Supporting Information

1. Optimization of PVDF gel percentage and electrospinning parameters

Mixing of 10%, 12%, 15%, and 18% PVDF with solvent solution comprising DMF and acetone (2:1); followed by stirring the mixture by magnetic stirrer for 4 to 6 h, preferably at 70-80 ºC at 100-300 rpm. Electrospinning was done at room temperature with optimized parameters like extrusion rate of 0.5 mL/h under an applied voltage of 18 kV to 25 kV [1] (preferably 22.5 kV) while the distance between the needle tip and collector was at 15 cm, as depicted in table 1. Notably, major fabrication in the present process was to integrate aligned PVDF nano/micro fiber on D-FS through electrospinning, as mentioned the patent application by Dhara et al., 2025 [2].

Table 1: Process parameters for electrospinning of PVDF solution and integration with demineralized fish scale (D-FS) matrix.

Composition	Solution Percent	Applied Voltage	Tip to Collector Distance	Flow Rate
Electrospinning of PVDF	10	18 kV to 25 kV, preferably at 22.5 kV	~ 15 cm	~ 0.5 mL/h
	12			
	15			
	18			
Electrospinning of PVDF over D-FS	12	~ 22.5 kV	~ 15 cm	~ 0.5 mL/h

2. Swelling and Degradation Study

The swelling and degradation behaviours of the C-FS, D-FS, and E-FS composites were systematically evaluated to understand the influence of demineralization and electrospun PVDF integration on hydrophilicity and structural stability. Figure S1.a shows a sharp increase in swelling ratio within the first 30 minutes, followed by gradual stabilisation. The D-FS showed the highest swelling ratio throughout the immersion period, followed by the E-FS and C-FS. The pronounced swelling in D-FS can be attributed to the removal of mineral components such as hydroxyapatite, which exposes the underlying collagen fibrils and increases the number of hydrophilic functional groups (–COOH, $–NH_2$) available for water absorption [9]. In contrast, the CFS exhibited limited swelling due to its dense mineralized

structure that restricts water diffusion into the matrix. In contrast, E-FS sample demonstrated an intermediate swelling ratio, indicating that the electrospun PVDF layer provides a semi-permeable barrier that controls water uptake while maintaining flexibility. This controlled swelling behavior is advantageous for maintaining mechanical integrity and ensuring effective nutrient diffusion under physiological conditions, as reported by Su et al. (2025) [10]. The degradation profile in Figure S1.b revealed a gradual mass loss for all samples over a 14-day period. D-FS exhibited the highest degradation rate due to its susceptibility to hydrolytic/enzymatic attack (i.e., higher water uptake, exposed collagen). In contrast, C-FS retained most of its mass, confirming that the mineralized phase significantly resists degradation. The E-FS displayed a moderate degradation rate, slower than D-FS but higher than C-FS, suggesting that the electrospun PVDF network partially protects the matrix and reduces direct enzymatic degradation by forming a protective fibrous layer [11]. Representative sample images further illustrate the morphological changes after degradation and corroborate the observed weight loss percentages, as shown in Figure S1(c). D-FS shows greater matrix loss, while CFS remains compact and retains its shape up to 14 days of study. E-FS preserves shape better than D-FS and retains visible fibrous coating. That supports the idea of a semi-permeable electrospun barrier slowing degradation while permitting fluid access. This balance between structural protection and biodegradability is desirable for biomedical scaffolds that require temporary support during tissue regeneration. The observed degradation trend aligns with the findings of Dai et al. (2020) [12], which indicate that polymer-collagen composites with electrospun interfaces exhibit tunable degradation and enhanced biointegration properties. Overall, the swelling and degradation studies demonstrate that D-FS enhances hydrophilicity and degradability, while the electrospun PVDF coating effectively regulates these properties, ensuring mechanical stability and controlled bioresorption suitable for implantable applications.

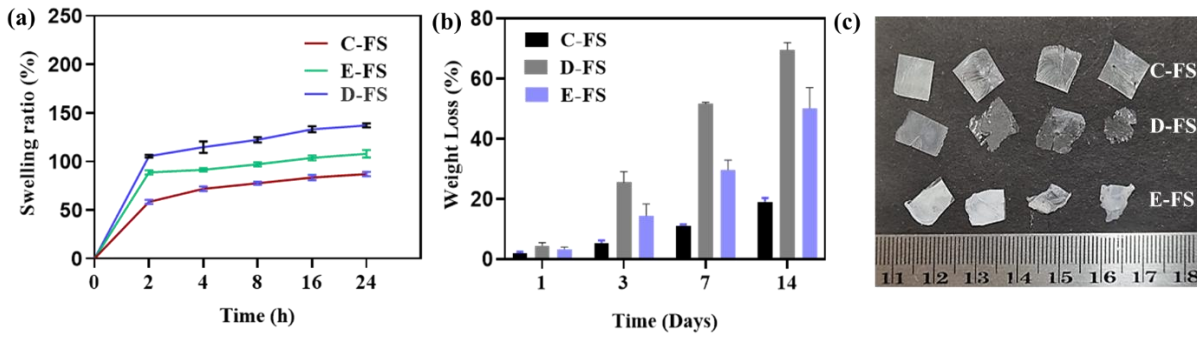

Figure S1: Swelling and degradation behaviour of C-FS, D-FS, and E-FS respectively: (a) swelling ratio percentage as a function of immersion time, showing enhanced water uptake in D-FS compared to E-FS and -CFS. (b) degradation profile over 14 days, indicating faster mass loss in D-FS and controlled degradation in E-FS. (c) representative images of samples after degradation, illustrating morphological changes and confirming the weight loss trend observed in (b).

3. SAXs data analysis for PVDF

The Small-Angle X-ray Scattering (SAXS) analysis of control, unaligned PVDF, and aligned PVDF samples elucidates their structural organization and changes in crystallinity, as depicted in Figure 3.d, 3.e, 3.f. The scattering intensity curves reveal that the control sample demonstrates a pronounced drop, indicating a more compact and less ordered structure with increased crystallinity. The unaligned PVDF exhibits a more gradual decline in intensity, indicating a decrease in crystallinity resulting from the random orientation of polymer chains. The oriented PVDF exhibits an intermediate trend, indicating a partial augmentation of crystallinity due to orientation.

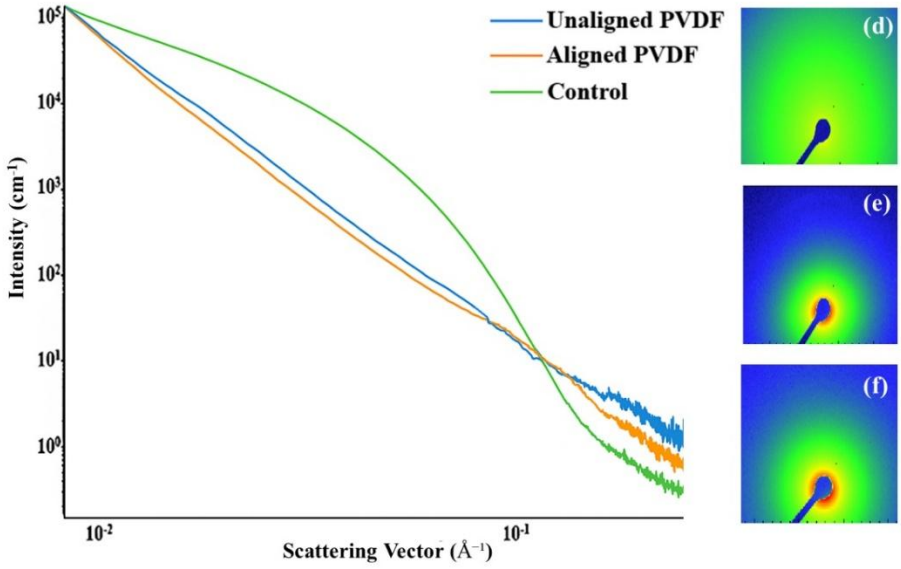

Figure S2: Merging of scattering pattern and plot using SAXS analysis between (d) Control (α-PVDF), (e) Unaligned, and (f) Aligned PVDF nanofibers.

References:

1. *He, Zhongchen, et al. "Electrospun PVDF nanofibers for piezoelectric applications: A review of the influence of electrospinning parameters on the β phase and crystallinity enhancement." Polymers 13.2 (2021): 174.*
2. *Dhara, S., et al. (2025). Mechano-stimuli device for electrical impulse using biotemplate nanohybrid and application thereofIndian Patent Application No. 202531043608, filed October 2025, India.*