## **European Journal of Organic Chemistry**

**Supporting Information** 

A Solid Alkylation: Highly Recyclable, Flow Chemistry-Ready, Resin-Supported Thioimidazoliums Alkylate Sulfur-Centered Nucleophiles

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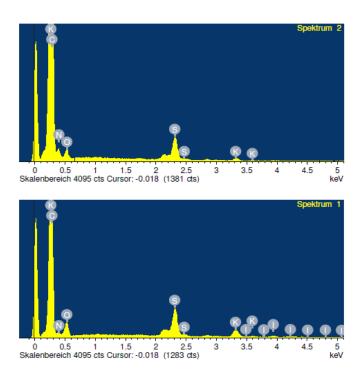
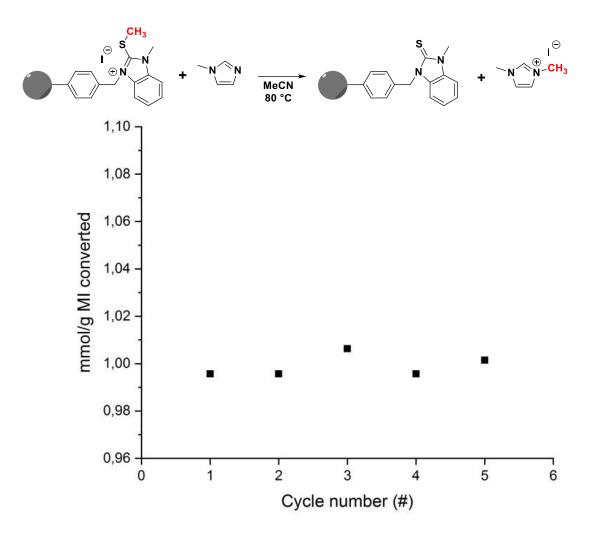


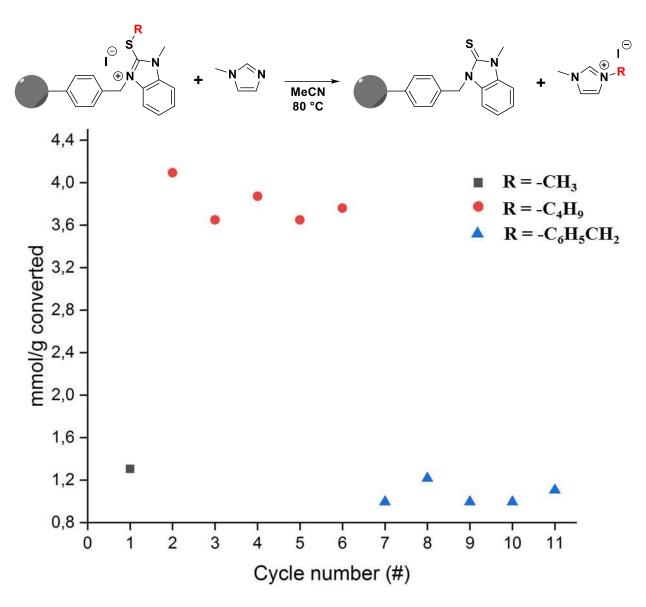
Figure S1. EDX spectrum before and after the functionalization of the resin with CH<sub>3</sub>I to prepare 2a.



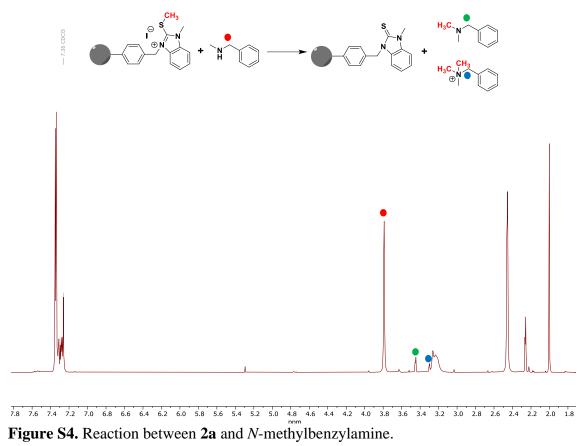
**Figure S2.** Alkylation efficiency of **2a** after five alkylation-regeneration cycles. The reaction investigated is reported above the graph.

**Table S1.** Influence of the regeneration solvent on the alkylation capacity (mmol/g) of resin **2a** as a function of regeneration solvent (determined by 1-methylimidazole conversion).

CH <sub>3</sub> CN	DMF	DMSO	MeOH	EtOH
1.106	0.774	0.996	1.217	1.438
1.216	0.774	0.885	1.217	1.217
1.106	0.885	0.885	1.217	1.217
1.106	0.885	0.885	1.106	1.327
0.996	0.774	0.995	1.106	1.327
1.106	0.819	0.929	1.173	1.305
	1.106 1.216 1.106 1.106 0.996	1.106 0.774   1.216 0.774   1.106 0.885   1.106 0.885   0.996 0.774	1.106 0.774 0.996   1.216 0.774 0.885   1.106 0.885 0.885   1.106 0.885 0.885   0.996 0.774 0.995	1.106   0.774   0.996   1.217     1.216   0.774   0.885   1.217     1.106   0.885   0.885   1.217     1.106   0.885   0.885   1.106     0.996   0.774   0.995   1.106



**Figure S3**. Alkylation efficiency of **2a** in the transfer of different alkyl chains. The reaction investigated is reported above the graph.



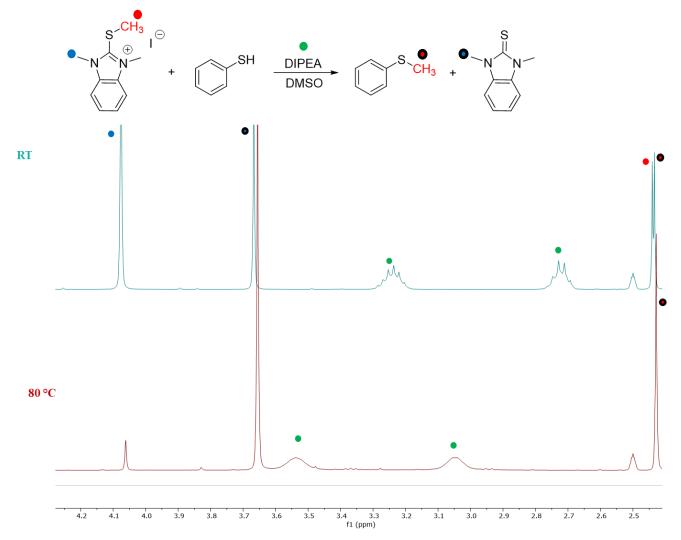


Figure S5. <sup>1</sup>H NMR for the reaction between IL-2a and thiophenol at RT and 80 °C.

Table S2. Comparison of the conversion of pyridine by CH<sub>3</sub>I and IL-2a DMSO-d6 at room temperature.

Alkylator	t0	10min	60min	72h
CH <sub>3</sub> I	23%	36%	58%	66%
IL-2a	1%	37%	47%	56%

**Table S3.** Comparison of the conversion of thiophenol by CH<sub>3</sub>I and **IL-2a** DMSO-d6 at room temperature.

Alkylator	t0	10min	60min	72h
CH <sub>3</sub> I	23%	36%	58%	66%
IL-2a	1%	37%	47%	56%

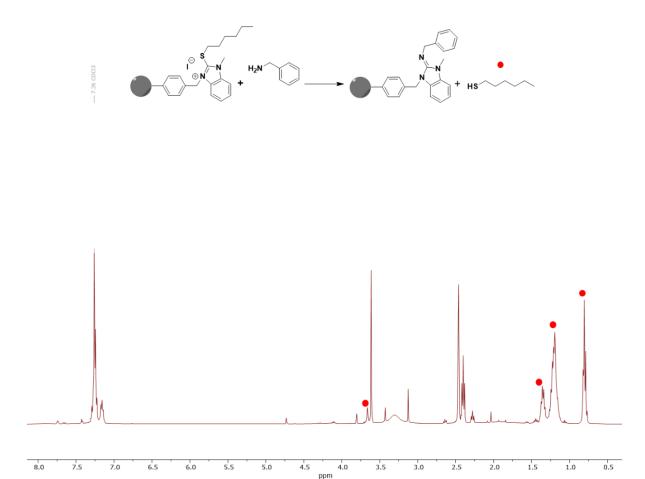
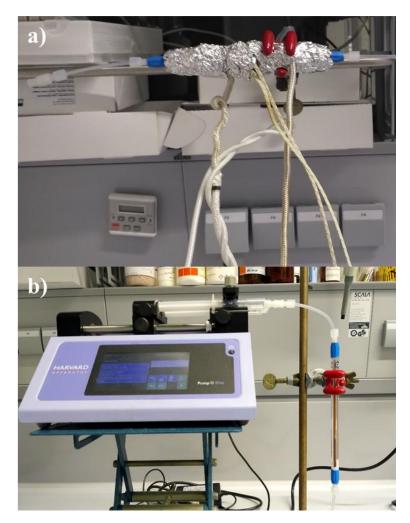


Figure S6. Formation of 1-hexanethiol due to poisoning of 2a by benzylamine.



**Figure S7.** a) Resin loaded glass tube surrounded by heating bands; b) Resin loaded glass tube connected to a syringe pump apparatus.

## **Author Contributions**

The project was designed by RG and JFT. Experimental data was collected by VAC, computational calculations by SMT and SM. Funding acquired and project administration by JFT and RG. VAC and SMT wrote the first draft, edited by VAC, SMT, SM, JFT, and RG.