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# Kinetic selection of polymeric guanosine architectures from dynamic supramolecular libraries

## *Sélection cinétique des architectures polymères de guanosine à partir des bibliothèques supramoléculaires dynamiques*

Anca Meffre<sup>1</sup>, Eddy Petit, Didier Cot, Mihail Barboiu\*

Adaptive Supramolecular Nanosystems Group, Institut européen des membranes, ENSCM–UMII, UMR CNRS 5635, place Eugène-Bataillon, CC047, 34095 Montpellier cedex, France

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## ABSTRACT

We report an original strategy to transcribe and to fix supramolecular guanosine architectures in self-organized polymers. In the first resolution step, the G-quartet and G-quadruplex architectures are pre-amplified in solution in the presence of  $K^+$  cations from a dynamic pool of ribbon-type or cyclic supramolecular architectures. Then in a second selection polymerization step, the G-quadruplex is kinetically fixed in a covalent polymethacrylate network via an irreversible amplification step. Both supramolecular and polymeric components mutually (synergistically) adapt their spatial constitution during simultaneous (collective) formation of micrometric self-organized hybrid domains. This contributes to the high level of adaptability and correlativity of the self-organization of the supramolecular G-quadruplexes and of the polymeric systems. Biomimetic-type hybrid systems can be generated by using this strategy.

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## 1. Introduction

Constitutional self-assembly provides evolutionary approaches for the generation of functional systems through the implementation of reversible exchanges between different complex architectures of variable functionality [1,2]. Much efforts continue to be undertaken on complex systems, which have been identified as an especially promising means to explore the chemical space with natural selection of their structural and functional

behaviours [3]. These new concepts may be connected today with the simple definition of Dynamic Multicomponent Self-assembly in chemical and biological systems [4]. They concern collections of molecular/supramolecular objects reversibly exchanging and continuously organizing at the nano- or macroscopic levels.

A representative example is related to the architecture, the H-bonded supramolecular macrocycle formed by the self-assembly of four guanosines and stabilized by alkali cations. It may embody an important constitutional (spatial and interactional) reorganization with different G-networks, dynamically exchanging between G-ribbons and G-quartets or their stacked tubular G-quadruplexes (Fig. 1) [5a].

Although discovered in the 1960s [5b], the functionality of the artificial G-quadruplexes as ion-channels and their direct quantification in human cells have been only

\* Corresponding author.

E-mail address: mihail-dumitru.barboiu@univ-montp2.fr (M. Barboiu).

<sup>1</sup> Present address: Université de Toulouse, INSA, UPS, laboratoire de physique et chimie des nano-objets, LPCNO, 135, avenue de Rangueil, 31077 Toulouse, France.

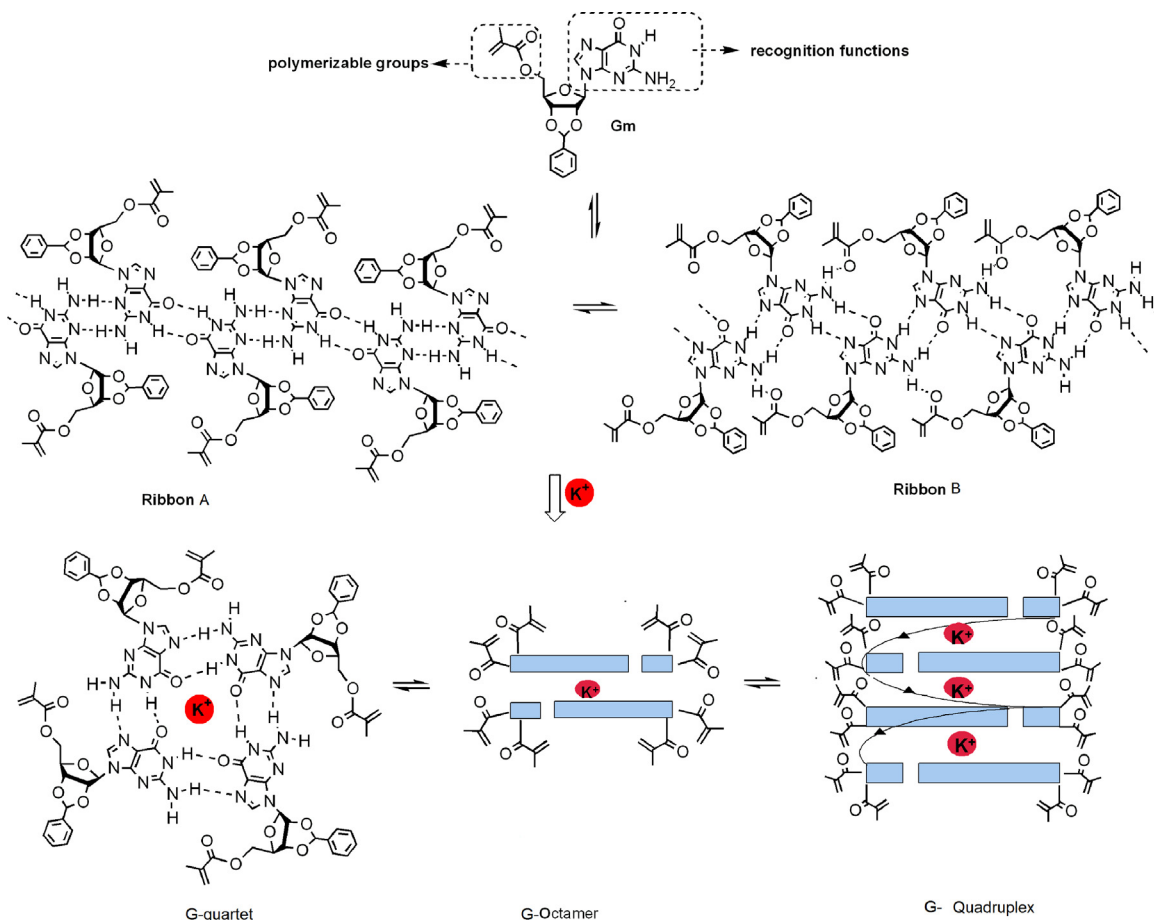


Fig. 1. Dynamic exchanges between supramolecular architectures G-ribbons, G-quartets or G-quadruplexes in the presence of ionic chemical effectors (red sphere).

recently emphasized [6]. Liposomes [7], surfaces [8], dynamers [9], hybrid materials [10,11], mesoporous silica [12] or silicon [13] have been used as scaffolding matrices to stabilize and to orient the anisotropic directional G-mesophases.

We have recently showed that *sol-gel selection* can be used as an irreversible kinetic process in order to stabilize the *G-quadruplex* and to provide long-range amplification of its supramolecular chirality into hybrid organic-inorganic twisted nanorods. This may be followed by a second *inorganic transcription* via calcination into inorganic replica materials, when silica microsprings can be obtained [10]. A second used strategy is related to the implementation of dynamic hydrophobic non-covalent [11a] or reversible-covalent iminoboronate [11a] interactions between the supramolecular *G-quadruplexes* and the siloxane/polymeric constituents. The reversibility of the interactions between components represents a crucial factor and, accordingly, a dynamic reversible hydrophobic interface might render the emergence of organic/inorganic mesophases self-adaptive. They mutually may adapt their 3D spatial distribution based on their own structural constitution, during the simultaneous formation of hybrid self-organized domains. [14] It is known that the multicomponent self-assembly of the *G-quadruplexes*

superstructures can be also achieved by using the reversible metathesis polymerization reaction, as previously demonstrated by Davis et al. [15].

We hypothesized that the stabilization of guanine supramolecular architectures may be obtained *via* a classical polymerization process. Herein we describe this strategy by using 5'-methacrylate guanosine,  $G_m$ , self-assembling in solution in *G-quadruplexes*, followed by their irreversible transcription in an organic polymeric network *via* intermolecular cross-linking of methacrylate end groups. These results provide new insights into the basic features that control the convergence of supramolecular self-organization and polymerization process toward functional materials at the nano(micro)level.

## 2. Experimental

### 2.1. Materials and methods

All reagents were obtained from Aldrich and used without further purification. All organic solutions were routinely dried by using sodium sulfate ( $Na_2SO_4$ ). Polymerizations were performed under an inert atmosphere of nitrogen, using standard Schlenk techniques. Tetrahydrofuran was dried and distilled prior to use over Na. The

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