Effect of horizontal shift between fabric layers on the meso-scale-void formation in liquid composite molding

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Abstract. Accurate prediction of void content is the foundation of process optimization for Liquid Composite Molding (LCM). During mold filling, void formation is a three-dimensional flow process, while at present the void formation is treated as a result of in-plane air entrapment, the resin and air flows on the thickness direction are totally neglected. In this paper, resin flows in the adjacent layers are considered to analyze the three-dimensional air entrapment and the meso-scale-void formation. Firstly, the horizontal shift between fabric layers is analyzed, the mesopore coverage modes under different shift states are summarized. Then based on the mathematical models of micro and meso flows, the three-dimensional void entrapment processes under different cladding flow modes are studied in detail, a mathematical model for the prediction of meso-scale-void formation is established. Finally, a series of mold filling and void measurement experiments are carried out to verify the above model.

Keywords: polymer composites, liquid composite molding, void formation, modeling and simulation

1. Introduction

Void type defect is one of the most common process defects in the composite parts produced by Liquid Composite Molding (LCM), which greatly degenerates mechanical performances of the final products such as inter-laminar shear strength, flexural strength and compressive strength [1–3]. It has been reported that the inter-laminar shear strength of the laminate decreases more than 20% compared to a void free laminate when the void content is increased to 5% [4–6]. According to American Aeronautics Standard, final products with more than 2% of void defects should be rejected [7, 8].

The mechanical air entrapment behavior caused by the non-uniform resin flow is the main reason for the void formation in LCM. LCM process usually adopts the fabric preform, a fabric is interlaced by a lot of yarns with millimeter-sized sections, and a yarn is

composed by thousands of filaments with micrometer-sized sections. As a result, the millimeter-sized mesopores and micrometer-sized micropores are formed between and inside yarns, respectively. Therefore, resin flow in preform can be divided into meso flow in mesopores and micro flow in micropores. Since the mesopores have relatively large sizes, the capillary force is negligible, the injection pressure is the primary drive force of the meso flow; while the micropores have small sizes, capillary wicking is more important and becomes a main drive force of the micro flow. Furthermore, there is a huge difference between the flow resistances of mesopores and micropores. As a consequence, the non-uniformity of flow front between meso flow and micro flow occurs, which is known as the fingering phenomenon, the micro and meso voids will be formed if air is entrapped by flow fronts.

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Once voids are formed during mold filling, it is difficult to eliminate them completely [9, 10]. So studying the effects of preform structure and mold-filling parameters on void formation, and optimizing the process parameters to avoid or decrease the void formation become a significant method to improve the part quality. Researchers have conducted a large amount of studies on the modeling and prediction of void formation. Refs. [11–17] summarized and analyzed a mass of experimental data, found a relationship between void content of LCM part and the modified capillary number (Ca^*) as shown in Equation (1):

$$Ca^* = \frac{\mu \bar{u}}{\gamma \cos \theta} \quad (1)$$

where μ is the resin viscosity, γ is the surface tension of resin, θ is the contact angle between resin and fibers, and $\bar{u}$ is the global resin velocity. At low Ca^* , the global resin velocity is small but the micro flow velocity is high because of the capillary force, so meso-scale-voids are easily formed. At high Ca^* , on the contrary, the formation of micro-scale-voids is favored. Theoretical analysis method was adopted in Refs. [18–25] to analyze the micro and meso flows in a unit cell, and void formation prediction models were established by comparing the flow velocity difference. The shape, size and position of the formed void in the unit cell can be predicted and the relation between void content and Ca^* can be explained properly based on the models, so the theoretical analysis method has been widely approved. With the development of computer geometric modeling and numerical simulation, some scholars started to simulate the micro and meso flows in a unit-cell to predict void formation, such as Refs. [26–29]. They carried out a series of flow simulations using the cross section models of the unit cell to predict the resin flow behavior and void formation. The numerical methods used for solving the flow equations include finite element method, control volume method, VOF technology and boundary element method.

From above analysis, the formation of void is caused by the non-uniform resin flow in meso and micro pores, so the geometric constructions of pores have a significant impact on void formation. From the literature review we can find that the research object of all the current studies is a single unit cell of preform, and it is supposed that the void formation in the unit cell is only affected by its pore structure and size, the influence of other unit cells is not considered.

Actually, the preform used in practical application is usually obtained by lay-up of woven fabric layers, the interconnection of pores between adjacent layers definitely has some influences on the void formation. For example, with the variation of horizontal shift between fabric layers, the interconnection state of mesopores between adjacent layers changes, so the resin and air flow paths are unavoidably influenced and the void formation is ultimately affected. However, so far no research on the relation between inter-layer pore interconnection and void formation can be found.

In this study, the horizontal shift between fabric layers and its influence on void formation are analyzed, the mesopore coverage modes under different shift states are summarized. After studying the micro and meso flows on different coverage modes, a mathematical model for the prediction of meso-scale-void formation and its size is established. A series of mold filling and void measurement experiments are carried out to verify the correctness of the above model.

2. The horizontal shift between fabric layers

At present, void formation during LCM is predicted by analyzing the in-plane air entrapment induced by the velocity difference between micro and meso flows. As shown in Figure 1, if the mesopore has not been filled by resin when the two micro flow fronts converge, a meso-scale-void is formed. But actually, the resin and air flows are both three-dimensional, void is formed only when air is entrapped in three-dimensional space. As shown in Figure 2, the two micro flow fronts in the unit cell have converged, while the resin in the adjacent layer has not completely clad the mesopore, the entrapped air can still escape along the thickness direction, so the meso-scale-void has not formed completely.

Figure 1. The in-plane air entrapment.

Figure 2. Air escape along the thickness direction.

As can be seen from the above analysis, the difference of resin flow fronts on adjacent layers has direct influence on the formation of meso-scale-void, which is caused by the horizontal shift between fabric layers. Figure 3 shows a dual-layer unit-cell of woven fabric with no horizontal shift, the mesopore positions of the upper and lower layers are the same to each other. The dual-layer unit-cells of woven fabric with horizontal shifts are shown in Figure 4 and Figure 5, there is malposition between mesopores of different fabric layer.

Figure 4 shows a dual-layer unit-cell of woven fabric with horizontal shift in one direction, which means the relative shift of the two unit-cells occurs only on warp or weft direction. As the relative shift increases, the mesopore of the lower unit-cell is gradually covered by the yarn of the upper unit-cell, as shown in Figure 4a, until it is completely covered as shown in Figure 4b. Figure 5 shows a dual-layer unit-cell of woven fabric with horizontal shift on two directions, the relative shift of the two unit-cells occurs on both warp and weft directions. The mesopore of the lower unit-cell is also gradually covered by the yarns of the upper unit-cell, as shown in Figure 5a and 5b.

Figure 3. A dual-layer unit-cell of woven fabric with no horizontal shift.

Figure 4. A dual-layer unit-cell of woven fabric with horizontal shift on one direction. a) The mesopore is partially covered, b) the mesopore is completely covered.

Figure 5. A dual-layer unit-cell of woven fabric with horizontal shift on two directions. a) The mesopore is partially covered, b) the mesopore is completely covered.

Selecting the mesopore of the lower unit-cell as research object, the formation of meso-scale-void in this mesopore is affected by the covering yarns and its fiber orientations. Figure 4 and Figure 5 only demonstrate two horizontal shift directions between two neighbouring layers, actually, the relative shift on any direction can occur. However, the coverage of the present mesopore by the upper layer can be summarized to four basic modes, as shown in Figure 6, represented by mode A, B, C and D respectively. All possible horizontal shifts between two adjacent layers can be represented by one of the 4 coverage modes. Mode A and mode C are partial coverage, mode B and mode D are complete coverage.

In general, a fabric layer in mould has two adjacent layers, an upper layer and a lower layer, both them have effects on the formation of meso-scale-void in the present layer. The relationship between the present mesopore and the lower layer can also be represented by the four coverage modes shown in Figure 6. Therefore, there are 16 coverage modes for the present mesopore covered by the upper and lower

Figure 6. The basic coverage modes of the present mesopore by an adjacent layer.

layers, i. e., random combinations of the 4 basic coverage modes.

3. Mathematical modeling of void formation

3.1. Meso and micro flows

During Mold-filling, meso flow is the resin flow in mesopores outside yarns. Micro flow is the resin flow in micropores inside yarns, according to the flow direction, micro flow can be divided into longitudinal flow and transversal flow.

For the meso flow, the capillary force can be ignored since the relatively large pore size, the Darcy's law as shown in Equation (2) can be adopted to calculate the flow velocity:

$$u_c = -\frac{1}{\phi_c} \cdot \frac{K_{\text{unsat}}}{\mu} \cdot \nabla p \quad (2)$$

where u_c is the meso flow velocity, ϕ_c is the mesopore volume fraction which is derived by dividing the volume of the mesopore by the whole fabric volume, p

is pressure, K_{unsat} is the unsaturated permeability of the preform which can be obtained by the regression analysis of measurement data of global flow front position with time.

For the micro flow, the capillary force should be considered because of the small micropore size. Since the widths of a single yarn and a unit-cell are so small compared with the global flow distance, we can assume that the global resin pressure gradient is identical both in mesopores and in micropores and keeps unchanged while void is created at the flow front. Therefore, the micro flow velocity can be obtained from Equation (3):

$$u_t(K_t, P_c) = -\frac{1}{\phi_t} \cdot \frac{K_t}{\mu} \left(\nabla p - \frac{p_c}{l_s} \right) \quad (3)$$

where u_t is the micro flow velocity, K_t is the permeability of the yarn, which can be the longitudinal permeability $K_{t,l}$ or transversal permeability $K_{t,t}$, ϕ_t is the porosity of the yarn, l_s is the distance from the edge

of the yarn to the flow front, p_c is the capillary pressure, which can be the longitudinal capillary pressure $p_{c,t}$ or transversal capillary pressure $p_{c,l}$.

The permeabilities of yarn can be obtained from empirical formula, such as the widely used Gebart model [25] shown in Equation (4):

$$K_{t,l} = \frac{8}{53} \frac{(1 - v_{f,t})^3}{v_{f,t}^2} r^2$$

$$K_{t,t} = \frac{16}{9\pi\sqrt{6}} \left(\sqrt{\frac{v_{fmax,t}}{v_f}} - 1 \right)^{5/2} r^2 \quad (4)$$

where $v_{f,t}$ is the fiber volume fraction of yarn, $v_{fmax,t}$ is the maximal volume fraction of the tow which is 0.91 for the hexagonal packing, r is the radius of the fiber. The capillary pressure in Equation (3) can be obtained from Equation (5) and Equation (6) [27, 30]:

$$p_{c,t} = \frac{\gamma}{\pi r_e} \int_{-\pi/2}^{\pi/2} \frac{\cos(\theta + \eta)}{\sqrt{\frac{\pi}{2\sqrt{3} v_{f,t}} - \cos \eta}} d\eta \quad (5)$$

$$p_{c,l} = \frac{4\gamma \cdot \cos \theta}{r_e} \quad (6)$$

where r_e the capillary radius which can be approximated to half of the distance between the fibers, η is the angular location of the resin-air meniscus during flow between neighboring fibers.

3.2. Formation of meso-scale-void

The in-plane air entrapment in a single-layer unit-cell is shown in Figure 7, l and w are the length and width of the mesopore. From the meso flow path we can find that the meso flow distance for filling the mesopore is l , therefore, the mesopore filling time Δt_c can be computed by Equation (7):

$$\Delta t_c(l) = \frac{l}{u_c} = - \frac{l}{\frac{1}{\phi_t} \cdot \frac{K_{unsat}}{\mu} \cdot \nabla p} \quad (7)$$

Figure 7. The in-plane air entrapment in a single-layer unit-cell.

From the micro flow path shown in Figure 7 we can find that the micro longitudinal flow distance for entrapping air in the mesopore is $l + w/2$, so the time Δt_t required for the converging of the two micro flow fronts can be calculated with Equation (8):

$$\Delta t_t \left(l + \frac{w}{2}, K_{t,l}, p_{c,l} \right) =$$

$$= - \frac{l + \frac{w}{2}}{\frac{1}{\phi_t} \cdot \frac{K_{t,l}}{\mu} \cdot \nabla p} \left[1 + \frac{p_{c,l}}{(l + \frac{w}{2}) \cdot \nabla p} \ln \left(1 - \frac{(l + \frac{w}{2}) \cdot \nabla p}{p_{c,l}} \right) \right] \quad (8)$$

If $\Delta t_c < \Delta t_t$, the mesopore will be filled by meso flow before the micro flow fronts converging, no air will be entrapped; on the contrary, if $\Delta t_c > \Delta t_t$, the micro flow fronts will converge before the mesopore is filled, the in-plane air entrapment will occur, and the length of the entrapped area l_v can be calculated by Equation (9):

$$l_v = l - \int_0^{\Delta t_t} u_c dt \quad (9)$$

From the previous analysis in Section 2, it should be noted that the three-dimensional void has not been completely formed when the in-plane air entrapment occurs, the resin flow in the upper and lower adjacent layers should be considered. Using the coverage mode D shown in Figure 8 as an example, the cladding flow path in the coverage layer consists of two parts, the transversal flow part and longitudinal flow part, the flow distances of the two flow parts are l_1 and l_2 , respectively, so the flow times of the two parts Δt_1 and Δt_2 can be obtained from Equation (10) and Equation (11):

$$\Delta t_1 = \Delta t_t(l_1, K_{t,t}, p_{c,t}) =$$

$$= - \frac{l_1}{\frac{1}{\phi_t} \cdot \frac{K_{t,t}}{\mu} \cdot \nabla p} \left[1 + \frac{p_{c,t}}{l_1 \cdot \nabla p} \ln \left(1 - \frac{l_1 \cdot \nabla p}{p_{c,t}} \right) \right] \quad (10)$$

$$\Delta t_2 = \Delta t_t(l_2, K_{t,l}, p_{c,l}) =$$

$$= - \frac{l_2}{\frac{1}{\phi_t} \cdot \frac{K_{t,l}}{\mu} \cdot \nabla p} \left[1 + \frac{p_{c,l}}{l_2 \cdot \nabla p} \ln \left(1 - \frac{l_2 \cdot \nabla p}{p_{c,l}} \right) \right] \quad (11)$$

Therefore, the cladding time Δt_a required for resin in the coverage layer to completely clad the mesopore of the present layer is given by Equation (12):

$$\Delta t_a = \Delta t_1 + \Delta t_2 =$$

$$= \Delta t_t(l_1, K_{t,t}, p_{c,t}) + \Delta t_t(l_2, K_{t,l}, p_{c,l}) \quad (12)$$

Figure 8. The cladding flow in the coverage layer.

According to the position of the coverage layer, the cladding time can be divided into the upper layer cladding time Δt_{au} and the lower layer cladding time Δt_{al} . The total cladding time Δt_l can be obtained by Equation (13):

$$\Delta t_l = \max\{\Delta t_{au}, \Delta t_{al}\} \quad (13)$$

To accomplish the formation of a meso-scale-void, the in-plane air entrapment and mesopore cladding by resin in the adjacent layers need to be completed firstly. If $\Delta t_l > \Delta t_i$, the mesopore has been entirely cladded when the in-plane air entrapment, so the trapped air will not change and the meso-scale-void is fully formed, its length can be obtained from

Equation (9). If $\Delta t_l < \Delta t_i$, the mesopore has not been entirely cladded when the in-plane air entrapment occurs, the entrapped air can escape along the thickness direction until the mesopore is fully cladded, the final size of the meso-scale-void can be computed from Equation (14):

$$l_v = l - \int_0^{\Delta t_l} u_c dt \quad (14)$$

In conclusion, the length of the meso-scale-void can be obtained from the Equation (15):

$$l_v = \begin{cases} l - \int_0^{\Delta t_l} u_c dt & \Delta t_l < \Delta t_c, \Delta t_l > \Delta t_i \\ l - \int_0^{\Delta t_i} u_c dt & \Delta t_l < \Delta t_c, \Delta t_l < \Delta t_i \\ 0 & \Delta t_l \geq \Delta t_c \end{cases} \quad (15)$$

3.3. Influence of coverage mode and flow direction

According to the different coverage modes and flow directions, 16 different cladding flow modes can be obtained as shown in Figure 9, represented by A1, A2, ..., D3, D4. Shown in Figure 8 is the cladding flow mode D1. The cladding times for different cladding flow modes can be computed by Equation (16):

Figure 9. The cladding flow modes.

$$\Delta t_a = \begin{cases} \Delta t_t(l_1, K_{t,t}, p_{c,t}) + \Delta t_c(l_2), & \text{A1, A3, C1, C3 modes} \\ \Delta t_c(w), & \text{A2, A4 modes} \\ \Delta t_t(l, K_{t,t}, p_{c,t}), & \text{B1, B3 modes} \\ \Delta t_t(l, K_{t,t}, p_{c,t}), & \text{B2, B4 modes} \\ \max\{\Delta t_t(w, K_{t,t}, p_{c,t}), \Delta t_t(w_1, K_{t,t}, p_{c,t}) + \Delta t_c(w_2)\}, & \text{C2, C4 modes} \\ \Delta t_t(l_1, K_{t,t}, p_{c,t}) + \Delta t_t(l_2, K_{t,t}, p_{c,t}), & \text{D1, D3 modes} \\ \max\{\Delta t_t(w, K_{t,t}, p_{c,t}), \Delta t_t(w, K_{t,t}, p_{c,t})\}, & \text{D2, D4 modes} \end{cases} \quad (16)$$

Since the meso-scale-void is only formed under the low flow velocity, the cladding times of modes A2, A4, C1 and C3 are calculated under the hypothesis that meso flow is slower than micro flow.

4. Experiments and discussions

4.1. Materials and equipment

A series of mold filling and void measurement experiments were carried out to verify the above model. The preform used is a glass plain fabric EWR600 (Hualike New Materials, Changzhou, China), its structure sizes of unit cell are shown in Table 1.

The fiber volume fraction of yarns is 79%, the mean diameter of fibers is 13 μm . According to Equation (4), the longitudinal and transversal permeability values of the fiber tows are $1.1 \cdot 10^{-13}$ and $1.54 \cdot 10^{-14} \text{ m}^2$. According to experimental measurements [26], the fiber volume fraction of EWR600 fabric is 45.6%, the unsaturated permeability values of the fabric on warp and weft directions are $4.27 \cdot 10^{-10}$ and $5.26 \cdot 10^{-10} \text{ m}^2$, respectively.

The resin system used in the experiments is 191 unsaturated polyester resin (Thousands Chemicals, Jiangsu, China) and H-03 styrene hardener (Hualike New Materials, Changzhou, China), which can be cured at room temperature. In order to slow down the curing speed and reduce the change of resin viscosity during mold filling, the mix ratio of resin with hardener (parts by weight) is 100:0.5. The resin system has a viscosity of 0.3 Pa·s and a surface tension of 0.02 N/m at 25 °C which were measured respectively by NDJ-8S rotational viscometer (Shanghai

Precision Instrument Co. Ltd., Shanghai, China) and maximum bubble pressure method. The resin is assumed to have a perfect wettability, so its contact angle is set to 0°.

In order to cut the cured test specimen without demoulding, the acrylic plates were used as the upper and lower dies, the hot melt adhesive was used to seal the three sides of the mould except the outlet, then a cavity was obtained. The acrylic plates with 8 mm thickness were applied to increase the rigidity. The length and width of the mould used in the experiments are 200 and 100 mm, a 20 mm length free flow region was set as shown in Figure 10a to obtain a flat flow front. The inlet was set in this region on the upper die, resin fills the free flow region rapidly during mold-filling, then infiltrates the fabric with a 1D flow. Seven-layer fabrics were applied as the preform in experiments to avoid the influence of mould on air

Figure 10. The schematic diagram and photo of the mould used in the experiments. a) The detailed dimension of the mould, b) the actual photo of the mould.

Table 1. Geometric parameters of EWR600 fabric unit cell.

Description	Value
Width of the warp yarns [mm]	2.95
Thickness of the warp yarns [mm]	0.30
Length of the mesopore, l [mm]	0.73
Width of the weft yarns [mm]	3.24
Thickness of the weft yarns [mm]	0.30
Width of the mesopore, w [mm]	0.50

Figure 11. The mold filling equipment.

escape. The middle layer (the forth layer) was selected as the research object, its adjacent layers (the third and fifth layers) were both the coverage layers, there horizontal shift relative to the middle layer were identical. Figure 10b shows an actual mould used in the experiments.

The mold filling equipment is shown in Figure 11, constant flow rate injection at normal pressure was adopted, the TJ-3A/W0109-1B injection pump (Longer Corporation, Baoding, China) was employed which can adjust the flow rate on a large scale by changing syringe and driving velocity. The silicone tube was applied to connect the syringe outlet and mould inlet.

After mold filling, resin was cured at room temperature, and then the composite specimen is cut without demould by a milling machine, the cut position was the middle line of the forth layer fabric. The super deep scene microscope VHX-2000 (Keyence Corporation, Ōsaka, Japan) was used to observe and

Figure 12. Measurement of the meso-scale-void.

measure the size of meso-scale-voids after machining as shown in Figure 12. In order to increase data accuracy, five composite specimens were made at each process condition, and twenty voids in each specimen were measured to average, therefore, the size of a meso-scale-void at each process condition is obtained by the average of one hundred measurements.

4.2. Results and discussions

The experiments were implemented according to Table 2, the sizes of meso-scale-voids under A1, B1, C1, D1 cladding flow modes were measured, the coverage parameters and filling speed were varied to investigate there effects on the size of meso-scale-void. According to Equation (16), A1 and C1 modes have the same cladding times, so the same length of the meso-scale-voids will be generated under the same filling speed based on Equation (15). Shown in Figure 13 is the relationship between the length of meso-scale-void and l_1 under A1 and C1 modes, the parameter w_1 in C1 mode was 0.25 mm. As can be seen from the figure, with the increase of l_1 , length of meso-scale-void increases. Which is because the cladding micro-flow path increases with l_1 , the cladding time decreases since the micro-flow is faster than meso-flow, so the three dimensional air entrapment will be completed in advance and the length of meso-scale-void increases. The variation tendencies of meso-scale-void length of experiment measurement results and model prediction results are similar, while the experimentally measured length changes slower, which

Table 2. Experiment arrangement.

Cladding flow mode	Ca^*	l_1 [mm]	w_1 [mm]
A1, C1	$5.95 \cdot 10^{-5}$	0	0.25 (only for C1 mode)
		0.1	
		0.2	
		0.3	
		0.4	
		0.5	
		0.6	
B1, D1	$1.19 \cdot 10^{-5}$	0.365 (only for D1 mode)	—
	$2.38 \cdot 10^{-5}$		
	$3.57 \cdot 10^{-5}$		
	$4.76 \cdot 10^{-5}$		
	$5.95 \cdot 10^{-5}$		
	$7.14 \cdot 10^{-5}$		
	$8.33 \cdot 10^{-5}$		

Figure 13. Relationship between void length and l_1 under A1 and C1 modes.

may be caused by the plugging of the air escape path or the void evolution during curing process. From the figure we can find that C1 mode produce larger voids than A1 mode, this is because the yarn covered area of the present mesopore is larger in C1 mode, which has certain positive effects on the cladding flow. The difference between the longitudinal capillary pressure and transversal capillary pressure is quite small in the experiments, which causes the cladding flow of the mesopore is completed before in-plane air entrapment under B1 and D1 modes, i.e. $\Delta t_t > \Delta t_l$, in which case the length of meso-scale-void is only related to the modified capillary number. Presented in Figure 14 is the relationship between void length and modified capillary number under B1 and D1 modes. With the increase of Ca^* , length of meso-scale-void gradually decreases. This is because the increase of the macro flow speed has little effect on the micro flow (since capillary force is the main driver of the micro flow), while it accelerates the meso flow, which reduces the entrapped air in the mesopore. The model prediction results agree well with the experimental results.

Figure 14. Relationship between void length and modified capillary number under B1 and D1 modes.

5. Conclusions

Void content is an important quality indicator of composite part, accurate prediction of the void content is the foundation of process design and optimization for LCM. The existing models predict the void formation based on the in-plane air entrapment analysis, the resin and air flows on the thickness direction are totally neglected, which is obviously not rigorous. In this paper, the horizontal shift between adjacent fabric layers was analyzed firstly, four basic coverage modes of mesopore by an adjacent layer were summarized. Then based on the mathematical models of micro and meso flows, the three-dimensional void entrapment processes under different coverage modes and flow directions were studied in detail, and the mathematical model for the prediction of meso-scale-void formation and its size was established. Finally, a series of experiments were carried out to verify the above model. The results indicate that the air formation is a three-dimensional flow process, which is largely affected by the resin flow in the adjacent fabric layers, it should be considered in void content prediction.

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