

## Introduction

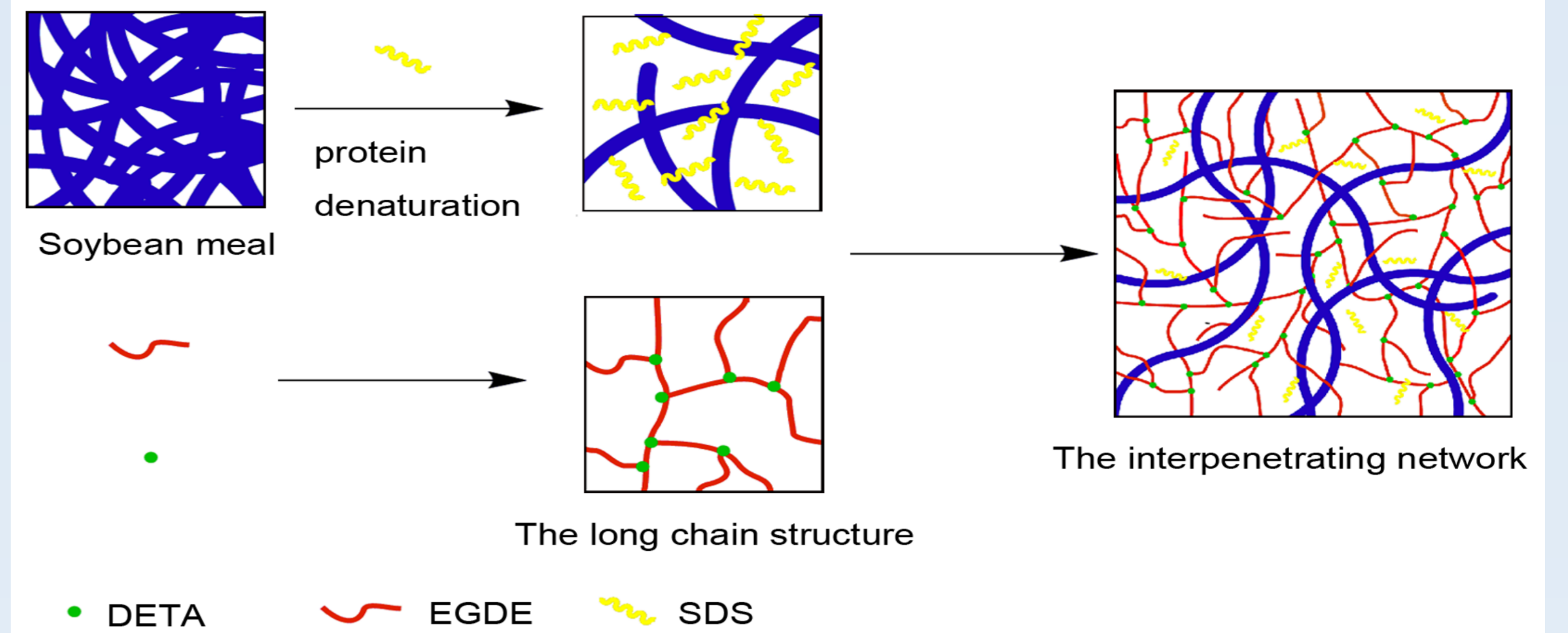
With the increasing concerns over environmental, deployment of biodegradable and renewable biomass for the production of wood adhesives is not only inevitable but also responsive to suppressing the impact caused by formaldehyde-based adhesives.

Soybean meal is an attractive raw material for the bonding wood. It is abundant, renewable, low price, and environmentally friendly. The potential issues for the soybean meal adhesive are the bonding strength and the water resistance.

## Objective

The objective of this study was to improve performance of the soybean meal-based adhesive by an ethylene glycol diglycidyl ether (EGDE) and diethylenetriamine (DETA). Three-ply plywood was fabricated to measure wet shear strength. The functional groups, cross section, crystallinity, thermal behavior, and physical properties of the resultant adhesives were characterized in detail.

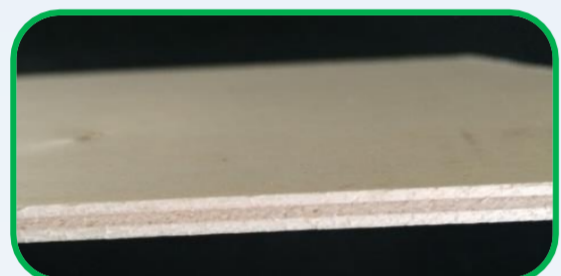
## Graphic Abstract



## Materials and Methods

### Preparation of soybean meal-based adhesives

Sample	Adhesive formulation
A (SM adhesive)	Soybean meal flour (28 g); Deionized water (72 g)
B (SM/SDS adhesive)	Soybean meal flour (28 g); Deionized water (72 g); Sodium dodecyl Sulfate (1 g);
C (SM/SDS/EGDE adhesive)	Soybean meal flour (28 g); Deionized water (72 g); Sodium dodecyl Sulfate (1 g); EGDE (10 g)
D (SM/SDS/EGDE/DETA adhesive)	Soybean meal flour (28 g); Deionized water (72 g); Sodium dodecyl Sulfate (1 g); EGDE (10 g); DETA (0.6 g)

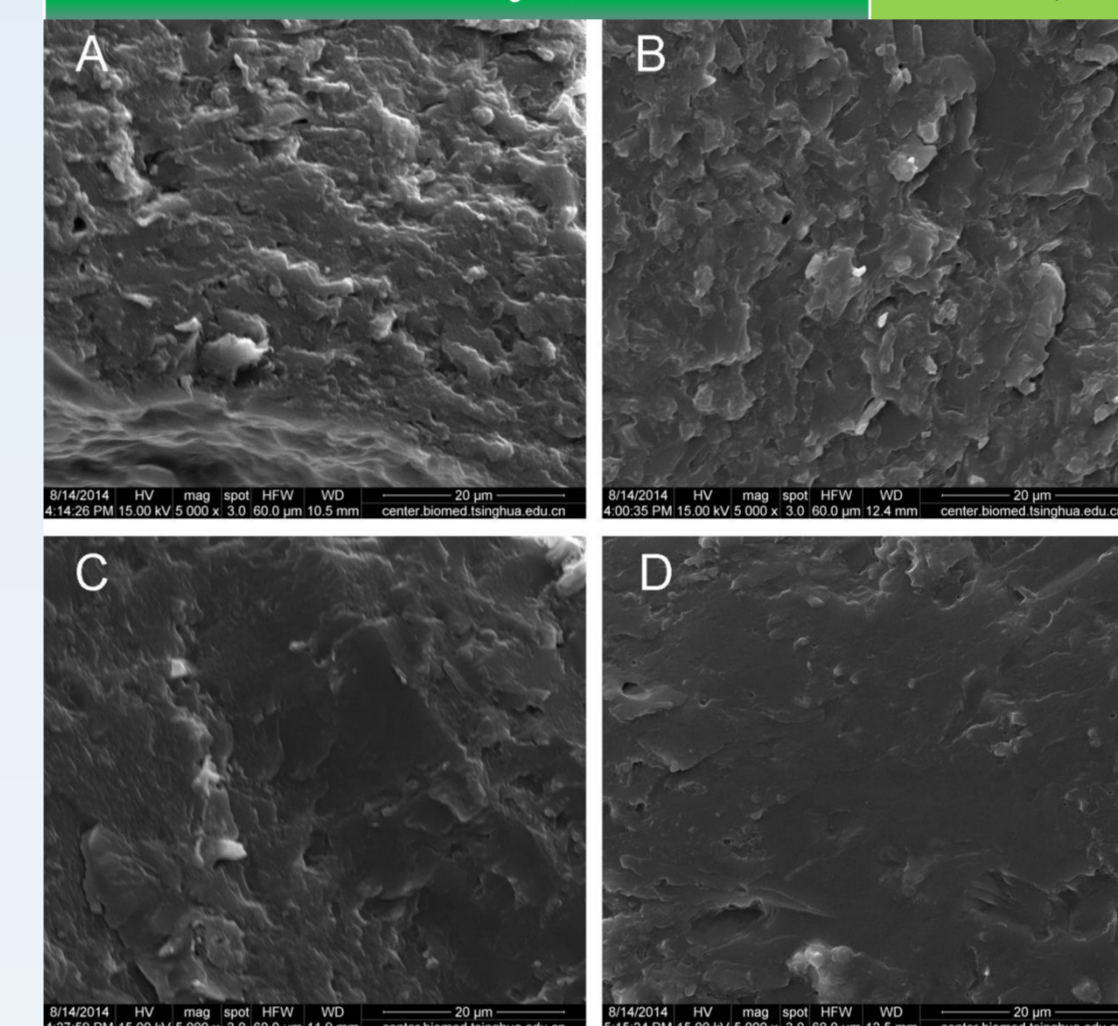


Three-ply plywood specimens were fabricated with the resulting adhesives and their wet shear strength was tested. And adhesive properties were further characterized.

## Results and Discussion

Table 1 Initial viscosity of different adhesives.

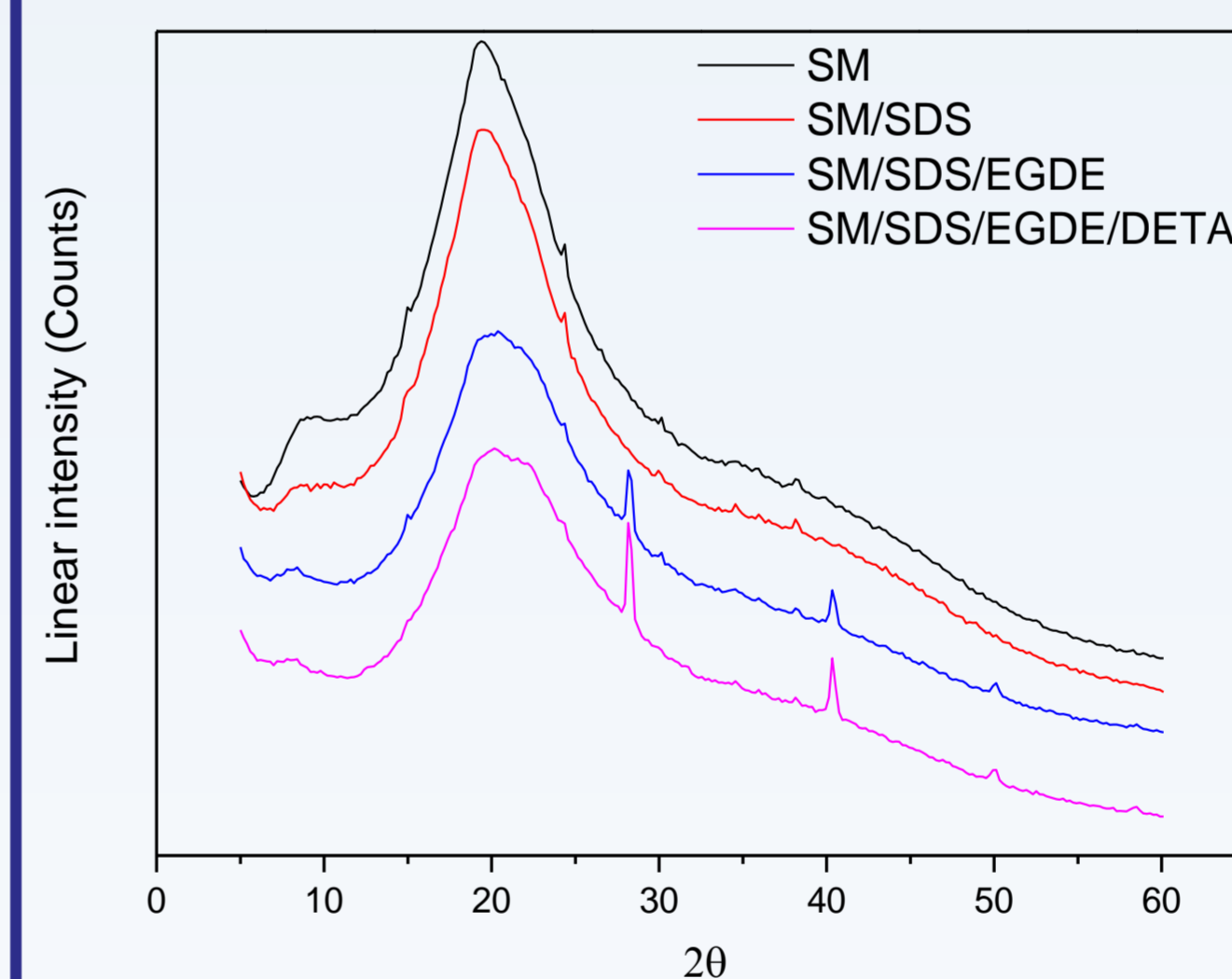
Adhesive formulations	A	B	C	D
Initial viscosity (mPa·s)	36,980	45,040	14,410	22,400



The incorporation of EGDE dramatically decreased the initial viscosity of the adhesive C by 68% from 45,040 to 14,410 mPa.s. This was because EGDE, which was a small molecular substance, could distribute in SM as a molecule lubricant to decrease the viscosity of the adhesive.

After DETA was introduced, a much smoother and more homogeneous surface was observed in the fracture surface of the adhesive D.

Fig. 4. The morphology of the fracture surface for the cured adhesives in different formulations.

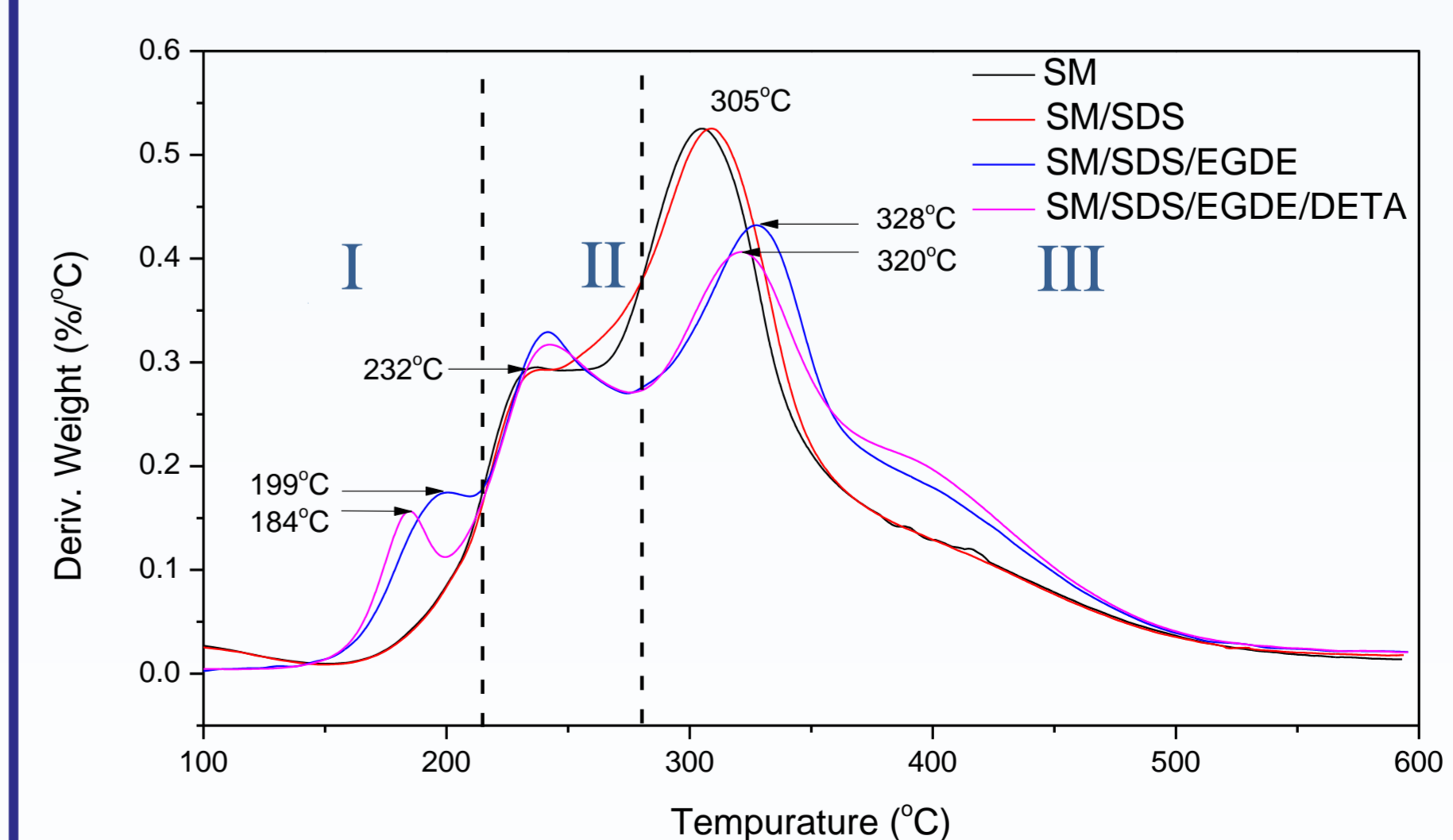


In general, increasing the cross-linking degree of the adhesive will lead to its crystallinity reduction. When adding EGDE to the adhesive B, the crystallinity of the adhesive C decreased, indicating that the cross-linking degree of the adhesive C was increased by the reaction between the EGDE and soy protein. After using DETA, the crystallinity of the adhesive D further decreased, which meant the DETA addition further increased the cross-linking degree of the adhesive.

Fig. 5. The XRD pattern of the different adhesive formulations:

Table 2 The crystallinity of the different adhesives:

Adhesive formulations	A	B	C	D
The crystallinity (%)	14.5	16.8	11	10.5



After incorporating EGDE/DETA, the post-cure process at stage I move to a lower temperature (184°C), indicating the reactivity of the EGDE/DETA was higher than that of the single EGDE.

Fig. 6. The DTG curve of the different adhesive formulations.

## Results and Discussion

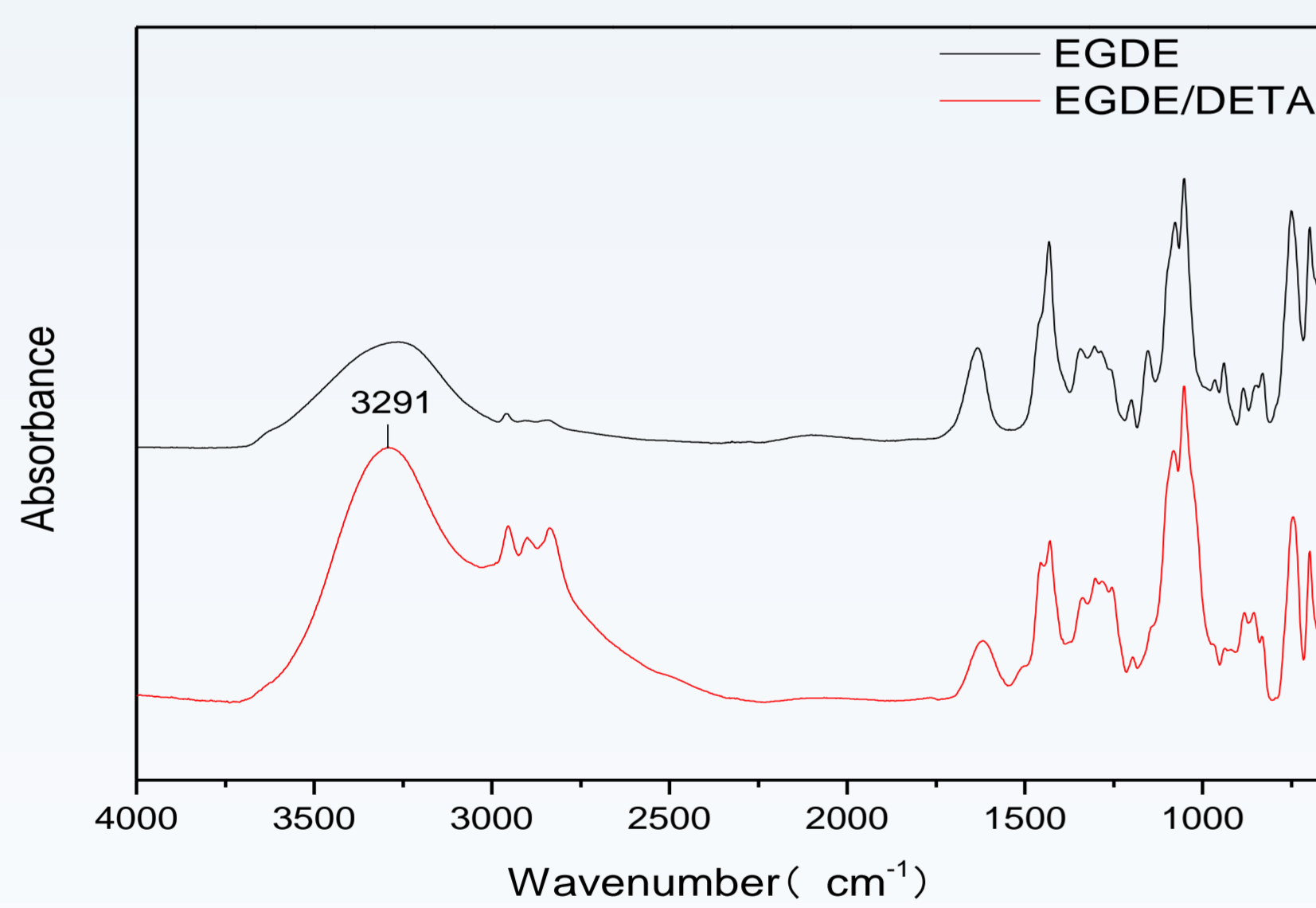
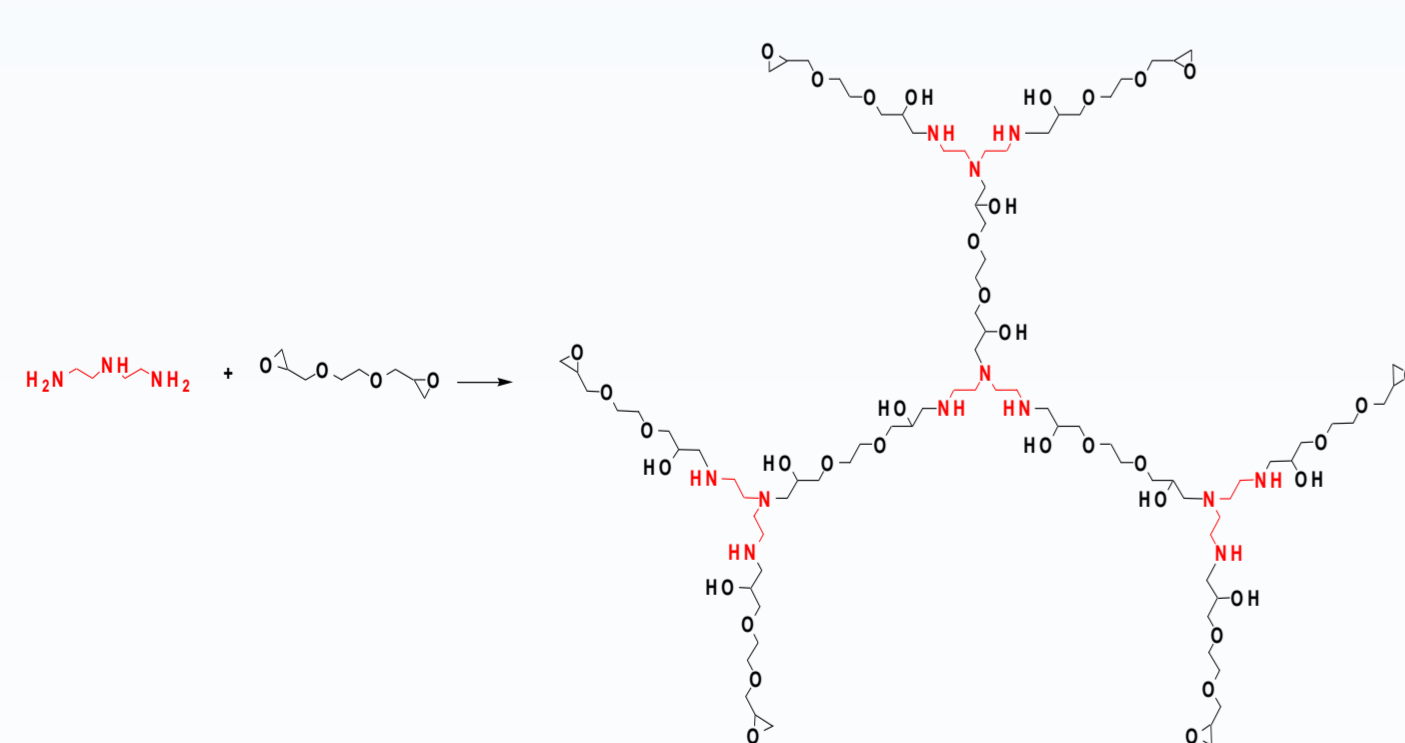
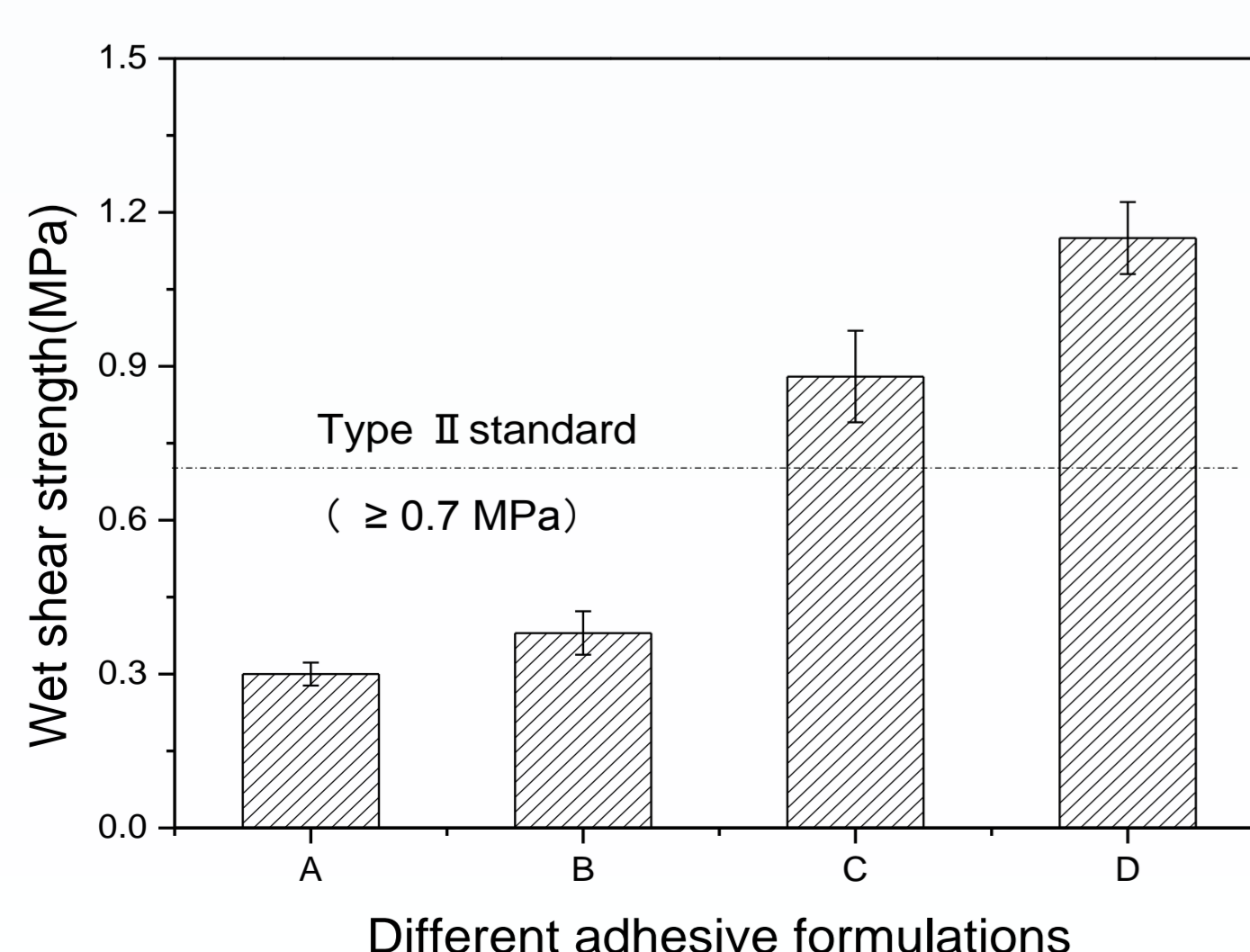


Fig. 1 The FTIR spectrum of EGDE and EGDE/DETA.

The reaction between DETA and EGDE increased the length of the molecular chain, which benefited for increasing the toughness of the EGDE. While, this reaction also increased the number of epoxy groups in a single molecule, which increased the cross-linking degree and benefited the water resistance improvement of the resultant adhesive during the curing process. The reaction was shown in Scheme A.



Scheme A The synthesis procedure for EGDE/DETA and its chemical structure.



When mixing the EGDE/DETA into the adhesive B, the wet shear strength was further improved by 30.7% to 1.15 MPa which was suitable for industrial application ( $\geq 1.0$  MPa).

Fig. 2. The wet shear strength of the plywood specimens bonded by different adhesives.

## Conclusions

1. DETA reacted with EGDE to form a long chain structure with epoxy groups, which cross-linked the soy protein molecules to form a denser cured adhesive layer and combine with the soy protein molecules to form an interpenetrating network. Both of them improved the water resistance of the resultant adhesive.
2. The wet shear strength of the plywood bonded by the SM/SDS/EGDE/DETA adhesive was 1.15 MPa and the adhesive viscosity was 22400mPa.s, which made the resultant adhesive practical for plywood industrial application.
3. After the incorporation of EGDE/DETA, the cured adhesive layer showed a smooth cross section, high cross-linking degree, and post-cure process, which benefited the water resistance improvement.