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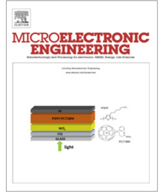

Review Article

Directed self-assembly of block copolymers for nanocircuitry fabrication

M.A. Morris *

CRANN, Trinity College Dublin, Dublin, Ireland
Department of Chemistry, University College Cork, Cork, Ireland

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ABSTRACT

This paper is a perspective on progress that has been made in the use of block copolymers as potential, non-UV lithographically formed, on-chip etch masks for the fabrication of ultra-small circuit elements. Rather than focus on a comprehensive review of the fundamentals of the self-assembly process, the article will provide a personal, retrospective appraisal of what has been done and what needs to be done using published and unpublished material. It is hoped it will provide a summary of the field to as wide an audience as possible. The science of the self-assembly of these systems will be briefly reviewed. We will provide a brief overview how block copolymers can produce ordered nanopatterns, how they can be 'directed' to provide long-range order and how the patterns formed can be transferred to a substrate. In the article we will review key developments over the last 10 years or so that have moved these materials from the laboratory to a point where they are close to use in integrated circuit (IC) manufacturing. We will centre on challenges that remain in the chemistry and processing of block copolymer films that if successfully met would allow their insertion into current IC manufacturing. In particular we will explore issues such as: definition of pattern alignment and orientation, defectivity in the as formed systems and development of reliable process flows. During the article, we will examine emerging challenges such as the need to develop systems that can reach feature sizes of sub 10 nm, techniques such as solvent-annealing that can be used to generate patterns in short times and polymers that can be designed or modified to give high etch contrast during pattern transfer. We also will look at the future of these systems and where they may add value to the production of nanowires, nanodots etc. for application in micro- and nanoelectronics.

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1. Introduction

The micro-, nano-electronics industry has developed around the use of photolithography to create surface features [1]. A light sensitive polymer (becoming either more or less soluble in a chosen solvent on light irradiation) is used as 'resist' which can be patterned using ultraviolet light exposure through a photomask. Subsequent removal of exposed areas of the resist in a selective solvent yields a topographic polymer pattern which can be used as an etch mask for transfer to the underlying substrate. Miniaturisation of transistor size and the corresponding increase in their density has driven the microelectronic industry for over 60 years and enabled the mass production of chips of staggering processor speed, operating at low powers and manufactured at low cost; currently, pitch and feature sizes are at sub 20 nm dimensions [2,3]. Photolithography has been at the very heart of this progress. Feature dimensions have shrunk and are now below the wavelength of light used (193 nm) [1] to pattern them. However, the use of 193 nm wavelength light for photolithography has been ongoing and successfully implemented for over ten years since the advent of commercial argon fluoride laser sources. Efforts to move to shorter wavelengths (extreme ultraviolet (EUV) at 13.5 nm) for patterning, which are consistent with shrinking device dimensions, have been faced with technical challenges such as EUV source power output, the availability of defect free masks/detection systems and EUV's considerable cost [4]. Research and development work to address these technical challenges is ongoing and maturing towards a manufacturing ready EUV patterning solution. As of today, state-of-the-art devices are currently manufactured using multiple patterning (MP) methods (using 193i lithography) that require the use of repeated exposures and very accurate wafer positioning so that feature sizes can be trimmed [5]. These MP methods require nm accuracy in terms of mask-sample positioning (for overlay and registry) between exposures and this reduces wafer throughput and increases capital and other costs dramatically.

* Corresponding author at: Department of Chemistry, University College Cork, Cork, Ireland.

E-mail address: m.morris@ucc.ie

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As feature sizes have shrunk, they have entered ranges where dimensions are similar to those achievable by molecular self-assembly [6]. Self-assembly is the name given to the way in which molecules, molecular aggregations or particulates spontaneously form well-defined arrangements or, in some cases, patterns of long-range order in an effort to minimise the free energy of the system [6]. The advantages of a self-assembled process are that it can be rapid, require low capital costs and lead to very small feature sizes (sub 5 nm) unobtainable by conventional photolithography. The challenge, of course, can be attaining a similar structural regularity of a pattern formed by chemical interaction to that of a physically defined pattern such as that formed in photolithography. A number of appropriate reviews from a large volume of work are provided [7]. To be considered as a viable manufacturing alternative to photolithography, self-assembly needs to overcome four important challenges. Firstly, the pattern arrangement formed must be similar to that used in current device technologies (i.e. lines, spaces, holes). Until recently, this was not the case because chip layouts were much more complex than the limited periodic arrangements offered by self-assembly [5]. Secondly, there has been industry resistance to the introduction of new material sets because of integration concerns. However, the introduction of novel masks, new resist compositions, high and low dielectric constant materials and device integration (e.g. magnetics and electronics) has led to a 'quiet' revolution in the number and variety of on-chip materials [8]. Thirdly, with shrinking device dimensions and within layer pattern density increasing, addressing the challenge of acceptable pattern placement error and registry to previous layers in an integrated circuit, is one of the more significant technical challenges to implementing self-assembly as a new potentially complimentary lithography IC manufacturing technique. Lastly and most significantly, the pattern formed by self-assembly must have defect densities similar to those formed by photolithography [9]. By necessity, the integrated circuit industry has become ever more receptive to these emerging technologies but it is the defect challenge that has prevented their introduction. In simple terms, it is this statistical nature of self-assembly which leads to high defect densities and dimensional variation from feature-to-feature. In recent years, one form of self-assembly has emerged as having potential to form highly periodic arrangements over wafer and other large area surface dimensions. This is block copolymer self-assembly and it is this methodology that is the focus of this review. In particular, photolithography for IC production is the main considered application of BCPs in this review. However right across the developing nanotechnology spectrum, where any form of surface or material nanostructuring is needed, BCP's are being considered as materials sets for use. A number of recent reviews have been published which highlight the progress and challenges associated with these new and novel applications [7].

2. Background to microphase separation in block copolymer systems

The self-assembly of block copolymers (BCPs) is more properly described by the term microphase separation. A block copolymer consists of linear chains or star-like arrangements of two or mole molecular units arranged in distinct segments. The simplest system is a diblock copolymer which can be represented as $A_n - B_n$ where A and B are the molecular units and n and m are the numbers of each unit. Microphase separation results from the system minimising repulsive (high energy) interactions between dissimilar blocks and maximising attractive, low energy, interactions between similar blocks [10]. Effectively, the system attempts to minimise the interfacial surface area between unlike blocks. Because of the covalent bond between blocks, phase separation can only occur on a scale similar to that of the polymer chain size and the polymers segregate into well-defined domains of the individual blocks. In depth studies of this phenomenon are relatively recent and Gompper and Schick provide a good review of the very early literature [11].

Theory and understanding of microphase separation in bulk systems are well developed and phase diagrams relating polymer composition, molecular weight and the chemical dissimilarity have been defined [12,13]. The result of the intermolecular interactions between blocks is a series of well-defined, ordered arrangements of the chemically distinct domains. These arrangements are similar to the structures formed in lipid and micellar systems and consist of several different phases: a lamellar structure, a structure consisting of hexagonally packed cylinders, a body-centred cubic arrangement of spheres and a bicontinuous cubic phase with *la3d* symmetry (the gyroid structure) [14]. It should also be emphasised, that 'minority phases' or combination phases can be observed in the regions on a phase boundary between structural or morphological arrangements [14]. This is particularly evident for thin films of BCPs where interface effects are strong. An example is the perforated lamellar structure where a one of the blocks in a lamellar structure can support cylinders of another block in the surface region [15]. These phases cannot be ignored because complex 3D morphologies can be challenging for both analysis and eventual pattern transfer to a substrate [16].

For simplicity, and to mirror the work carried out to demonstrate applicability to nanocircuitry fabrication, we will confine our discussion to simple diblock copolymers. It should be noted that more complicated BCPs such as triblocks might have significant relevance because their structural complexity may have future importance as 'smart' masks [17]. For application as an alternative and complimentary to conventional photolithography (i.e. block copolymer lithography), two of the structural phases described above have become all important. The first is the lamellar structure where the individual block domains can form well-defined stripes (Fig. 1A). The second is the hexagonal arrangement of cylinders where one block forms cylinders within a matrix of the other block (Fig. 1B). Their dominance reflects their potential use to create device, interconnect and via/hole structures (i.e. line and dot patterns).

Despite the obvious practicality of these two arrangements, considerable thought must be given to how these systems interact

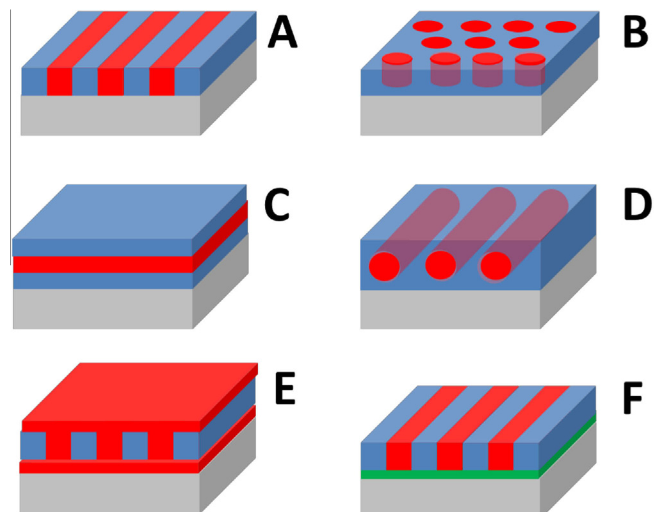

Fig. 1. Microphase separated block copolymer patterns with potential to be used in nanocircuitry fabrication. See text for details.

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