MULTI-SCALE SIMULATIONS OF STIRRED LIQUID–LIQUID DISPERSIONS

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Abstract: Direct numerical simulations (DNS) of liquid–liquid dispersions in a three-dimensional periodic domain have been performed by means of the lattice–Boltzmann method. The dispersion was agitated by a random force field such as to produce isotropic turbulence. The turbulence levels and its history were based on a fluid parcel in a turbulently stirred tank traversing the impeller swept region. The DNS provide detailed insight in the coalescence and break-up events brought about by the turbulent flow, the evolution of the drop size distribution in time, and the interaction of the small-scale flow and the droplets as witnessed in terms of the spectral characteristics of the turbulence.

Keywords: liquid–liquid dispersion; break-up; coalescence; turbulence; lattice–boltzmann; numerical simulation.

INTRODUCTION

Formation of droplets in laminar and turbulent flow by breaking up a continuous phase liquid occurs in many engineered and natural systems. If we limit ourselves to man-made systems, examples are water-in-oil emulsions in food production, formation of monomer droplets by agitation to start an emulsion polymerization process, droplets in extraction columns. The subject has received extensive attention from experimentalists and theoreticians over the years. Classical results in the field are due to Rayleigh (stability and break-up of jets), and Kolmogorov and Hinze (drops in turbulent flows) (Kolmogorov, 1946; Hinze, 1955). Dependent on the nature of the flow, the problem of interfacial dynamics and drop formation has been approached differently. In laminar flow, clean experiments in e.g., Couette devices (De Bruijn, 1993) or four-roll mills (Bentley and Leal, 1986) reveal the stretching of drops and their breakage in great detail. Also coalescence (the inverse of break-up) has been studied in simple sheared systems experimentally. Regime maps with many degrees of freedom (interfacial tension, density and viscosity ratios, shear rates) have been determined experimentally.

In turbulent flow of immiscible liquids the formation of droplets is directly related to the dynamics of the flow. Break-up and coalescence occur simultaneously since the flow is able to rupture interfaces, and at the same time stimulates drop–drop collisions leading to coalescence. Therefore, the size distribution of a population of droplets in a turbulent flow is a continuously changing function. Its dynamics (including the quasi steady-state shape of the drop size distribution) depend on the characteristics of the turbulence as expressed in the distribution of the turbulence activity over various length scales (the turbulence spectrum), and on the properties of the liquid-liquid system (viscosities, densities and surface tension). Experiments in dense (10% disperse phase and up) turbulent suspensions are difficult because of limited optical accessibility and small time-scales preventing an approach as detailed as in laminar flow. In this area, experimental research focuses on the resulting drop size distributions (DSDs), rather than on the details of the break-up and coalescence events. For instance DSDs as a function of the parameters that determine the strength of the turbulence (like the speed of an impeller in a mixing tank), and the fluid properties have been measured with intrusive (Ritter and Kraume, 2000) and non-intrusive (Colenbrander, 2000) techniques. From such experiments important practical correlations can be derived, e.g., relating the mean drop diameter with the power inserted in the system. Such correlations have been used for many decades in the design and scale-up of industrial processes involving liquid–liquid dispersions.

Largely due to computational and experimental limitations, an extensive part of fundamental research on turbulent liquid–liquid dispersions focuses on dilute suspensions. Dilute suspensions better allow for (non-intrusive) optical access, and are tractable with Eulerian–Lagrangian type of numerical

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simulations that keep track of DSDs in turbulent flow where break-up phenomena are coupled to local turbulent conditions, and coalescence hardly occurs. Practical (industrial) flow systems, however, are operated at high loadings (up to phase inversion). The major practical questions focus around the relation between the turbulent flow and the DSD. For instance, in some specific industrial process it was found that using static mixers instead of agitated mixing tanks greatly reduced the amount of very fine droplets. This favoured the use of static mixers since it made the subsequent separation step much easier. Furthermore, the flow physics at high loadings has many interesting and challenging aspects, like how the small-scale turbulence interacts with the interfaces. In this respect one could anticipate an inverse cascade due to turbulent kinetic energy inserted at the small (drop) scale due to relative motion of drops and continuous phase liquid.

In this paper, we propose a numerical, multi-scale approach to turbulently agitated liquid–liquid dispersions. Starting point is the direct numerical simulation (DNS) of a small-scale system containing a dispersed system of two immiscible liquids. This system we agitate as if it was a fluid package travelling through a stirred tank. It experiences bursts of turbulence when it traverses the impeller stream and/or the impeller-swept region. It also spends significant amounts of time in the more quiescent parts of the tank. The turbulence results in droplet motion and deformation leading to break-up and coalescence and a continuously changing DSD. This approach forms an alternative to population balance modelling (see e.g., Ramkrishna, 2000), where the emphasis is strongly on the parameterization of the micro-scale phenomena (in terms of kernel functions relating break-up and coalescence to local flow and turbulence conditions). Since especially in dense dispersions the microscopic phenomena are very complex, we believe that a more detailed approach to it (via DNS) could add to the quality of predictions.

We aim at a detailed description by means of DNS of the phenomena occurring in a mixture of two immiscible liquids in turbulent motion. The disperse phase volume fraction is chosen to be significant (15% and up). We limit ourselves to systems without walls in which homogeneous isotropic turbulence is generated by randomly forcing a three-dimensional periodic domain (Alvelius, 1999). This periodic box contains the two fluids that (for reasons of simplicity) have equal density and viscosity. As a pair, the fluids have a surface tension. The box is agitated in such a way that the turbulence intensity mimics the passage of a fluid package through the impeller stream in a lab-scale stirred tank. Our goal is to reveal the dynamics of the two-fluid mixture, including coalescence and break-up, and (eventually) to relate the turbulence properties and the drop size distribution (DSD). We use the lattice–Boltzmann method to solve the velocity field and an index function field that contains the thermodynamics of the fluid-fluid interaction. More specifically, we use the scheme proposed by He, Zhang and co workers. (He *et al.*,1999a, b; Zhang *et al.*, 2000).

NUMERICAL SETUP

In the lattice–Boltzmann method, fluid flow is mimicked by a system of fictitious particles residing on a lattice and having discrete velocities such that the particles travel to neighbouring lattice sites during one time step. At the lattice sites, particles coming from the various lattice directions interact (collide). With a single set of particles and the proper lattice topology and collision rules the system recovers the incompressible, single-phase Navier–Stokes equation (Chen and Doolen, 1998). In the lattice–Boltzmann method there are various ways to introduce a second fluid. The procedure we use is based on a pressure distribution function and an index function (or order parameter) that tracks the interfaces between the two fluids (He *et al.*, 1999a, b; Zhang *et al.*, 2000). These two distribution functions now involve two sets of lattice–Boltzmann particles. The immiscibility of the fluids is brought in by defining a free energy function that (among other things) is a function of the order parameter. This function has two minima, giving rise to an equilibrium state where Fluid 1 is preferred, and a state where Fluid 2 is preferred. The free energy also is a function of the spatial gradients of the order parameter. This way we incorporate molecular interactions that mimic surface tension. Our flow domain is a fully periodic, three-dimensional cube without any walls. In this cube we generate homogeneous, isotropic turbulence with well-defined properties through random forcing (Alvelius, 1999; Ten Cate *et al.*, 2006).

Relation with Stirred Tank Flow

The turbulence and fluid properties in the periodic cubic domain are loosely based on the experiments by Schulze *et al.* (2000). They measured DSDs of toluene–water mixtures in baffled tanks of various sizes agitated by Rushton turbines. We specifically will focus on their experiments in a $T = 150$ mm diameter tank operated at a tank-average specific power input of 510 W m^{-3} (in this case the impeller speed approximately is $N = 10$ rev s^{-1}). Results of these experiments have been summarized in figure 7 of the paper by Schulze *et al.* (2000).

In order to assess the levels of turbulence a fluid package would encounter in this tank, we performed a single-phase large-eddy simulation (LES) of the flow and tracked fluid packages through it. The LES was done in a manner similar to that presented by Derksen and Van den Akker (1999), and Derksen (2003), including the use of the Smagorinsky subgrid-scale model. In Figure 1 we show a part of the history of the energy dissipation rate of a fluid package (please note the logarithmic ε-scale). Typically once every 10–20 impeller revolutions the fluid package traverses the impeller swept volume and/or the impeller stream leading to a burst of turbulence with dissipation peak levels of approximately $\varepsilon = 10\,D^2N^3$ (being about 40 times the tank-average value). It is expected that these ε-peaks are decisive in

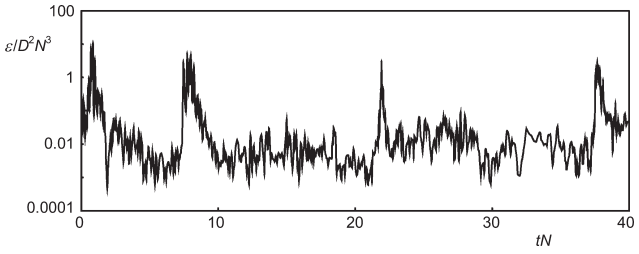

Figure 1. LES result: time history of a fluid package traveling through a Rushton stirred tank operated at $Re = 30\,000$ in terms of the energy dissipation rate ε (scaled with impeller diameter D, and impeller speed N in rev s^{-1}).

Figure 2. Demixing simulation. Cross sections through a fully periodic 128^3 domain. Time proceeds from left to right, the time interval between subsequent images being 500 time steps.

the drop formation process. The Kolmogorov length and time scale related to the dissipation peak levels are $\eta_K = 15\ \mu m$ and $\tau_K = 0.23$ ms, respectively. The latter values we use to specify the turbulent conditions of the DNS in the periodic domain containing the liquid–liquid mixture.

Definition and Scaling of the DNS Cases

In the liquid–liquid DNS we have a cubic grid with linear cell size Δ. In the turbulence cases that will be discussed here, our three-dimensional periodic domain consists of 256^3 cells. When randomly forcing turbulence in periodic domains, we introduce energy at low wavenumbers, i.e., large scales (Alvelius, 1999). The energy cascade then takes care of the development of smaller scales. The random forcing is fully specified by the spectrum of forcing, i.e., the distribution of forcing amplitudes over wavenumber space, and by the total power input into the system. In steady state, the power inserted will be completely dissipated and therefore the time- and volume averaged dissipation rate in this homogeneous system equals the inserted power.

The inserted power and the viscosity (both fluids have the same viscosity, see below) were specified in such a way that the Kolmogorov scale is $\eta_K = 1.8\,\Delta$. The RMS velocity follows from $u' = \sqrt[3]{P/k_f}$ with P the inserted power per unit mass, and k_f the main (central) forcing wavenumber.

The method we use allows for fluids with different density and viscosity. For reasons of simplicity, however, we have chosen to give the two fluids the same viscosity and density. In this sense we depart from the situation discussed by Schulze *et al.* (2000) since toluene is less viscous than water, and also has a density slightly lower than that of water. The interfacial tension between the two fluids at room temperature is $\sigma = 0.036\ N\ m^{-1}$. We use the Weber number based on velocity fluctuation levels ($We = (\rho u'^2 d/\sigma)$, with d the drop diameter) to relate the surface tension in the experiment (Schulze *et al.*, 2000) to the surface tension in the DNS. In the impeller stream, turbulent kinetic energy levels typically are $0.18\ v_{tip}^2$ (Derksen and Van den Akker, 1999) from which value we can estimate u'. With a typical drop size of $d = 250\ \mu m$ (Schulze *et al.*, 2000) $We \approx 1.6$.

In the turbulent liquid–liquid DNS the disperse phase volume fraction has been fixed to 16%, which is a value that is well within the range investigated by Schulze *et al.* (2000).

RESULTS

Demixing

In order to qualitatively demonstrate the thermodynamic behaviour of the liquid–liquid system in the lattice–Boltzmann context, we first consider its demixing properties without additional turbulent fluid flow. This simulation was done in a periodic box with size 128^3. We start with a random mixture of the two fluids (i.e., on any computational node we made a random choice between the white and the black liquid, each having a 50% chance). In the series of pictures shown in Figure 2, one sees the separation of the two fluids in their tendency to minimize surface energy.

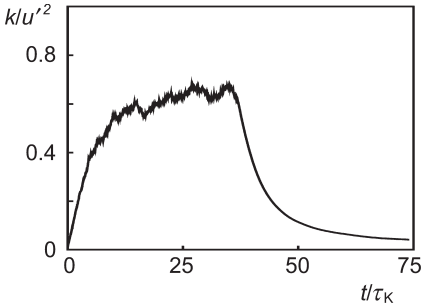

Figure 3. Time series of the turbulent kinetic energy in the periodic domain.

Droplet Dynamics

The direct simulation of a turbulent dispersion starts with a zero velocity field, and a set of spherical droplets having an initial diameter of $d_0 = 20\,\Delta$ (Δ being the lattice spacing; in physical units $d_0 = 180\ \mu m$). At $t = 0$ we switch on the random forcing to generate turbulence. It then takes some time for the turbulence to develop. After some time we switch off the forcing and turbulence dies out. This way we mimic what a fluid package in a stirred tank experiences when it approaches, traverses and leaves the impeller region (see Figure 1). The width of the peaks in Figure 1 typically is 1/10N. With $N = 10\ rev\ s^{-1}$ and the Kolmogorov time scale being $\tau_K = 0.23$ ms, the peak width is some 50 τ_K. The history of the turbulent kinetic energy in the periodic box simulation has been displayed in Figure 3. The settings were chosen such that the fluid experiences a rapidly increasing turbulent activity, a quasi steady situation with intense levels of turbulence during approximately 30 τ_K, followed by a rapid decay.

The dispersed phase volume fraction was 16%. With a domain size of 256^3, $d_0 = 20\,\Delta$, and a disperse phase volume fraction of 16% this implies that we start with 640 droplets. In Figure 4 we show how the liquid/liquid system responds to this turbulence history. It should be reminded that the figure shows a cross section through a three-dimensional field; the drops in the first panel all have approximately the same size. The plane of view, however,

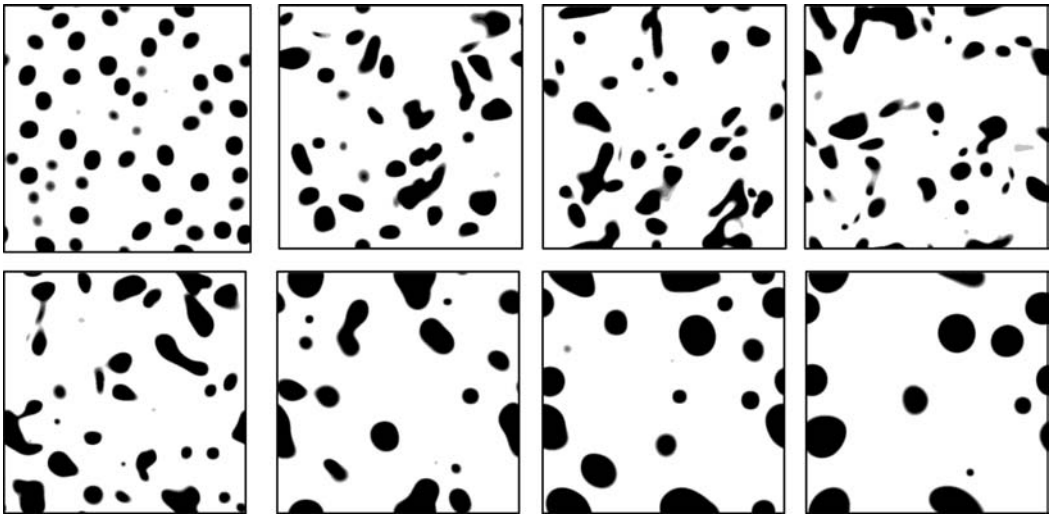

Figure 4. Cross sections through a fully periodic 256^3 domain. Time proceeds from left to right, and then from top to bottom; the time interval between subsequent images is 9 τ_K.

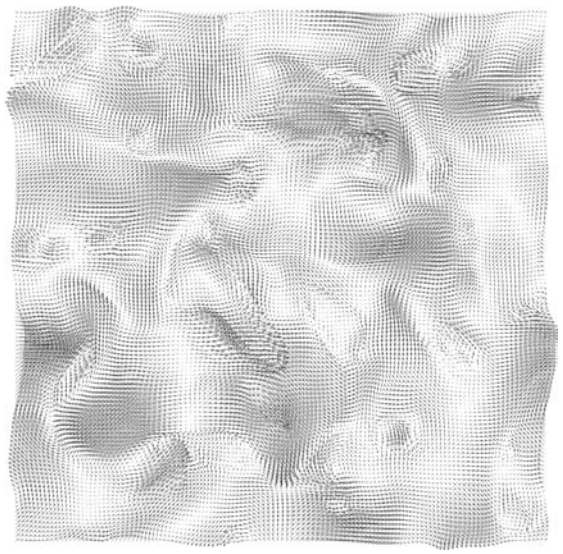

Figure 5. Typical cross-section through an instantaneous velocity field.

cuts each one differently. In Figure 5 we show a velocity field that corresponds to the fourth panel in Figure 4. The sequence in Figure 4 shows the deformation of droplets due to fluid flow, and the formation of big (due to coalescence) and small (break-up) drops. The net effect, however, seems to be an increase in the average drop size. This will be further verified below.

In analysing the simulation results, the emphasis will be on the DSD. As can be seen in Figure 4, drops are in general not spherical. A drop is defined as a connected piece of black liquid, the volume of which (V) can be readily determined. Drop sizes are expressed in terms of their volume-equivalent diameter $d = \sqrt[3]{6V/\pi}$. The series of DSD's in Figure 6 shows that the turbulence widens the DSD (that started at $t = 0$ as a delta-function at $d = d_0$). Gradually more and more very small drops are formed. Once the turbulent forcing has been switched off (at $t = 37\ \tau_K$), coalescence dominates the evolution of the DSD: it shifts to the right.

The Sauter mean diameter d_{32} as a function of time (Figure 7) can be well interpreted in terms of the history of the turbulence (Figure 3): in the initial stage (rising turbulence) the net effect of coalescence and break-up brought about by the turbulence is apparently an increase of d_{32}. The system tends to a steady state when $t \approx 30\ \tau_K$. Then the forcing is switched off and turbulence quickly dies out. In the dying-out phase coalescence dominates over break-up and the d_{32} monotonically increases. It levels off at the end of the time series.

Turbulence Spectra

The relation between drops and turbulence has a two-way character with not only turbulence acting on the DSD but also drops influencing the turbulence spectrum. To demonstrate the two-way coupling, we compare in Figure 8 a turbulence spectrum of a single-phase system and a liquid–liquid dispersion obtained on the same grid and with the same agitation. The latter shows enhanced turbulence levels at small scales (high wave-numbers). This is due to flow being generated at the small scales by the motion of the interfaces. Ten Cate *et al.* (2004) observed similar effects when performing DNS of liquid–solids suspensions.

SUMMARY AND OUTLOOK

In this paper we have shown first results of a novel and fundamental approach to turbulent liquid–liquid dispersions. By means of direct numerical simulations based on the lattice–Boltzmann method we solve the dynamics of a liquid–liquid dispersion in a fully periodic, three-dimensional domain in which homogeneous, isotropic turbulence is generated by means of random forcing. This turbulence has well defined properties in terms of its energy dissipation rate and spectral characteristics. During the simulation we keep track of the DSD.

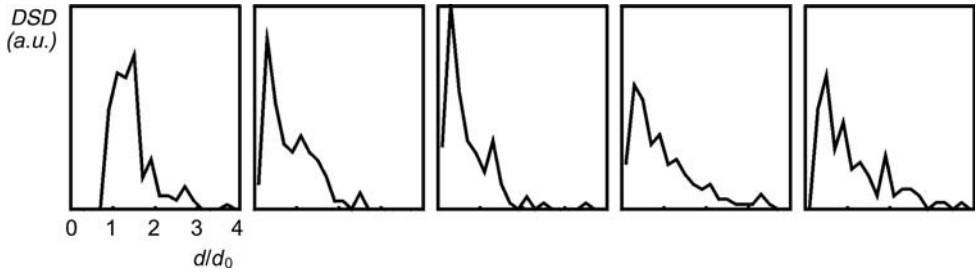

Figure 6. DSD (by number of drops) at (from left to right) $t = 13\ \tau_K$, 26 τ_K, 39 τ_K, 52 τ_K and 65 τ_K.

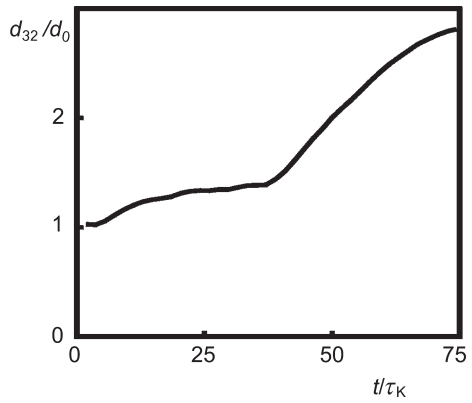

Figure 7. Predicted Sauter mean diameter as a function of time.

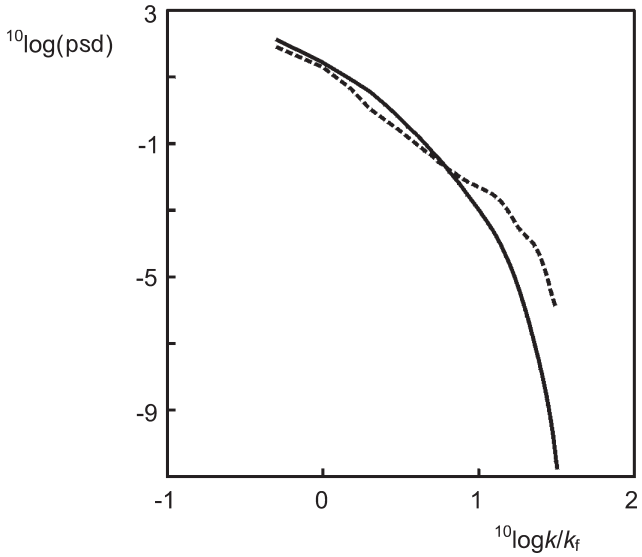

Figure 8. Turbulence spectra of a single phase (drawn line) and liquid–liquid dispersion (dashed line). The power-spectral density has arbitrary (lattice–Boltzmann) units, the wavenumber k has been non-dimensionalized with the main forcing wavenumber k_f.

The turbulence history given to the liquid–liquid dispersion in the direct numerical simulation is derived from the history a fluid package in a stirred tank experiences when it traverses the highly turbulent impeller region. The latter has been estimated from a single-phase large-eddy simulation. Results of the DNS show in great detail the dynamics of the liquid–liquid dispersion. The DSD and the Sauter mean diameter were tracked as a function of time. It was demonstrated that the presence of droplets changed the turbulence spectrum due to the generation of small-scale turbulence in the continuous phase.

Future work will focus on the sensitivity of the results with respect to the spatial resolution. We expect at least some sensitivity since in our numerical method the liquid–liquid interface has a thickness of typically two to three lattice spacings. In the present simulation the interface thickness therefore was comparable to the Kolmogorov length scale. In physical reality, however, the interface is much thinner than any of the other scales relevant in the turbulent two-phase system. We also need to run more simulations in order to determine DSDs and spectra with a better statistical accuracy.

Once these numerical issues are resolved we are in a position to study in great detail the relation between turbulence and the DSD of practical liquid–liquid dispersions. To couple specific industrial processes and process equipment to these fundamental results, we will perform single-phase large-eddy simulations (LES) of the flow in dispersion devices (stirred tanks, static mixers). We subsequently will perform a DNS in the periodic domain containing the dispersion with a turbulence history reflecting the path fluid packages travel in a realistic flow field. Representative turbulence history paths are extracted from the LES. This way the dispersing performance of a real process or apparatus may be evaluated. In the longer run simulations that take into account the role of surfactants and/or stabilizers are foreseen.

NOMENCLATURE

D	impeller diameter, m
N	impeller speed, rev s^{-1}
P	inserted power, W
T	tank diameter, m
d_0, d	(initial) drop diameter, m
d_{32}	Sauter mean diameter, m
k_f, k	(forcing) wavenumber, $1/m$
t	time, s
u'	RMS of velocity fluctuations, m s^{-1}
v_{tip}	impeller tip speed, m s^{-1}
Δ	grid spacing, m
ε	energy dissipation rate, W kg^{-1}
η_K	Kolmogorov lenght scale, m
ρ	density, kg m^{-3}
σ	interfacial tension, N m^{-1}
τ_K	Kolmogorov time scale, s
Re	Reynolds number
We	Weber number

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