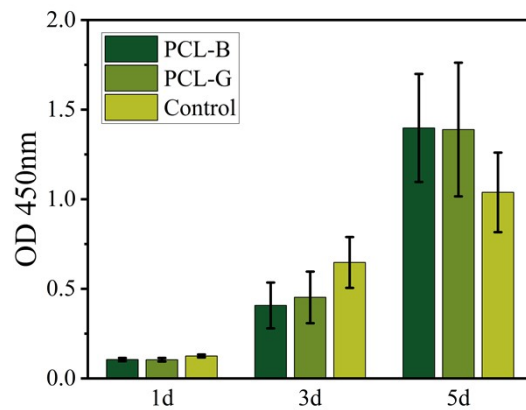


Supplementary Figure 1: Microscopic morphology of the bilayer membrane:

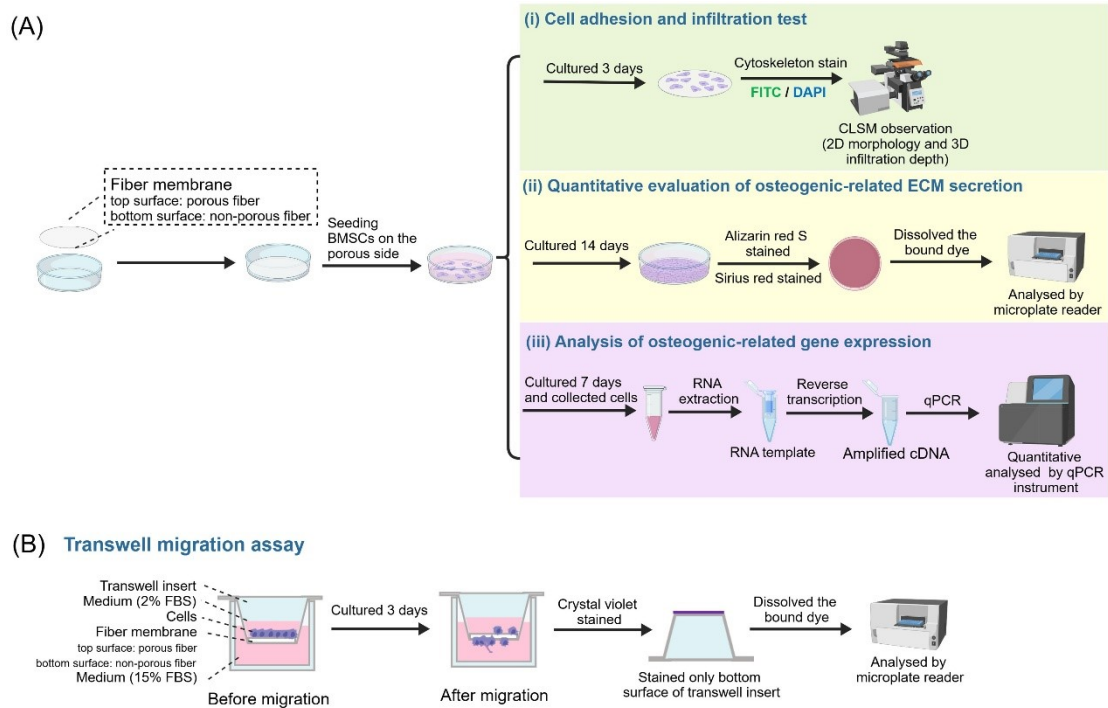
(A) Cross-sectional SEM image of the bilayer membrane, showing a distinct interlayer interface (indicated by the white dashed line). (B) SEM image of the dense layer surface facing the soft tissue side, showing relatively ordered fiber alignment, a smooth surface, and low porosity. (C) SEM image of the porous layer surface facing the bone defect side, exhibiting randomly arranged fibrous networks, a surface with micro- and nano-pore structures, and high porosity.



Supplementary Figure 2: Macroscopic demonstration of the flexibility of the bilayer membrane (twisting with tweezers)



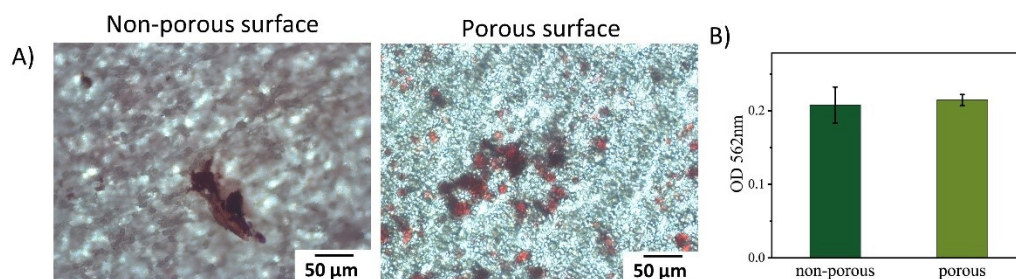
Supplementary Figure 3: Cell viability of BMSCs cultured on PCL-G and PCL-B membranes for 1, 3, and 5 days, evaluated by CCK-8 assay. Data were presented as mean  $\pm$  SD (n = 3).



Supplementary Figure 4: Schematic illustration of in vitro cell experiments.

(A) Schematic workflow of the cell adhesion, infiltration, osteogenic differentiation, and gene expression analysis assays: (i) For cell adhesion and infiltration assessment, BMSCs were seeded on the porous side of the fiber membrane, cultured for 3 days, and subsequently subjected to cytoskeleton/nuclei staining (FITC/DAPI) followed by CLSM observation to analyze 2D morphology and 3D infiltration depth. (ii) For quantitative evaluation of osteogenic-related ECM secretion, BMSCs were cultured on the membrane for 14 days, then Alizarin Red S staining and Sirius Red staining were performed. The bound dyes were dissolved and the absorbance was measured using a microplate reader for semi-quantitative analysis of calcium deposition and collagen secretion, respectively. (iii) For gene expression analysis, BMSCs were cultured for 7 days, collected for RNA extraction, and reverse transcription was conducted to synthesize cDNA,

followed by quantitative real-time polymerase chain reaction (qPCR) to quantify the expression levels of osteogenesis-related genes. (B) Schematic setup of the Transwell migration assay. BMSCs were seeded in the Transwell insert with the porous side of the fiber membrane facing upward, and the lower chamber was filled with complete medium (15% FBS) as a chemotactic stimulus. After 3 days of culture, cells that migrated to the bottom surface of the Transwell insert were stained with Crystal Violet. The bound dye was dissolved, and the absorbance was measured by a microplate reader to semi-quantitatively evaluate cell migration capacity.



Supplementary Figure 5: Mineralization of BMSCs on the non-porous and porous sides of PCL-G under non-osteogenic induction conditions for 21 days. (A) Representative images of Alizarin Red S staining, the red deposits indicate calcium nodules formed by BMSCs. (B) Semi-quantitative analysis of calcium deposition, measured as absorbance at 562 nm.

Supplementary Table 1: Primers sequences of Osteogenic marker genes

	Upstream primer	Downstream primer
<i>Gapdh</i>	CATGGCCTTCCGTGTTCCCTA	CCTGCTTCACCACCTTCTTGAT
<i>Runx2</i>	GAATGCACTACCCAGCCAC	TGGCAGGTACGTGTGGTAG
<i>Bmp</i>	CTGACCACCTGAACTCCAC	CATCTAGGTACAACATGGAG
<i>Col</i>	CCTGGTAAAGATGGTGCC	CACCAGGTTACCTTTCGCACC
<i>Opn</i>	TCCAAAGCCAGCCTGGAAC	TGACCTCAGAAGATGAACTC
<i>Ocn</i>	CAATAAGGTAGTGAACAGAC	CTTCAAGCCATACTGGTCT
<i>Osx</i>	GTCAAGAGTCTTAGCCAAAC	AAATGATGTGAGGCCAGATG
<i>Alp</i>	CCAACCTCTTTGTGCCAGAGA	GGCTACATTGGTGTGAGCTTTT

Video S1. gradient membrane and Video S2. bilayer membrane uniaxial stretching process. The videos clearly show the uniform extension of the gradient membrane until fracture, as well as the early-stage fracture of one layer during the stretching process of the bilayer membrane.