 4\sigma \cos\theta / d_p \sim 24.9$ kPa, which is much 393 higher than the measured steady-state gas pressure for 394 fermenting salted cabbage in the onggi, 4.1 kPa. The modi-395 fied Bond number may be defined as the ratio of the hydrostatic pressure $\rho g z_r$ and the capillary pressure at the pores σ/d_p . Note that there are two length scales at play here, which lead to

$$Bo = \frac{\rho g z_r}{\sigma/d_p}.$$
 (3.16)

The Bo is calculated to be 0.14, which implies that the water imbibition at the onggi pores is dominated by capillarity.

4. Results

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409 We used several cabbage preparations in this study. The first 410 was 'kimchi cabbage' which involves Napa cabbage (Brassica 411 rapa subsp. pekinensis) and traditional kimchi seasonings. The 412 fermentation of kimchi cabbage was captured by time-lapse 413 video. The second preparation was 'salted cabbage' which 414 only involves cabbage, salt and no other seasonings. The 415 salted cabbage was used in fermentation experiments in 416 which carbon dioxide is measured. The salted preparation 417 was performed over six times. For convenience, we provided 418 the minimal ingredients to trigger fermentation.

419 We filmed kimchi fermentation for 7 days (figure 1c; elec-420 tronic supplementary material, movie S1). After 24 h of 421 fermentation, the solution increased by 500 ml, which was 422 approximately 10% of the initial liquid, and comparable to 423 half of the volume of the original cabbage (which weighed 424 1 kg). Throughout the next few days, the cabbage generated 425 bubbles, presumably carbon dioxide due to the metabolism 426 of lactic acid bacteria which appears naturally on the cabbage. 427 The changing carbon dioxide in the vessel may in turn influ-428 ence the growth of the bacteria. We proceed by characterizing 429 the porosity and liquid permeability of the onggi wall, before 430 turning to its gas permeability.

431 Onggi releases gas through its permeable wall, which we 432 characterized using SEM microscope and CT scan. Figure 3a 433 shows the SEM image of a cross-section of the onggi wall. 434 Pore diameters ranged from 1 to 100 µm, which is compar-435 able to the range of $20 - 150 \,\mu\text{m}$ (accounting for 93.04% of 436 the pores) reported by Kim et al. [24]. Onggi pore formation 437 is driven by variables in the manufacturing process such as 438 the composition of ingredients, sintering temperature, and 439 sintering time. In comparison, porcelain's ingredients and 440 processing cause its uniformly fine particles to be imperme-441 able to gas [24].

We used a CT scanner to measure the porosity of the sample. Since the CT scan resolution was 2 µm voxel size, it could not resolve pores smaller than this size. Threshold masking produced a solid three-dimensional model of an onggi piece (figure 3b; electronic supplementary material, movie S5). We found a porosity of $\epsilon = 4.72 \pm 0.16\%$, which is comparable to $7.21 \pm 0.30\%$, measured using mercury por-osimetry [11], and four times smaller than $18.71 \pm 0.08\%$, the average value reported in Seo *et al.* [20]. We note that these other workers used indirect measurements of porosity through chemical absorption, while ours is a visualization of the porous structure using X-rays. We next performed water evaporation experiments to measure onggi's permeability to liquids. When Korean chefs ferment ganjang (soya sauce) in onggi, salt crystals pre-cipitate on the outside of the container, a phenomenon known as 'salt flower'. We demonstrated this process by film-ing an onggi halfway full of salty water (figure 2a,b; electronic supplementary material, movie S2). Within 8 h, a white layer of salt crystals appears on the outside, intensifying over time. Salt is initially limited to the water line but climbs above it after 24 h. Chefs (Onggi Village pers. comm.) believe the **Q2** sample. Since the CT scan resolution was 2 µm voxel size, it

appearance of the salt flower indicates a good quality fermentation vessel.

To better visualize the salt flower formation, we performed time-lapse videography of a small piece of onggi atop salt water using a microscope. Figure 2c and electronic supplementary material, movies S3,4 show a dry outer wall covered by a thin film of water and then by salt crystals. Based on these events, we surmise that salt water flows through the wall and evaporates at the outer surface, leaving salt crystals behind. Evaporation of water at the outer surface is replenished by water wicked through the wall.

Now that we see onggi is permeable to water, we measure the flow rate. We filled the onggi with 2000 g of water and sealed the top. The water flowed through the walls and evaporated at a constant rate of $Q_e = 0.75 \pm 0.18 \text{ g h}^{-1}$ ($R^2 = 0.989$) as shown in figure 3d. Using equation (3.4), we estimate the liquid permeability of the onggi sample to be $k = (4.58 \pm$ 1.10) × 10⁻¹⁵ m². Converting liquid permeability to hydraulic conductivity $K = k\rho g / \mu$ yields $K = (4.58 \pm 1.10) \times 10^{-8} \text{ m s}^{-1}$, where ρ is the density of water, *g* is the acceleration of gravity and μ is the dynamic viscosity of water [25]. Table 1 compares the liquid permeability of onggi with other well-characterized geological materials that are not tested here, but simply included for comparison. Onggi is comparable to silt and loess (a layer of windblown dust and silt covering 10 per cent of the earth). This makes sense since onggi is made mainly from natural mud, which has silt as a large component. We classify onggi as semi-permeable because it lies between permeable materials such as clean sand and gravel $(k \sim 10^{-7} - 10^{-10} \text{ m}^2)$ and impermeable materials such as granite $(k \sim 10^{-17} - 10^{-21} \text{ m}^2)$.

We may also compare the measured liquid permeability to predictions from theoretical models [27-29]

Kozeny – Carman model:
$$k = \frac{d_{par}^2}{180} \cdot \frac{\epsilon^3}{(1-\epsilon)^2}$$
,
Ergun model: $k = \frac{1}{66.7} \cdot \epsilon d_{par}^2$
and Happel – Brenner model: $k = \frac{2}{9}d_{ch}^2 \cdot \frac{2-(9/5)(1-\epsilon)^{1/3}-\epsilon-(1/5)(1-\epsilon)^2}{1-\epsilon}$

$$(4.1)$$

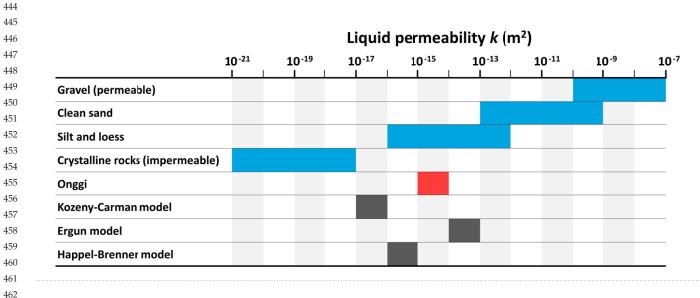
where ϵ is the porosity, d_{par} is the particle diameter, d_{ch} is the channel diameter and other coefficients are empirically

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Table 1. Onggi permeability compared to other geological materials and predictions from mathematical models. Onggi permeability is in the middle of the conventional range of permeable and impermeable materials. Data on rocks and sediments from [25,26].



465 found. Table 1 also shows the calculated permeability based 466 on these models, assuming d_{par} and d_{ch} equal to the pore size of 10 μ m, and ϵ equals 4.72 %. These models all give 467 values within 3 orders of magnitude of our measurement, 468 which was on the order of 10⁻¹⁵. Ergun and Happel–Brenner 469 470 models appear the most accurate, differing by only a factor of 471 100 from our measurement. We conclude that the onggi wall 472 is indeed a semi-permeable barrier to liquids. We proceed by 473 studying the effect of this permeability to carbon dioxide 474 on fermentation.

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475 We investigate the carbon dioxide generated by salted 476 Napa cabbage placed in an onggi and a glass jar. We perform 477 three experiments for each container and report the average 478 time course of carbon dioxide. We assumed that the salted 479 cabbage had negligible cellular respiration and generated 480 carbon dioxide mainly through bacterial fermentation. 481 Using equation (3.9), we converted carbon dioxide concen-482 tration to partial pressure. Figure 4c shows the time course 483 of the partial pressure of carbon dioxide for salted cabbage, 484 where the red line denotes onggi experiments and the blue 485 line denotes glass jar experiments. The onggi's permeable 486 wall clearly permits carbon dioxide to escape, resulting in a 487 slower increase in carbon dioxide and a lower plateau level. 488 Through the lens of our mathematical modelling, we will 489 see that the onggi involves greater carbon dioxide generation 490 than the glass container.

491 The main challenge with interpreting these results is that 492 the carbon dioxide generation rate is not explicitly known 493 since carbon dioxide is escaping the onggi's permeable 494 walls. To proceed further, we apply our mathematical 495 model, equation (3.11), to calculate the carbon dioxide gener-496 ation rate \dot{n}_g corresponding to each experiment. We constrain 497 our model with two conditions (1) to have a constant gas per-498 meability for each container and (2) to minimize the error 499 with respect to the time course of experimental carbon diox-500 ide. The modelling shows a good fit to our experimental 501 results, with R^2 values of 0.954 and 0.850, respectively, for 502 glass and onggi. In the electronic supplementary material, 503 we also show tests of cellular respiration for unsalted 504 cabbage.

Our calculation showed the glass container had a gas permeability k_g of $0.796 \times 10^{-18} \text{ m}^2$, while the onggi had more than double the value at $1.701 \times 10^{-18} \text{ m}^2$. We surmise that the permeability of the glass container was due to leaks from the three-dimensional-printed top. Since the onggi had a similarly manufactured top, its increased permeability was likely due to its porous walls. Previous researchers showed that the gas permeability of onggi depends on the glazing treatment, raw ingredients, and the kind of gas [3,20]. They presented permeance values only so we convert our permeability to permeance using equation (3.7). Previously measured gas permeance values for unglazed onggi lie between 0.68 and $19.75 \times 10^{-5} \text{ mol kPa}^{-1} \text{ m}^{-1} \text{ h}^{-1}$, which are on the same order of magnitude compared with our results, $1.70 \times 10^{-5} \text{ mol kPa}^{-1} \text{ m}^{-1} \text{ h}^{-1}$.

Figure 5*a* shows the calculated carbon dioxide generation rates (mmol h⁻¹) for onggi and glass containers. Salted cabbage carbon dioxide generation rates were 0.552 mmol h⁻¹, for glass and 0.695 mmol h⁻¹ for onggi. Thus, salted cabbage in onggi generated 26% more carbon dioxide than in the glass container, indicative of 26% more bacterial proliferation in the onggi. One-tailed *t*-test shows this difference was significant (P = 0.0498). Moreover, our carbon dioxide generation rates are also comparable to previous measurements. Given the carbon dioxide molecular mass of 44.01 g mol⁻¹, carbon dioxide generation rates per unit mass of cabbage for the glass and onggi were 2915 mg kg⁻¹ day and 3670 mg kg day, respectively, which were similar to that of fermenting kimchi in glass containers, 2531 mg kg day under 25°C and 2 wt% salt contents [30].

In conclusion, permeable onggi permits carbon dioxide to escape the container, which in turn accelerates the rate of fermentation. Porosity thus acts as a safety valve for carbon dioxide, maintaining its levels at less than half the values of hermetically sealed containers. Unlike a mechanical valve, there are no moving parts to break. The many pores permit a continuous outflow of gas, which helps reduce the entry of external contaminants. This oneway flow is shown schematically in figure 5*b*. In comparison to the passive gas control of the onggi, modern fermentation

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vessels use manual valves or other mechanical means to allow gas to escape.

5. Discussion

We discuss a few assumptions and caveats to be taken with our salted cabbage experiments. We assumed that the addition of salt to the cabbage triggered fermentation while impeding cellular respiration. Plants exposed to saline conditions experience physiological stress, disruptions to major biochemical processes, and inhibited mitochondrial activity [31,32]. While salt-tolerant plant species can maintain cellular respiration in saline conditions, we assume in this study Napa cabbage has inhibited cellular respiration under high salinity.

Given the high division rate of bacteria compared to plant cells, we infer that increases in carbon dioxide generation are due to bacterial proliferation. Since we cannot distinguish lactic acid bacteria from other bacteria, we assume that the increase in carbon dioxide is due to increased lactic acid bacteria alone. Our assumption is helped by the fact that salt supports lactic acid bacteria growth over other strains.

In our evaporation experiments, we found that onggi's 528 permeability can maintain most of the liquid inside while releasing the gases generated from fermentation. This ability to be semi-porous is a unique feature compared to other potteries. In comparison, terra cotta, a common pottery for houseplants, quickly leaks any water poured into it. For 533 plants, this property may be favourable by preventing the roots from sitting in stagnant water, but it's clearly poorly 535 suited for fermentation, a process where fermented vegetables should be kept moist.

The onggi in this study was unglazed. The traditional way to make onggi glaze is using natural ingredients such as water, tree ash and humus (leaf mould and soil), which come from 540 rotten grass and fallen leaves [3]. While the glaze is aesthetically pleasing, we surmise that it would reduce permeability 542 and in turn reduce its fermentation ability. 543

Our goal with designing the cover of the glass container was to make it hermetically sealed, but from our modelling, we find the glass container still had half the gas permeability of the onggi. We did not further improve the design because our focus in this study was on the onggi. For simplicity, we modelled both glass and onggi containers as having constant permeability. However, the permeability might be a function of pressure difference. For example, the glass container may be hermetic below a critical pressure, but then permeable as pressure increases. In fact, such nonlinear behaviour is how mechanical relief valves work: they have an energy storage mechanism in the form of a spring or gravity so that gas escapes when a critical pressure is reached.

The glass container only had half the volume of the onggi, and this difference may have influenced the bacteria's activity. Our mathematical model uses the container volume as a parameter to calculate the carbon dioxide partial pressure. However, the volume of the glass container could affect the interaction between the lactic acid bacteria and carbon dioxide. For example, if the container is large, the carbon dioxide may be diluted to a point where bacteria cannot detect any change. Such behaviour may provide an alternative explanation for the results observed.

Another assumption was a constant carbon dioxide generation rate \dot{n}_{g} , which reflects the average carbon dioxide rate generated by the lactic acid bacteria in the experimental interval of 48 h. In reality, the bacteria population will increase with time, and so will the carbon dioxide generation rate. However, we did not explore this scenario because it would require knowledge of the doubling time of the bacteria as well as their carbon dioxide generation.

The onggi is just one of many types of fermentation vessels [33,34] and cooking techniques [35,36] with ancient origins. Traditional fermentation vessels like oak barrels or amphora are mainly used for yeast-fermentation of alcoholic beverages, and especially the wooden barrels add flavour by ageing [37,38]. Onggi were designed to promote lactic acid fermentation, which provides the unique taste of kimchi.

6. Conclusion

We used fluid mechanics techniques to show that the onggi continuously 'breathes' carbon dioxide during fermentation, an idea that has been hypothesized for centuries, but had not been directly measured. We find that the onggi's porous walls are permeable to carbon dioxide, which reduces the carbon dioxide level inside the vessel. We surmise that this low carbon dioxide level is favoured by lactic acid bacteria, leading to their greater proliferation, and the increased carbon dioxide generation observed relative to hermetic glass containers. Our mathematical model predicts gas permeability that matches the range of previously reported values for onggi. We hope that this work draws interest to ancient cooking and food preparation strategies.

Data accessibility. The data and code supporting the findings of this study are available from the Github repository: https://github. com/Delicate-Kim/2023.

The data are also provided in electronic supplementary material [39]. Authors' contributions. S.K.: conceptualization, data curation, formal analysis, investigation, methodology, project administration, software, validation, visualization, writing-original draft; D.L.H.: funding acquisition, resources, supervision, writing-review and editing.

All authors gave final approval for publication and agreed to be held accountable for the work performed therein.

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