

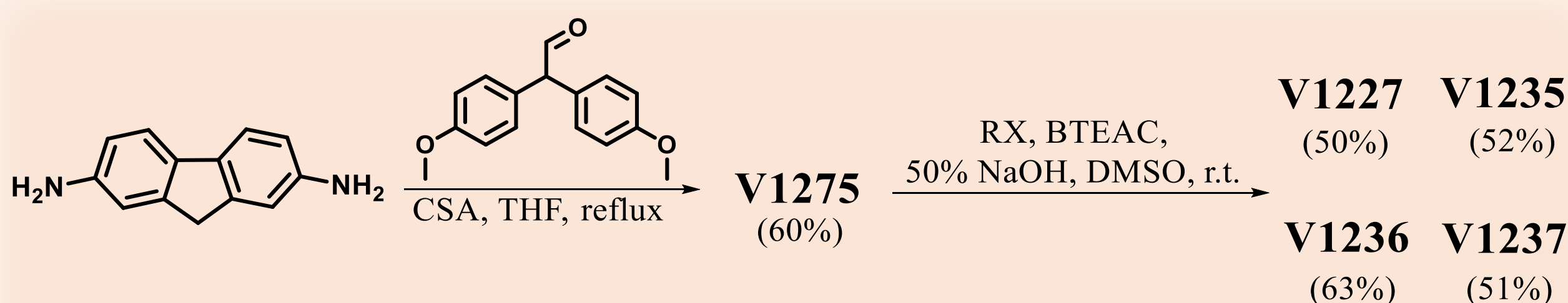
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Introduction

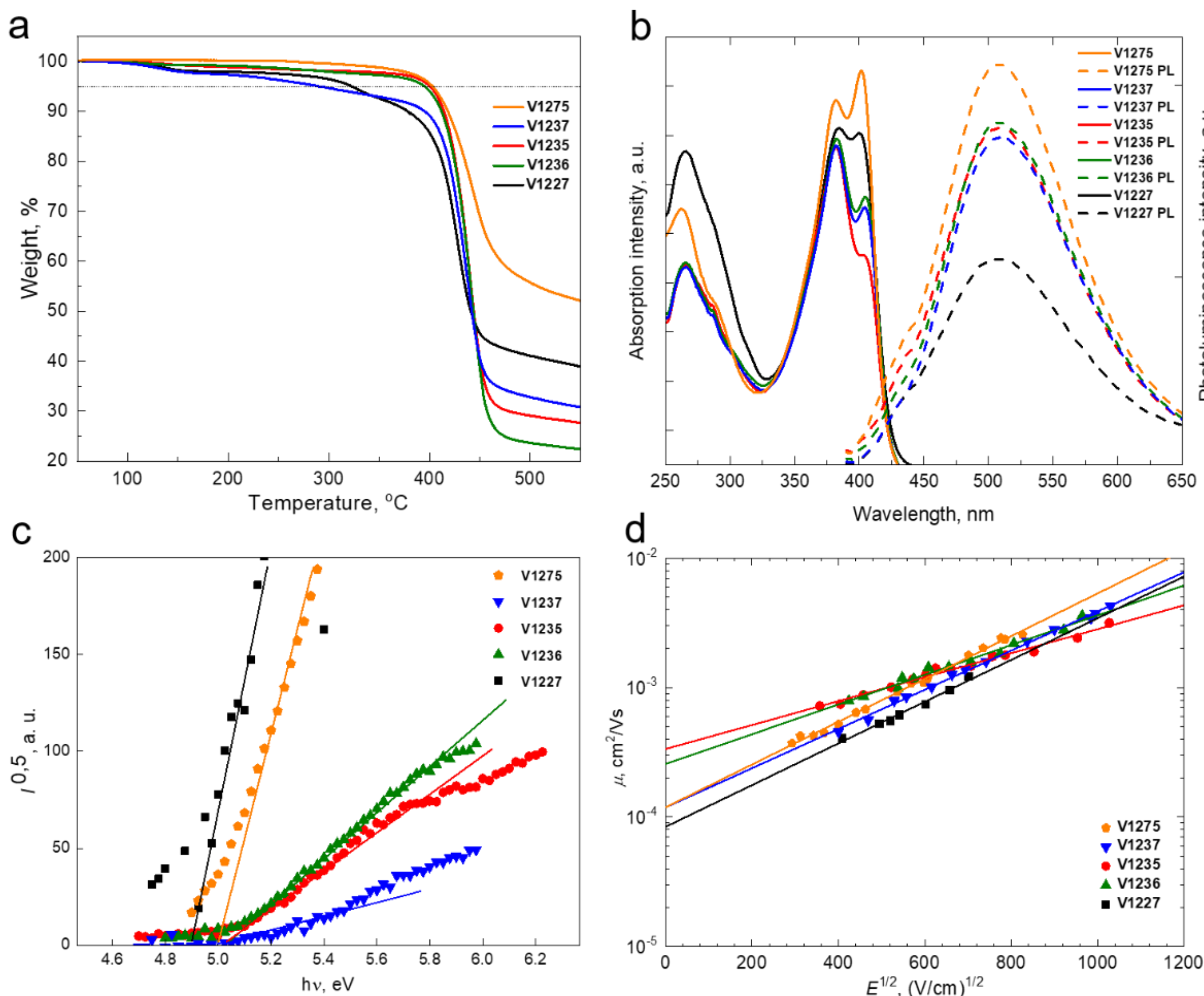
We report the synthesis, and a systematic study of the fluorene-based hole transporting enamines. Novel HTMs are easily attainable by a straightforward synthetic scheme ensuring cost-effective upscaling; in particular, **V1275** only required one-pot reaction condensing inexpensive commercially available reagents leading to the extremely low synthetic cost of approximately 10 €/g. The impact on different substitution in the central fluorene was revealed through the optical, electrochemical, photophysical, and photovoltaic measurements. PSCs using the V-series compounds were fabricated in doped- and dopant-free configuration. Synthesized materials exhibit very high hole mobility up to $3.3 \times 10^{-4} \text{ cm}^2/\text{Vs}$ leading to a light-to-energy power conversion efficiency exceeding 19% with the doped. The devices fabricated with dopant-free HTMs showed high efficiencies, exceeding 17%, with also excellent shelf-life stability. The results presented here show that HTMs prepared via simple condensation protocol can compete in performance with materials obtained via expensive cross-coupling methods at a fraction of their cost and may be very attractive low-cost and stable semiconductors solving one of the concerns for the near future commercialization.

Synthesis

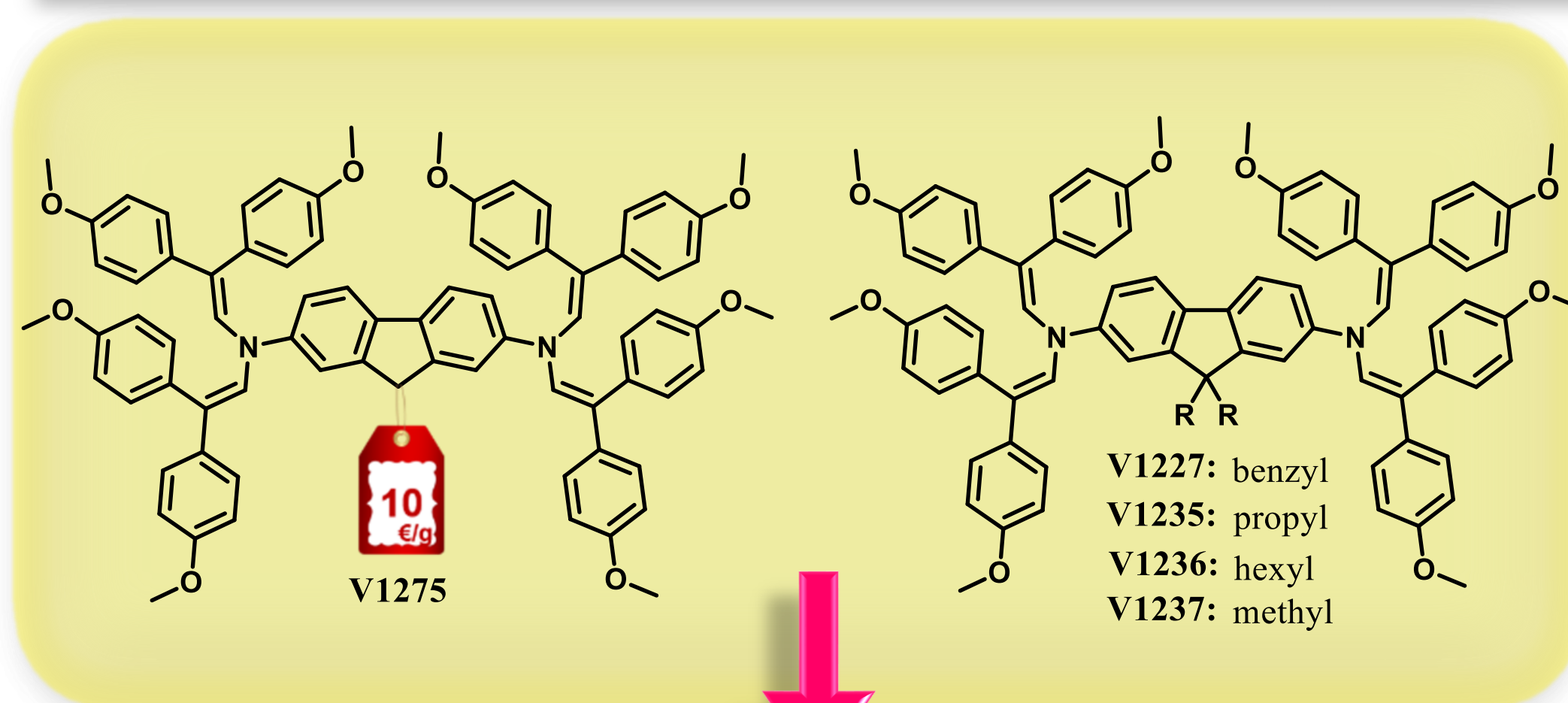


Properties

Fig. 1. (a) Thermogravimetric analysis (TGA) data; (b) UV-Vis absorption and photoluminescence spectra of V-series HTMs in THF solution 10^{-4} ; (c) ionization potential; (d) electric field dependencies of the hole-drift mobility in the synthesized HTMs



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Perovskite Solar cells

	HTM	PCE (%)
Doped-HTM	V1275	19.3
	V1237	19.2
	V1235	19.2
	V1236	19.1
	V1227	12.6
	Spiro-OMeTAD	19.7
Dopant-free HTM	V1275	17.1
	V1237	16.9
	V1235	16.6
	V1236	16.2
	Spiro-OMeTAD	10.4

Table 1. Thermal and photoelectrical properties of **V1275**, **V1237**, **V1235**, **V1236** and **V1227**.

HTM	T_m (°C) a)	T_c (°C) a)	T_g (°C) a)	T_{dec} (°C) a)	I_p (eV) e b)	μ_0 ($\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$) c)
V1275	255	-	150	403	5.01	1.2×10^{-4}
V1237	247, 267, 272	198	153	285	5.0	1.2×10^{-4}
V1235	273	159	120	399	5.03	3.3×10^{-4}
V1236	173, 195	-	90	393	5.03	2.6×10^{-4}
V1227	330	-	116	321	4.9	8×10^{-5}

a) Melting (T_m), crystallization (T_c), glass transition (T_g) and decomposition (T_{dec}) temperatures observed from DSC and TGA, respectively (10 °C/min, N_2 atmosphere); b) Ionization potential was measured by the photoemission in air method from films; c) Mobility value at zero field strength