

Microwave-Assisted Multicomponent Organic Syntheses: A Green Revolution in Synthetic Strategy

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Introduction

In recent decades, the increasing awareness of environmental sustainability and the urgent need to mitigate chemical waste have catalyzed the evolution of synthetic organic chemistry. Among the many innovative strategies aligned with the principles of Green Chemistry, microwave-assisted organic synthesis (MAOS) has emerged as a powerful tool that combines energy efficiency, accelerated reaction rates, and high product yields with operational simplicity. When merged with multicomponent reactions (MCRs)—which integrate three or more reactants in a single synthetic operation to form complex molecules—the result is a highly convergent, atom-economical approach that minimizes waste, reduces solvent use, and enhances selectivity.

The synergy between microwave technology and multicomponent reactions is particularly impactful in the synthesis of heterocycles, natural product analogs, and pharmaceutically relevant scaffolds. This approach not only aligns with the twelve principles of Green Chemistry but also provides chemists with a sustainable pathway to complex molecular architectures with operational ease and enhanced functional group tolerance.

This review presents a comprehensive account of microwave-assisted multicomponent reactions (MW-MCRs) with a focus on their green and sustainable attributes. It begins with an overview of the principles of Green Chemistry and the fundamentals of microwave heating. This is followed by a detailed discussion of various reaction types—solution-phase, solvent-free, and solid-supported—highlighting the role of reaction media, reactor design, and catalytic systems. Key synthetic examples and mechanistic insights are included to illustrate the efficiency and versatility of this technique. The review also emphasizes recent advancements, challenges, and future prospects in integrating microwave technology into greener synthetic protocols.

Green Chemistry

Eco-friendly synthetic strategies come under the domain of *Green Chemistry* which is a chemical philosophy encouraging the design of products and processes that reduce or eliminate the use and generation of hazardous substances. Whereas environmental chemistry is the chemistry of the natural environment, and of pollutant chemicals in nature, Green Chemistry seeks to reduce and prevent pollution at its source.

The Twelve Principles of Green Chemistry

1. It is better to prevent waste than to treat or clean up waste after it has been created.
2. Synthetic methods should be designed to maximize the incorporation of all materials used in the process into the final product.
3. Wherever practicable, synthetic methods should be designed to use and generate substances that possess little or no toxicity to human health and the environment.
4. Chemical products should be designed to effect desired function while minimizing their toxicity.

5. The use of auxiliary substances (e.g., solvents, separation agents, etc.) should be made unnecessary wherever possible and innocuous when used.
6. Energy requirements of chemical processes should be recognized for their environmental and economic impacts and should be minimized. If possible, synthetic methods should be conducted at ambient temperature and pressure.
7. A raw material or feedstock should be renewable rather than depleting whenever technically and economically practicable.
8. Unnecessary derivatization (use of blocking groups, protection/ deprotection, temporary modification of physical/chemical processes) should be minimized or avoided if possible, because such steps require additional reagents and can generate waste.
9. Catalytic reagents (as selective as possible) are superior to stoichiometric reagents.
10. Chemical products should be designed so that at the end of their function they break down into innocuous degradation products and do not persist in the environment.
11. Analytical methodologies need to be further developed to allow for real-time, in-process monitoring and control prior to the formation of hazardous substances.
12. Substances and the form of a substance used in a chemical process should be chosen to minimize the potential for chemical accidents, including releases, explosions, and fires.

MICROWAVE: AN ALTERNATE ENERGY SOURCE

Introduction

Microwave activation as a non-conventional energy source has become a popular and useful technology in organic synthesis¹. With the increase in environmental consciousness throughout the world, the non-conventional mode of heating has gained precedence and is widely accepted by the scientific community. People have recognised other potential applications for this mode of heating and scientists engaged in a number of disciplines have applied the rapid heating associated with microwave technology to a number of useful processes. These include the preparation of samples for analysis², application to waste treatment³, polymer technology⁴, drug release/targeting⁵, ceramics⁶ and alkane decomposition.⁷ This has also found use in a range of decomposition processes including hydrolysis of proteins and peptides.⁸

The growing concern about the impact of organic solvents and chemical wastes on the environment, the non-traditional synthetic approaches are attracting much attention that may reduce the pollution at source.⁹ In this context, Microwave Induced Organic Reaction Enhancement (MORE) chemistry has become a widely accepted technique as it restricts the solvent requirement, increases 'atom economy'¹⁰ by improving product selectivity and chemical yield.

The microwave region of the electromagnetic spectrum lies between infrared radiation and radio waves which is characterized by wavelengths between 1 mm to 1 m, corresponding to frequencies between 100 and 5000 MHz (0.3-300 GHz). Telecommunication and microwave radar equipment occupy many of the band frequencies in this region. In order to avoid interference, the wavelength at which industrial and domestic microwave apparatus intended for heating operates at 12.2 cm, corresponding to a frequency of 2.450 GHz.¹¹

Mechanism of Microwave Heating

Microwave radiations, like all electromagnetic radiations, also have both an electric field component and a magnetic field component. The former component is responsible for the dielectric heating, which is effected *via* two major mechanisms (i) Dipolar polarisation mechanism and (ii) Conduction mechanism.¹²

Dipolar Polarisation Mechanism

In this, interaction of electric field component of microwave with the molecule/matrix occurs. To generate heat when irradiated with microwaves, it must possess a dipole moment. A dipole is sensitive to external electric field and attempts to align itself with the field by rotation. The applied field provides the

energy for this rotation. In the microwave radiation region, the frequency of the applied irradiation is low enough so that the dipoles have time to respond to the alternating electric field and therefore rotate. The frequency is, however, not high enough for the rotation to follow the field. Thus, as the dipole re-orientates to align itself with the electric field, the field is already changing and generates a phase difference between the orientation of the field and that of the dipole. This phase difference causes energy to be lost from the dipole by molecular friction and collisions, giving rise to dielectric heating.

Conduction Mechanism

- A solution containing ions, or even a single isolated ion with a hydrogen bonded cluster, in the sample of ions will move through the solution under the influence of an electric field, resulting in expenditure of energy due to an increased collision rate, converting the kinetic energy to heat.
- The conductivity mechanism is much stronger interaction than the dipolar mechanism with regard to the heat generating capacity.
- To compare the abilities of different solvents to generate heat from microwave irradiation, their capabilities to absorb microwave energy and to convert the absorbed energy into heat is calculated using the following equation:

$$\tan \delta = \frac{\epsilon''}{\epsilon'}$$

δ = Loss angle or dissipation factor.

ϵ' = Dielectric constant or relative permittivity, represents the ability of a dielectric material to store electric potential energy under the influence of an electric field and polarised by an electric field.

ϵ'' = Dielectric loss which measures the efficiency with which the absorbed microwave energy is converted into heat.

So, the source of microwave dielectric heating lies in the ability of an electric field to polarise charges in a material and the inability of this polarisation to follow rapid reversal of the electric field. The total polarisation is the sum of a number of individual components.

$$\epsilon_t = \epsilon_e + \epsilon_a + \epsilon_d + \epsilon_i$$

ϵ_e = Electronic polarisation arising from realignment of electrons around nuclei.

ϵ_a = Atomic polarisation arising from relative displacement of nuclei due to unequal distribution of charge within the molecule.

ϵ_d = Dipolar polarisation resulting from the orientation of permanent dipoles by electric field.

ϵ_i = Interfacial polarisation resulting from buildup of charges at interfaces or Maxwell-Wagner effect which occurs when there is building up of charges at interfaces.

The time scale for polarisation and depolarisation of ϵ_a and ϵ_e is much faster than the microwave, therefore, no contribution to dielectric heating effect.

Experimental Considerations

In a microwave oven, the radiation is generated by magnetron, the microwaves are guided into the cavity by the waveguide and reflected by the walls of the cavity. If microwaves are not absorbed, they may be reflected back to the waveguide and damage the magnetron, thus it is essential to have a microwave active dummy load which will absorb excess of microwaves and avoid such damage.

Reaction Vessels

The reaction vessels made up of glass, teflon, polystyrene, ceramic, and quartz are suitable for carrying out microwave induced reactions as these materials are transparent to microwaves.

Most of the reported reactions have been carried out either in sealed vessels or in the solid state. However, if the reactions are to be carried out under pressure in sealed systems, the major concern is the ability of the vessel to withstand changes in temperature and pressure which is associated with the particular chemical transformation. Reactions have been conducted in sealed teflon vessels which can retain pressure up to 50 atm. This technique does not allow great deal of control over the reaction and suffers from following disadvantages:

- (i) The teflon vessels are not designed for high boiling solvents ($>200^{\circ}\text{C}$) and viscous solutions.
- (ii) It is not possible to monitor the temperature and pressure in the teflon autoclave.
- (iii) The porosity of teflon means that the reaction liners can be contaminated with absorbed impurities.
- (iv) There are no means of viewing reaction mixture during the progress of reaction.

The above mentioned, disadvantages are overcome by the development of a Fischer-Porter reactor¹³ made up of pyrex. Fischer-Porter reactor vessel can be admitted through the roof of a microwave oven using suitably designed port. It contains pressure measurement and control facilities that allows a visual examination of the reaction. Thus, it provides a convenient and an inexpensive method for performing superheating experiments resulting in large reduction in reaction time for inorganic and organometallic synthesis.

Types of Reactions

(a) Non-Solid-State Reactions

They are microwave accelerated solution phase reactions. In these types of reactions, reagents are dissolved in an organic solvent and subjected to microwave irradiation (MWI). For this, at least one of the substrates should be microwave active i.e. polar.

Choice of Solvent

It is customary to conduct the reaction under reflux to control the temperature of the reaction in an organic solvent taken as a reaction medium. For microwave induced reactions the solvent of choice is one which absorbs microwave energy efficiently thus gets heated rapidly under microwave and has a boiling point at least $20\text{--}30^{\circ}\text{C}$ higher than the desired reaction temperature. With MWI, solvents can be heated above their boiling points and this superheating leads to reaction rate enhancements.¹⁴

For microwave induced organic reactions, any solvent which has high dielectric constant and high boiling point is an excellent energy transfer medium. For example, using *N,N*-Dimethylformamide (DMF) (ϵ 3.67, b.p. 160°C), the reaction temperature rises to about 140°C without much vapourisation of the solvent. DMF retains the water formed in the reaction, thus obviating the requirement of Dean-Stark apparatus. Other commonly used solvents are water, ethylacetate, acetone, acetic acid, methanol, dichloromethane, etc.

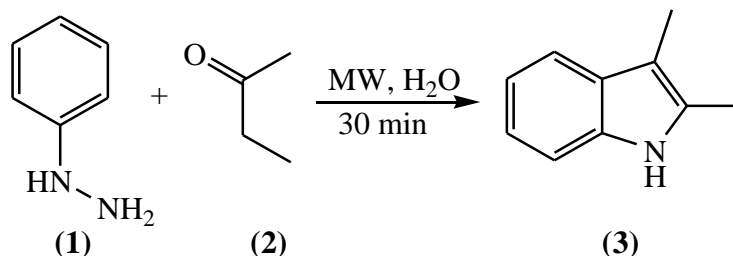
Hydrocarbon solvents like benzene, toluene, and xylene are not suitable for microwave assisted organic synthesis as they absorb microwaves poorly. The temperatures of 50 ml of solvents after heating for 1 min at 560 W is as follows:

Solvent	Temp. $^{\circ}\text{C}$	b.p. $^{\circ}\text{C}$
Water	81	100
Diethyl ether	32	35
Acetone	56	56
Chloroform	49	61
Methanol	65	65
Hexane	25	68
Carbon tetrachloride	28	77
Ethyl acetate	73	77
1-Chlorobutane	36	78
Ethanol	78	78
1-Propanol	97	97
Heptane	96	98
1-Bromobutane	95	101
1-Butanol	109	117
Acetic acid	110	119
1-Pentanol	106	137
<i>N,N</i> -Dimethyl formamide	131	153

Use of water as a green solvent

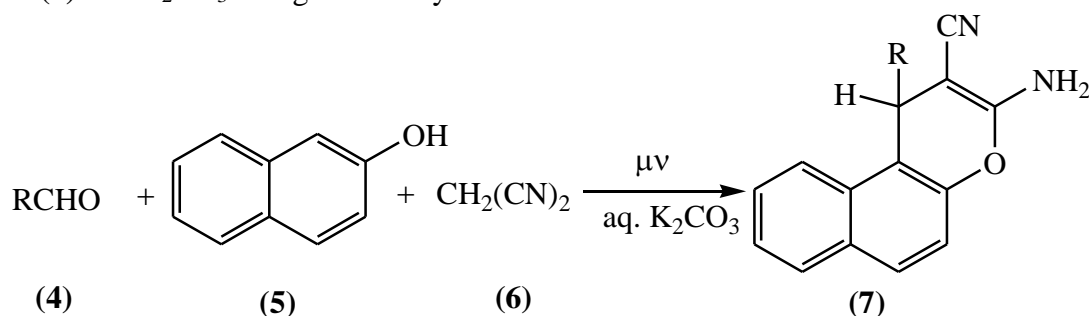
Although best solvent is 'no solvent' but if solvent is required water has a lot to recommend it as it is readily available, non-toxic and neither combustible nor explosive. Recently water¹⁵ has been reported as an alternate media in various transformation reactions with a considerable rate increase.¹⁶ This has been explained on the basis of its permanent dipole moment; due to which at elevated temperature, water facilitates the isolation of products due to the decreased solubility of organic materials. Other reasons to make water unique in comparison to organic solvents are its cheapness, non-inflammability and high dielectric constant properties.

The Fischer indole synthesis of 2,3-dimethylindole (**3**) was accomplished in water from phenylhydrazine (**1**) and butan-2-one (**2**) at 220 °C within 30 min thus circumventing the use of preformed hydrazone or any acid.¹⁷

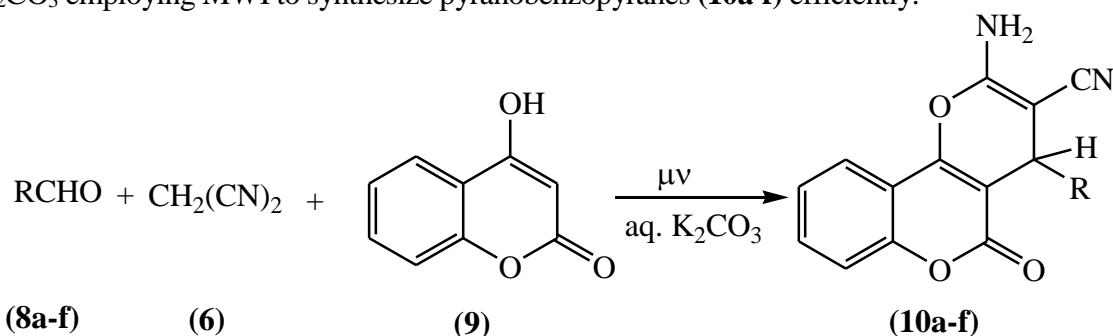


The formation of inorganic salts accounts for the major waste generation in the production of various pharmaceuticals and fine chemicals in view of the frequent neutralization of basic or acidic solutions. The microwave (MW) protocols using high-temperature water as the reaction medium are especially attractive in this context because of the selectivity and less usage of the acidic or basic agent that eventually reduces the requirement for neutralization.¹⁸

Recently, substituted-chromenes (**7**) were synthesized using β -naphthol (**5**), various aldehydes (**4**), malononitrile (**6**) and K_2CO_3 as a green catalyst in water under MWI.¹⁹



The three-component reaction of aldehydes (**8a-f**), malononitrile (**6**) and 4-hydroxy coumarin (**9**) was performed in aqueous K_2CO_3 employing MWI to synthesize pyranobenzopyranes (**10a-f**) efficiently.²⁰



R = phenyl, 4-chlorophenyl, 2-furyl, 3-indolyl, piperonyl, 2-chloro-3-quinolyl

(b) Solid State Reactions

The requirement of teflon vessels, specialised sealed vessels and the development of high pressure are some of the limitations of MW assisted solution phase reactions which have been circumvented in dry media solid state reactions.

Three types of solvent free procedures or dry media reactions can be coupled with microwave irradiations. These are as follows:

(i) Solid Supported Reactions

Heterogeneous reactions facilitated by supported reagents on inorganic oxide surfaces have received considerable attention both industrially as well as for laboratory purposes. These reactions are effected by the reagents immobilised on the porous solid supports and have advantages over conventional solution phase reactions because of the good dispersion of active reagent sites, associated selectivity and easier work-up. The recyclability of some of these solid supports (alumina, clay, silica, bentonite, zeolite, montmorillonite, anhydrous K_2CO_3 etc.) renders these processes into truly ecofriendly protocols. Reactants which are individually impregnated onto the solid support (saturated solution of reactants in an adequate solvent if they are solids with subsequent removal of solvent) undergo facile reaction with microwaves to provide high yields of pure products.²¹⁻²⁶

Aluminas, silicas, clays or zeolites can be selected as acidic or basic supports depending on the type of organic reaction.^{27,28}

Alumina alone can possibly act as a base towards acidic molecule but if a strong base is necessary, KF on alumina²⁹ can ionise carbon acids upto $pK_a = 35$.³⁰ On the other hand, montmorillonite³¹ clay like K10 or KSF offer acidities very close to nitric or sulphuric acids.

Alumina as solid support

Inorganic oxides (alumina, silica) do not absorb microwaves and so are not an obstacle for the transmission of microwaves.^{32,33} In dry media reactions, hydroxyl groups, water and organic compounds present on the surface of these inorganic oxides strongly absorb microwaves and these species are activated to microwaves. The thermal diffusion of inorganic oxides which are a barrier for thermal activation of reaction whereas microwaves are easily transmitted.³⁴

Studies of thermal behaviour of alumina (Figure I and II) indicate that temperatures reached depend on the quantity of irradiated solid. A maximum, connected with the optimal capacity of the oven, is obtained for about 200 g of alumina. It is to be noticed that a minimum amount of 4 g is required to observe any thermal effect. This observation confirms that the dimensions of irradiated samples (height and diameter) must be larger than $\square/10$ for the 2450 MHz domestic microwave oven with $\square = 12.2$ cm.

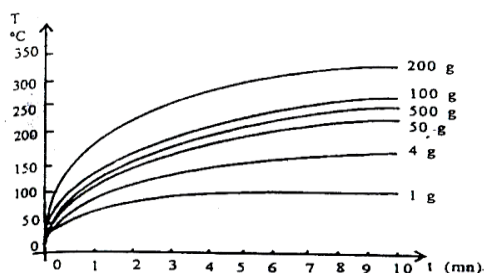


Figure I

Thermal behaviour of alumina as a function of the irradiation time and of alumina amount ($P = 600 \text{ W}$)

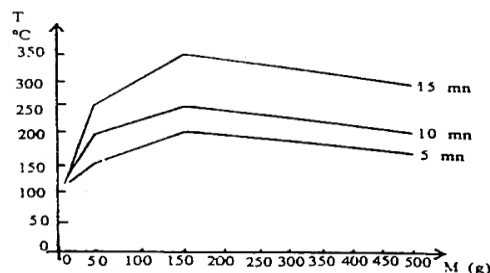


Figure II

Thermal behaviour of various amounts of alumina for different irradiation time ($P = 600 \text{ W}$)

It is observed that 1 g alumina sample cannot reach more than 100°C (Figure I) even with larger irradiation times (20 min). In these conditions, considerable energy losses cause overheating of the inner walls of the oven and of the pyrex vessels. This could lead to irreversible damage of the magnetron.

When only small amount of alumina is used, it is proposed to use an external bath³⁵ of about 150-200 g of alumina (Figure II) which allows fast heating of the reaction mixture, placed at its centre in an individual pyrex vessel, the reaction can then occur at temperatures lower than the boiling points of the products.

Montmorillonite as solid support

Clays are solid acidic catalysts, which can function as both Bronsted, and Lewis acids in their natural and ion exchanged forms. Modified smectite clays can be very sensitive catalysts for a wide range of organic transformations.^{36,37}

Clays are secondary minerals that have formed through the weathering and hydrous alteration of certain parent rocks and their constitution minerals. Silica and alumina are necessary precursors to the formation of clay minerals. Clay formed at the site of the parent rock are called primary clays even though they are secondary in origin after other minerals. Those formed after transformation and redeposition of the rock and mineral fragments or transported as clays from the origin site of origin are called secondary clays. The major clay sources are: kaolinite, halloysite, montmorillonite (bentonites), illite, chlorite, attapulgite and allophone. Bentonites structures are of two important varieties swelling and non-swelling bentonites. Swelling bentonites contains sodium in the platy molecular structure and has strong swelling properties and posses a high drying-bonding strength while non-swelling bentonites contain calcium in its structure and has lower capacity to swell.

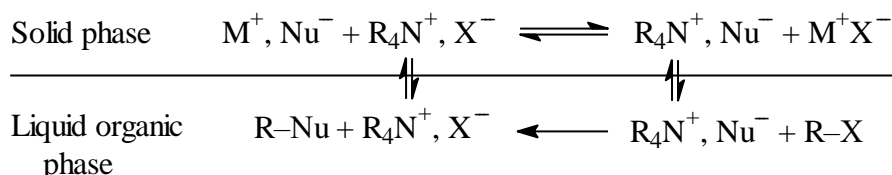
Montmorillonite is a subgroup of the smectite (swelling) clay and is main constitution of bentonites and fuller's earth. It is a common clay mineral with layered structured and a range of permanent charge, which cations and water molecules to enter the space between clay layers, also known as the interlayer. Montmorillonite has high degree of efficiency of metal cation exchange and the acidic and catalytic properties of montmorillonite can be easily altered by replacing interlayered cation in them montmorillonite without any change of the crystalline structure to suit the needs of synthetic organic reaction.³⁸ Chemically modified clays are considered to be potentially the most cost-effective nanomaterials, a variety of organic reactions have been carried out with great success using such clays as catalysts.

The Lewis acidity of clay is due to Al^{3+} and Fe^{3+} at the crystal edges and can be modified by the exchange of the interlayer Na^+ and Ca^{2+} cations. The Bronsted acid character of the clays arises mainly due to the dissociation of the intercalated water molecules coordinated to cations. Solid-acid catalysts are generally categorized by their Bronsted and Lewis acidity, the strength and number of these sites, and the morphology of the support (e.g. surface area, pore size). Layered clays have been extensively used as acid catalysts and as supports for oxidations (clayfen and claycop)³⁹ and Lewis acids (clayzic).⁴⁰ Hydrogen cyanide has been proposed as a probe of the Bronsted and Lewis nature of the acid sites in metal-oxide pillared clays.⁴¹

Montmorillonite K-10 is a type of acidic stratified silicate mineral with a three-layered structure with an ideal chemical formula of $(\text{Al}_{2-y}\text{Mg}_y)(\text{Si}_4\text{O}_{10})(\text{OH})_2 \cdot n\text{H}_2\text{O}$. In montmorillonite clay one octahedral aluminate layer is sandwiched between two octahedral silicate layers.⁴² The interlayer cations are exchangeable, which allows alteration of the acidic nature of the material by a simple ion-exchange procedure. They have been used in number of oxidation,⁴³ reduction, cyclocondensation for the synthesis of heterocyclic compounds⁴⁴ and many other reactions⁴⁵ as catalyst and also as solid support for reagents.⁴⁶

(2) Solid Liquid Solvent – Free Phase Transfer Catalysis (PTC)

This is specific for anionic reactions as it involves ionic activation.⁴⁷ A catalytic amount of tetralkylammonium salts or a cation complexing agent is added to the mixture of both pure reactants. Reactions occur in the liquid organic phase which consists only of the electrophile R-X . The presence of a solvent is prejudicial as it increases dilution of reactants and consequently, a decrease in activity. The electrophile, RX is therefore, both the reactant and the organic phase for the reaction.



(3) Neat Reaction

In this, neat reactants are subjected to MWI in the absence of solvent, solid support and catalyst. There has not been much advancement in this area as direct heating of reactants in the absence of solvent often leads to charring. But these no solvent reactions prove to be advantageous for environmental reasons and offer the benefits of shorter reaction times especially when coupled with microwaves or ultrasound.⁴⁸⁻⁵⁰

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